Supplementary Information for:

Possible Absence of Critical Thickness and Size Effect in Ultrathin Perovskite Ferroelectric Films

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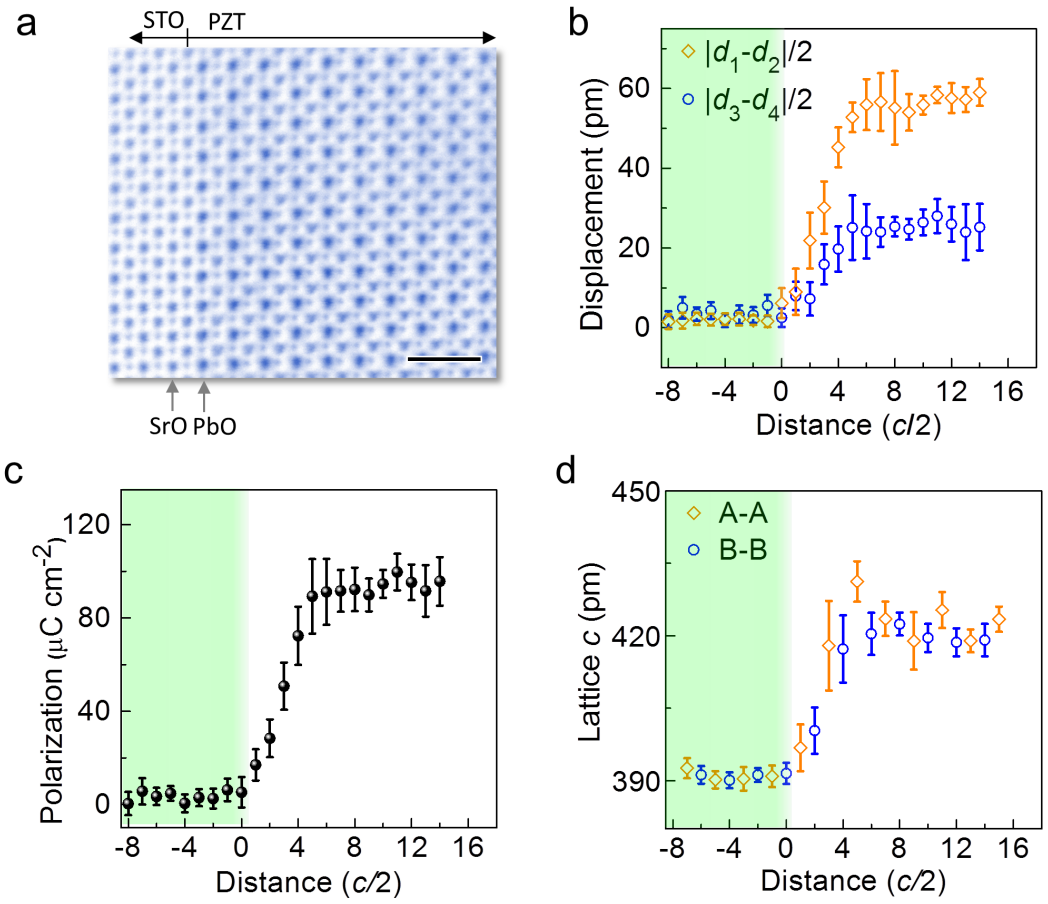
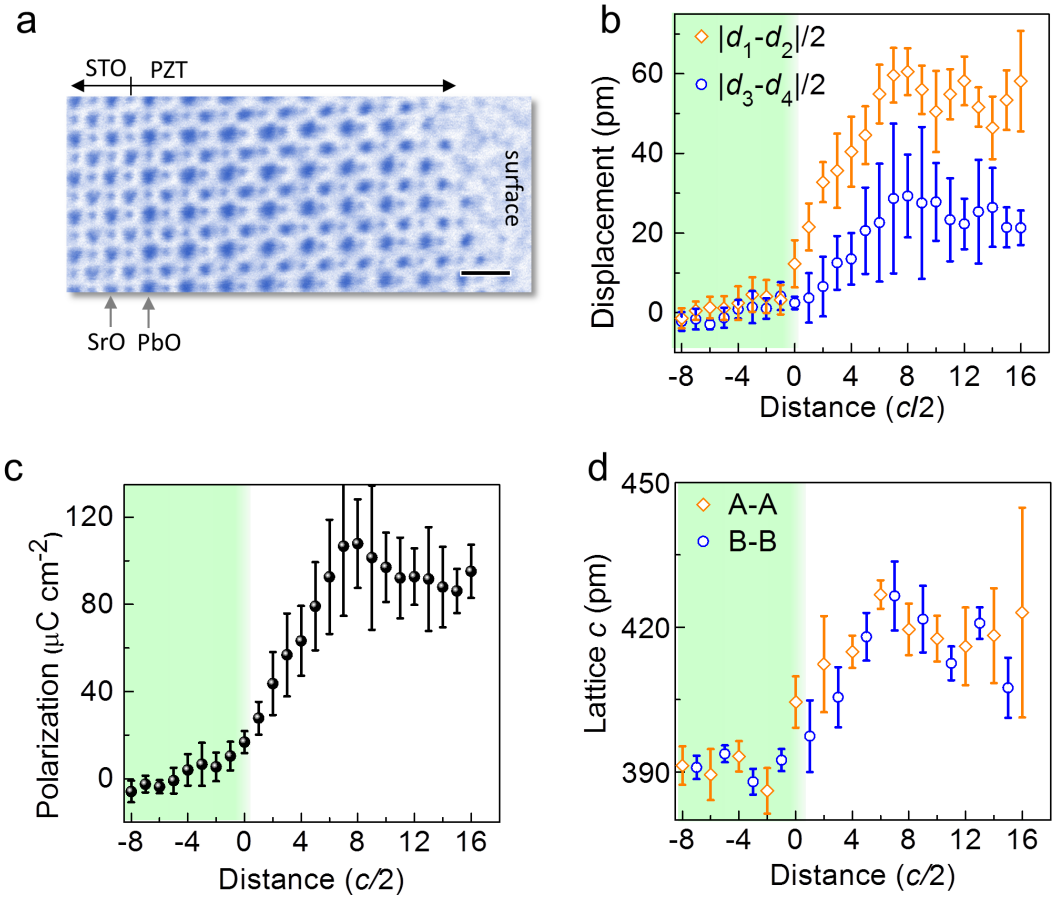
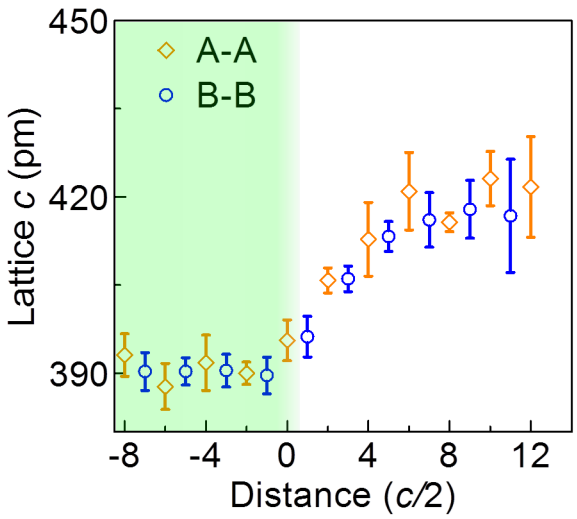


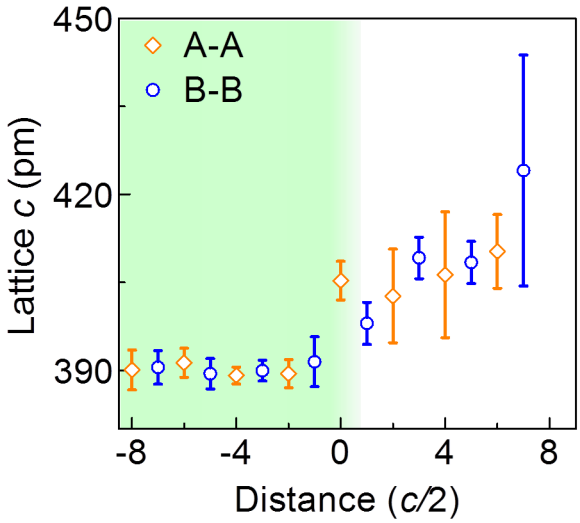
Supplementary Figure 1 | A region with thickness about 18–20 unit cells. (a) The annular bright filed (ABF) image. Scale bar: 1 nm. (b) Calculated displacements of A (Sr or Pb) respective to O and B (Ti or Zr/Ti) respective to O. The error bar is the standard deviation (s.d.). (c) Calculated total polarization. The error bar is the s.d. (d) Calculated lattice *c* from cation sublattice. The error bar is the s.d. These measured structural parameters of PZT thin film are basically in agreement with the previous X-ray study of $PbZr_{0.2}Ti_{0.8}O_3$ powder [1].



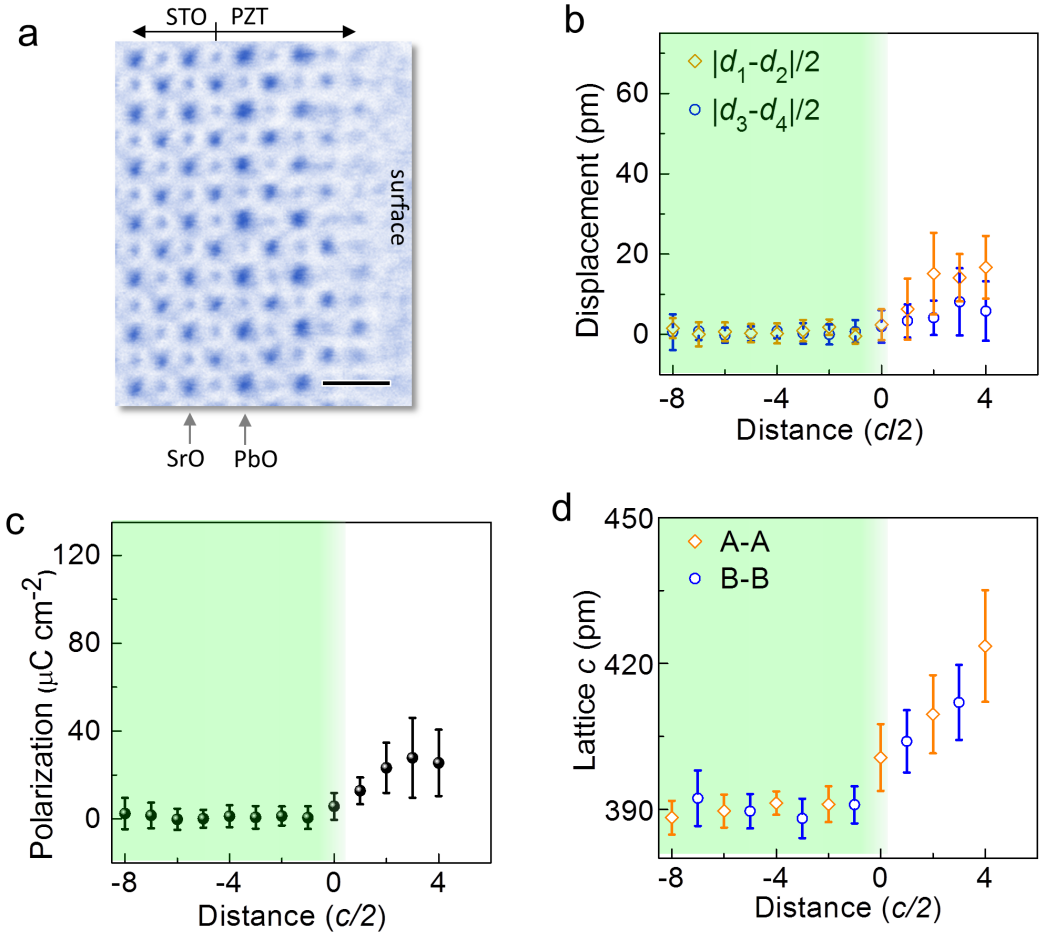
Supplementary Figure 2 | A region with thickness of 8.5 unit cells. (a) The ABF image. Scale bar: 0.5 nm. (b) Calculated displacements of A (Sr or Pb) respective to O and B (Ti or Zr/Ti) respective to O. The error bar is the s.d. (c) Calculated total polarization. The error bar is the s.d. (d) Calculated lattice *c* from cation sublattice. The error bar is the s.d.



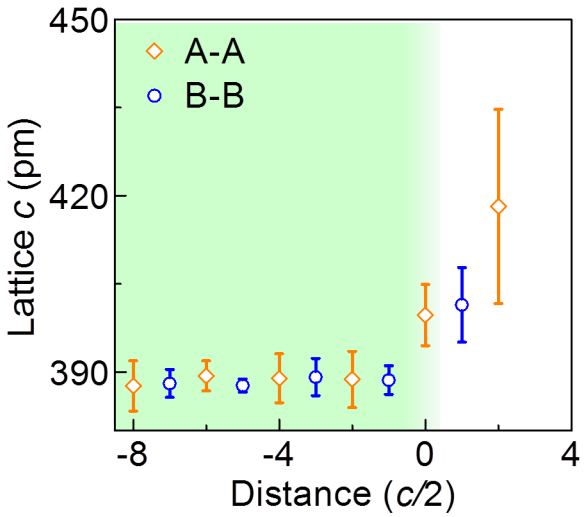
Supplementary Figure 3 | A region with thickness of 6.5 unit cells. Calculated lattice *c* from cation sublattice. The error bar is the s.d.



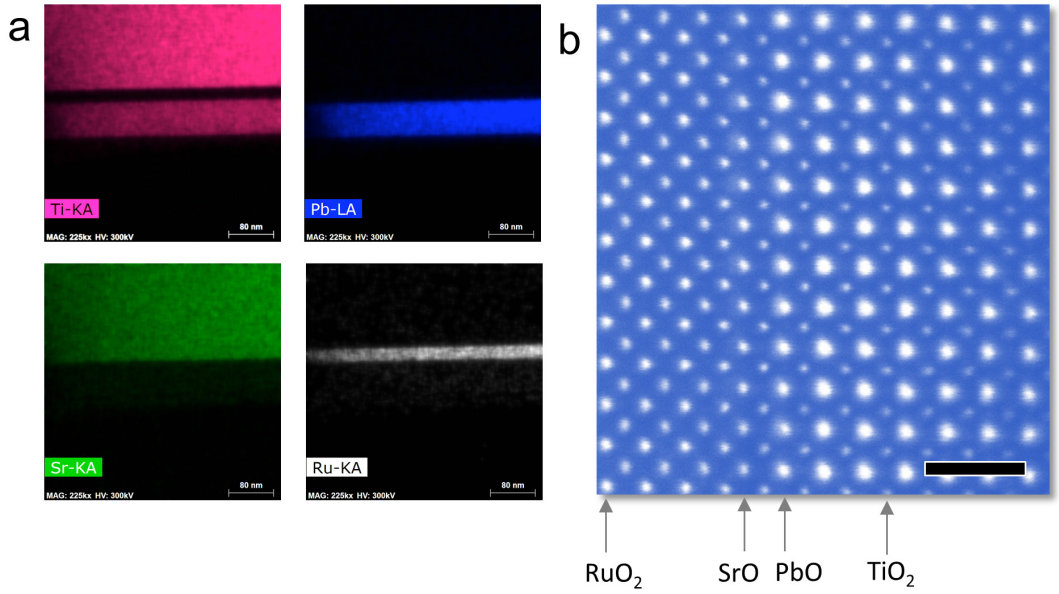
Supplementary Figure 4 | A region with thickness of 4 unit cells. Calculated lattice *c* from cation sublattice. The error bar is the s.d.



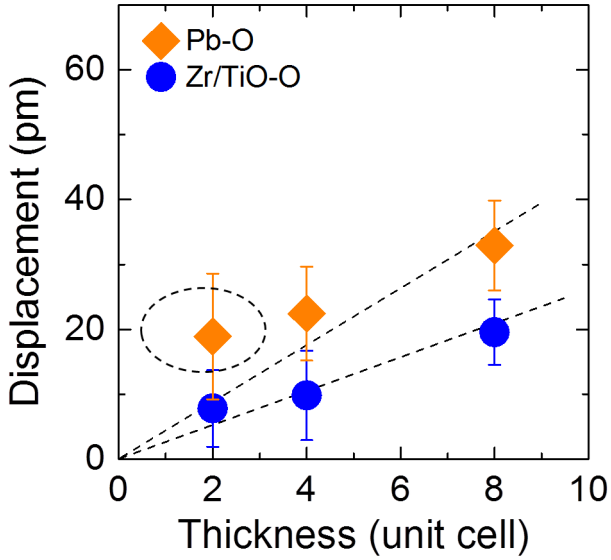
Supplementary Figure 5 | A region with thickness of 2.5 unit cells. (a) The ABF image. Scale bar: 0.5 nm. (b) Calculated displacements of A (Sr or Pb) respective to O and B (Ti or Zr/Ti) respective to O. The error bar is the s.d. (c) Calculated total polarization. The error bar is the s.d. (d) Calculated lattice *c* from cation sublattice. The error bar is the s.d.



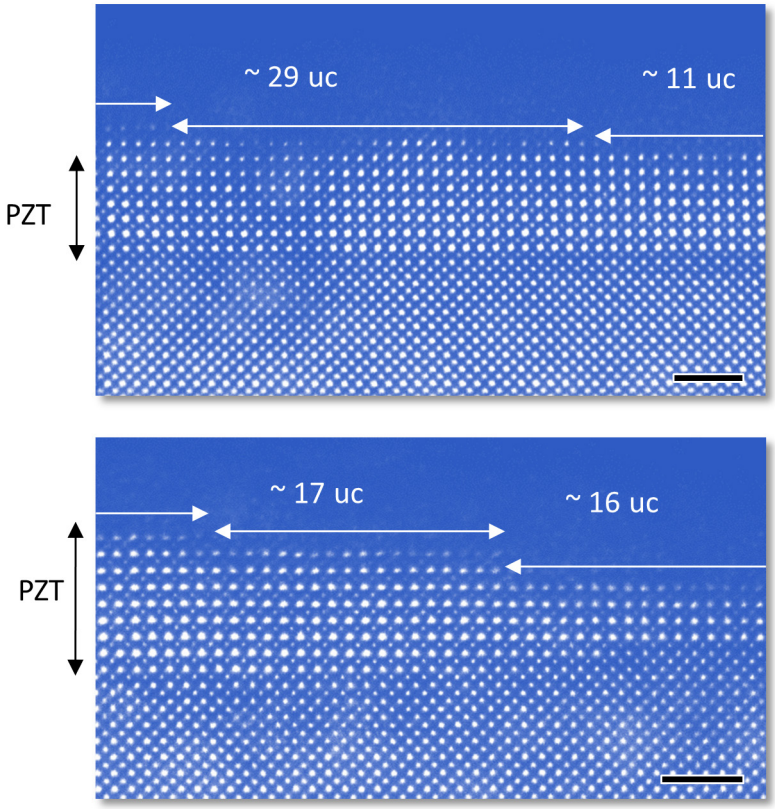
Supplementary Figure 6 | A region with thickness of 1.5 unit cells. Calculated lattice *c* from cation sublattice. The error bar is the s.d.



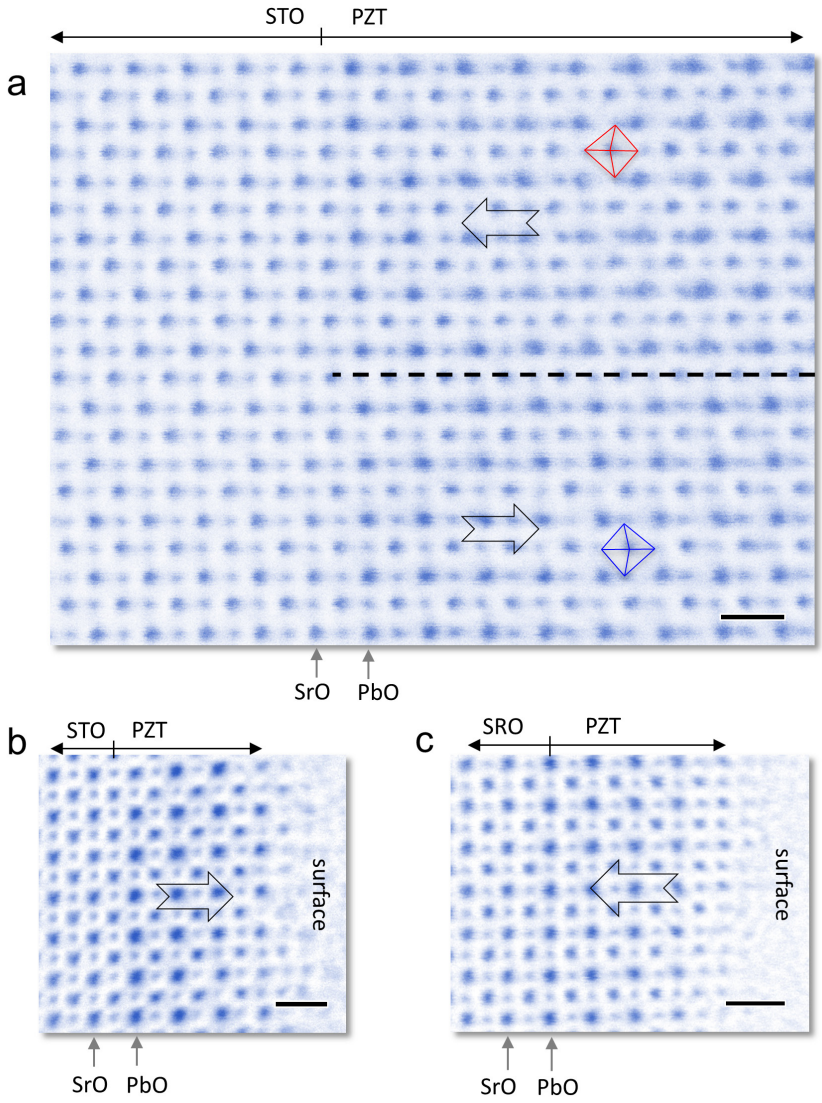
Supplementary Figure 7 | $PbZr_{0.2}Ti_{0.8}O_3$ thin film on $SrTiO_3$ substrate with $SrRuO_3$ bottom electrode. (a) Elemental mapping. (b) High angle annular dark field (HAADF) image. Scale bar: 1 nm.



Supplementary Figure 8 | Robust Pb-O bond in the ultrathin $PbZr_{0.2}Ti_{0.8}O_3$ film on $SrRuO_3/SrTiO_3$. Displacements of Pb respective to O and Zr/Ti respective to O are plotted as a function of the thickness. The error bars are the s.d.



Supplementary Figure 9 | The atomic terrace in the thin regions in $PbZr_{0.2}Ti_{0.8}O_3$ films. The typical width of a terrace is from 10 to 30 unit cells. Scale bar: 2 nm.



Supplementary Figure 10 | The switchable nature of polarization. (a) In a relative thick region, both upward and downward domains are present in $PbZr_{0.2}Ti_{0.8}O_3$ films on $SrTiO_3$ substrate, indicating the switchable nature. The domain wall position is highlighted by the dashed line. Scale bar: 0.5 nm. (b) Upward polarization in $PbZr_{0.2}Ti_{0.8}O_3/SrTiO_3$ heterostructure. Scale bar: 0.5 nm. (c) Downward polarization in $PbZr_{0.2}Ti_{0.8}O_3/SrRuO_3/SrTiO_3$ heterostructure. Scale bar: 0.5 nm.

Supplementary Reference

1 Joseph, J. *et al.* Structural investigations on $Pb(Zr_xT_{1-x})O_3$ solid solutions using the X-ray Rietveld method. *J. Mater. Sci.* **35**, 1571-1575 (2000).