

Electronic Supplementary Information

Ferroelectric $\text{Hf}_{0.5}\text{Zr}_{0.5}\text{O}_2$ films on $\text{SrTiO}_3(111)$

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1. Projected cation structures of o-HZO crystallites and LSMO

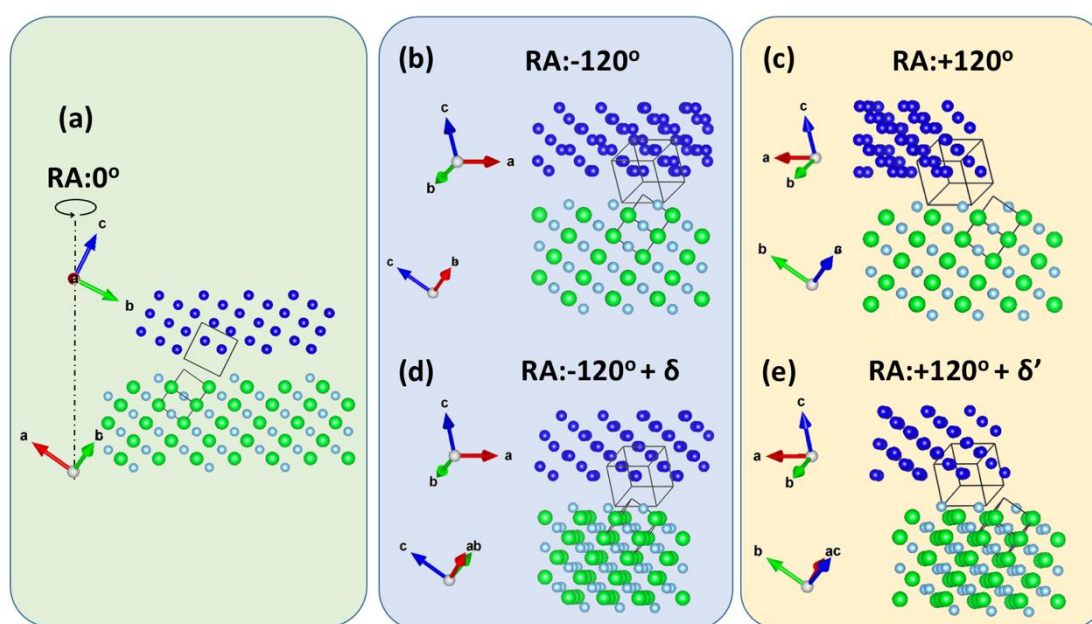


Fig. S1. (a), (b)-(d), and (c)-(e): Projected cation structures of one o-HZO crystallite (Hf/Zr atoms are represented by blue balls) and the LSMO electrode (La/Sr and Mn atoms are represented by green/light blue balls, respectively) along the three in-plane equivalent directions of LSMO(111)/STO(111). VESTA software was used to simulate the structures.¹ (a) View along [0-11] of LSMO. (b-c) View of both cation structures at +120° and -120° in-plane rotation around the rotation axis (RA) with respect to (a), where LSMO is viewed along one zone axis of the [0-11] type, but HZO is not properly oriented. (d-e) show a slightly rotated view of (b-c) (+120° and -120° and an additional small rotation denoted as δ and δ') with HZO properly oriented along the nearest zone axis in (b-c).

2. Cations distance and angle in the three families of orthorhombic crystallites

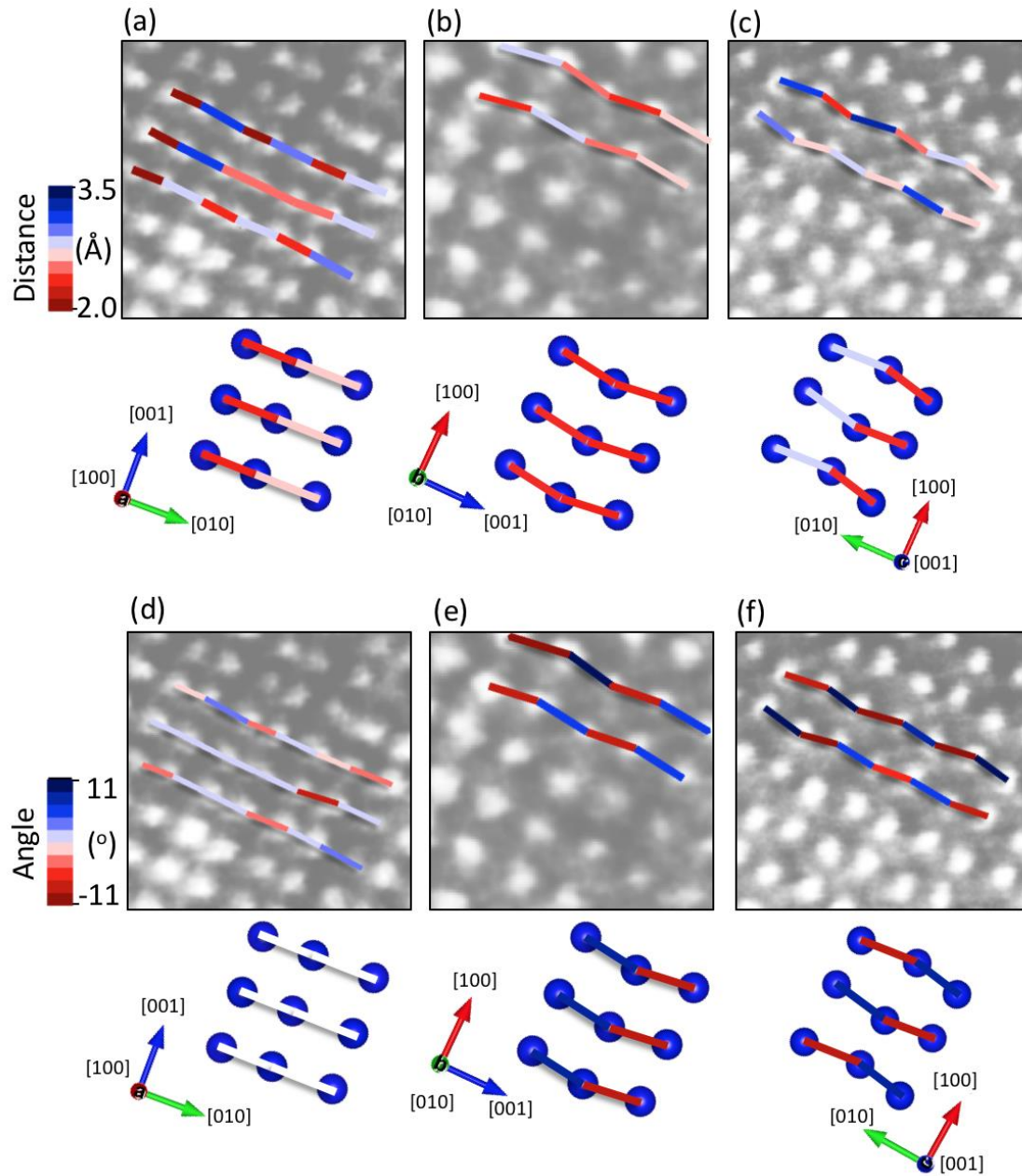


Fig. S2. (a-f) Cations distances (a-c) and angles (d-f) of the $Pca2_1$ phase on planes of the three families of orthorhombic crystallites. In the colored sticks onto the faded HAADF images, the color sticks represent the distance or angle between connected cations in the respective scales. Top panels correspond to the experimental measurement and bottom ones to the projected model of cations, with Hf/Zr atoms shown in blue. (a)-(d), (b)-(e) and (c)-(f) images correspond to the zoomed regions of Fig. 3 (b), (c) and (d), respectively. As expected from the model, colored sticks in (a) show the alternatively larger and smaller distance between adjacent cations along $[010]$, while in (b) more similar distance values are obtained along $[001]$, compatible with the constant spacing predicted by the model. Similarly, colored sticks in (c) show the alternatively larger and smaller distances and angles between adjacent cations along $[010]$. (d) shows a

dispersion of angle values around zero, but not periodically variation. On the other hand, (e) and (f) show periodically bigger and smaller angles, as expected from the respective models.

3. Semicoherent interface along the [01-1]LSMO direction

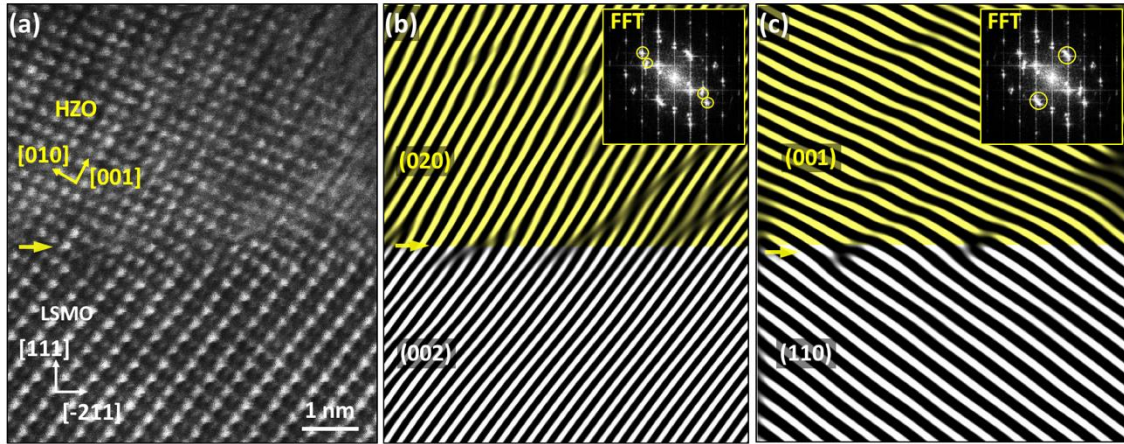


Fig. S3. (a) HAADF cross-sectional image of an HZO crystallite and the LSMO electrode. (b) Corresponding Fast-Fourier-Transform (FFT) filtered image. (020) HZO planes and (002) LSMO planes are selected in the FFT (see top-right side inset) and used to obtain the image containing only the selected planes. For clarity, planes in the HZO are shown in yellow while those in LSMO are shown in white. (c) FFT filtered image using the (001) HZO and (110) LSMO planes.

4. Polarization loop measured by PUND

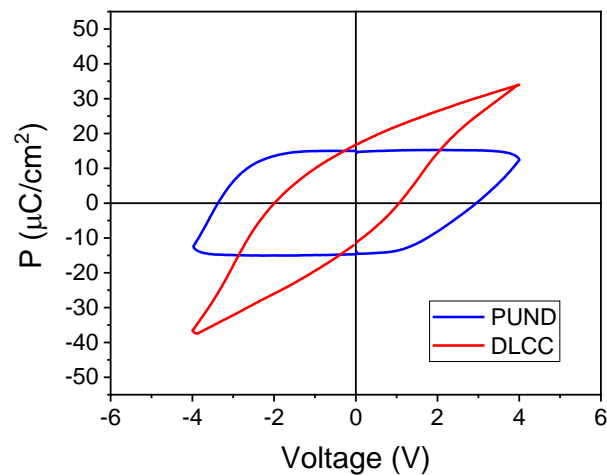


Fig. S4. Comparison between polarization loops measured using PUND (using 1 s delay time between pulses) and DLCC methods. The remanent polarization in both loops is above $14 \mu\text{C}/\text{cm}^2$. It can be observed that the loop collected using PUND shows larger coercive field than that collected using DLCC. This is because the presence of the so-called fluid imprint field.²

References

- 1 K. Momma and F. Izumi, *J. Appl. Crystallogr.*, 2011, **44**, 1272–1276.
- 2 P. Buragohain, A. Erickson, P. Kariuki, T. Mittmann, C. Richter, P. D. Lomenzo, H. Lu, T. Schenk, T. Mikolajick, U. Schroeder and A. Gruverman, *ACS Appl. Mater. Interfaces*, 2019, **11**, 35115-35121.