Electronic Supplementary Information (ESI)

Dramatic effect of thermal expansion mismatch on the structural, dielectric, ferroelectric and pyroelectric properties in low-cost epitaxial PZT films on $SrTiO_3$ and Si

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Content of the Electronic Supplementary Information (ESI)

- Azimuthal (in-plane) orientation and mosaicity of the PZT layers (Fig. ESI 1)
- Polar (out-of-plane) mosaicity of the PZT layers (Fig. ESI 2)
- PN versus PUND measurement method for ferroelectric hsteresis loops

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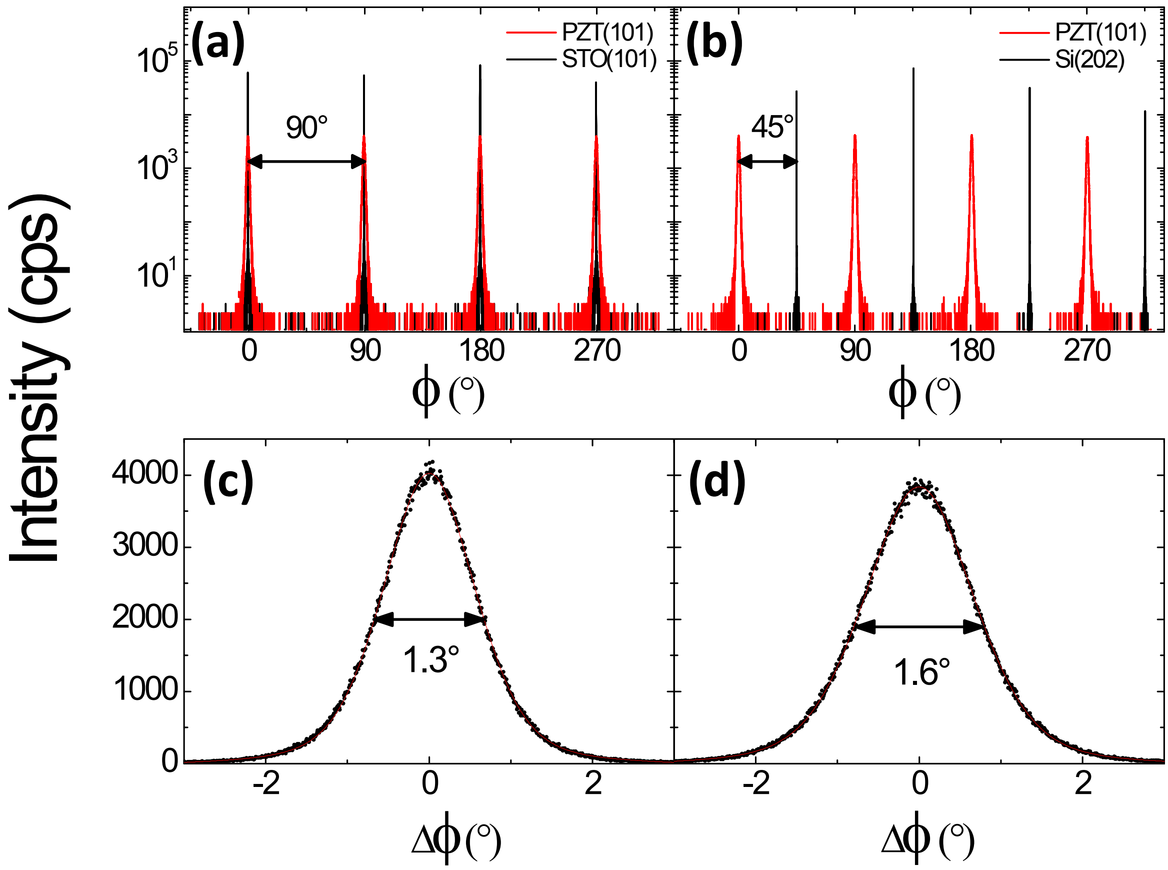

Fig. ESI 1: (a-b) **ϕ-scans** in the Bragg position of the (h0h) asymmetric reflections of the {00l}-oriented PZT films on the different substrates: (a) STO and (b) Si. These large ϕ-scans give the in-plane orientation of the PZT layer and its epitaxial relationship with the substrate. (c-d) ϕ-scans around one asymmetric (101) reflection of the PZT layer, giving its in-plane (azimuthal) mosaicity, (c) on STO and (d) on Si substrate.

The periodicity of asymmetric reflections measured by in-plane azimuthal scans (Fig. ESI 1) give the epitaxial relationship between the PZT layer and the substrate. As reported elsewhere, it is cube on cube on $SrTiO_3$ substrate having the same perovskite structure, and 45° in-plane rotated on Si(001)[1,2]. The complete epitaxial relationships can be written as follows: [100]PZT(001)//[100]SRO(001)//[100]STO(001)//[110]Si(001). The azimuthal mosaicity of the 500 nm thick PZT layer is quite good, about 1.3° and 1.6° on STO and Si substrate, respectively. The corresponding polar mosaicity is rather better, about 0.9° and 1° on STO and Si substrate, respectively (see Fig. ESI 2).

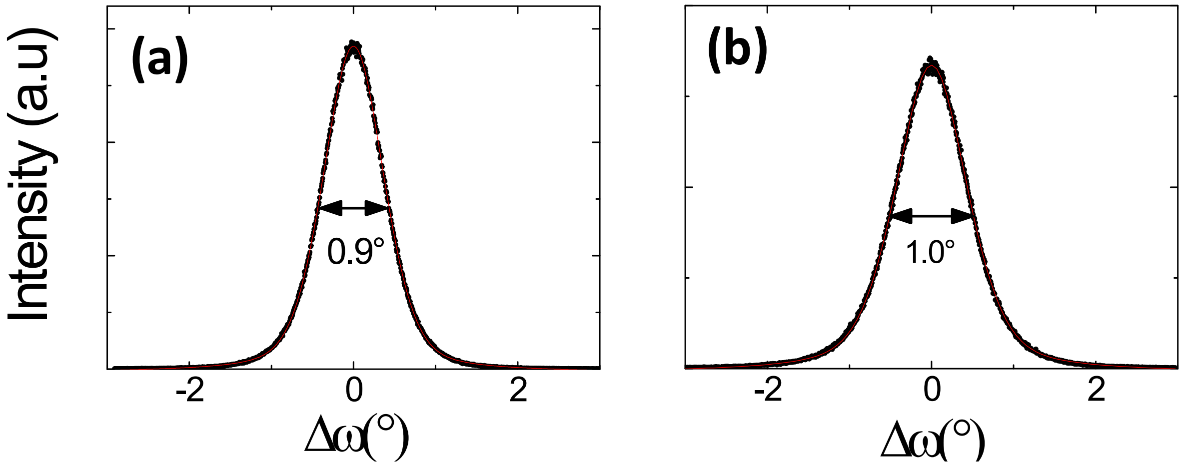

Fig. ESI 2: ω-scans around the {002} symmetric reflections of the PZT layers on (a) STO and (b) Si substrate, giving its polar mosaicity.

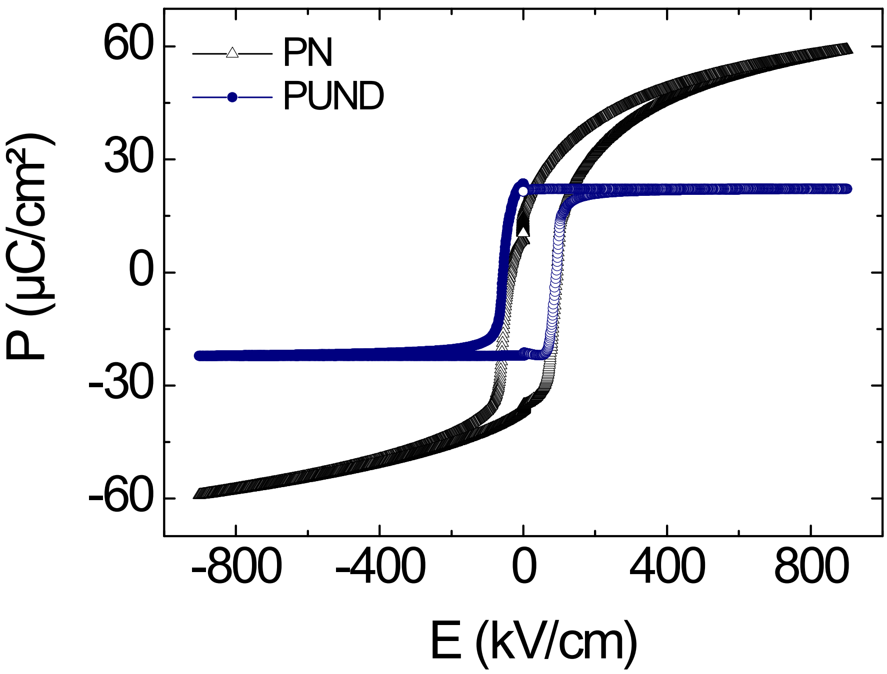

Fig. ESI 3: Ferroelectric hysteresis loops on the PZT/SRO/STO(001) heterostructure at room temperature with i) alternatively positive and negative (PN) polarization tension pulse trains (black triangles) and ii) alternatively two positive and two negative (Positive Up Negative Down, PUND) polarization tension pulse trains (blue triangles).

Fig. ESI 3 shows the ferroelectric hysteresis loop (electric polarization versus electric field) of the PZT/SRO/STO(001) heterostructure taking into account all the polarization contributions (by PN method) or only the remnant polarization (by PUND method). The PUND (Positive Up Negative Down) excitation pulse trains allowing to discriminate the polarization current from the dielectric displacement and leakage current.[3,4] So that, at 800 kV/cm for instance, the PN curve gives a polarization of about 60 $\mu C/cm^2$ whereas the PUND curve gives a polarization of 22.5 $\mu C/cm^2$ that corresponds to the remnant polarization. As specified in the main text, note that all the ferroelectric hysteresis loops are shown with the PUND method in the main communication in order to take only the polarization current contribution.

References

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