Supporting Information

Energy Storage Performance and Electric Breakdown Field of Thin Relaxor Ferroelectric PLZT Films using Microstructure and Growth Orientation Control

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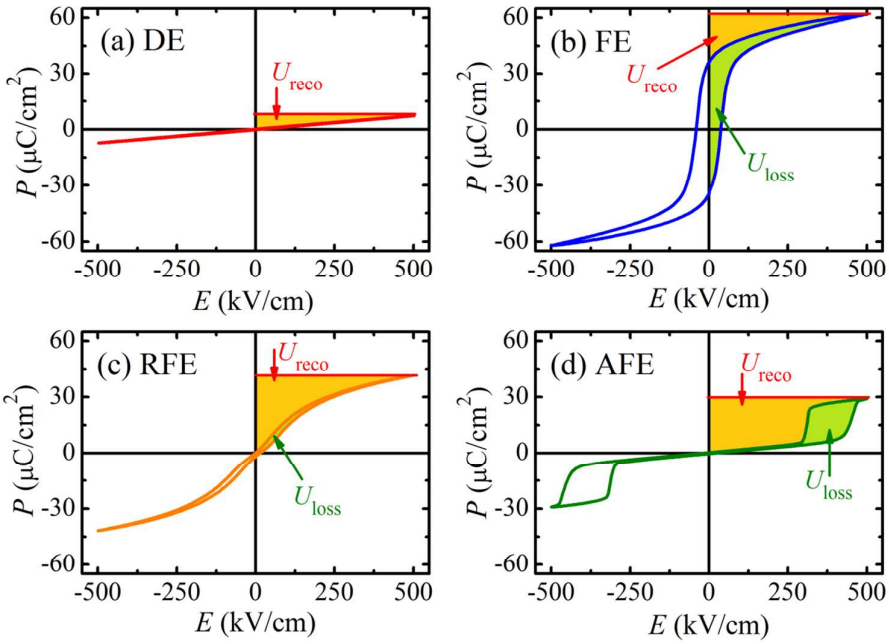

Figure S1. Schematic illustration of polarization (*P*-*E*) hysteresis loops, recoverable energy-storage density (U_{reco}) and energy-loss density (U_{loss}) for (a) linear dielectrics (*DE*), (b) ferroelectrics (*FE*), (c) relaxor ferroelectrics (*RFE*) and (d) antiferroelectrics (*AFE*).

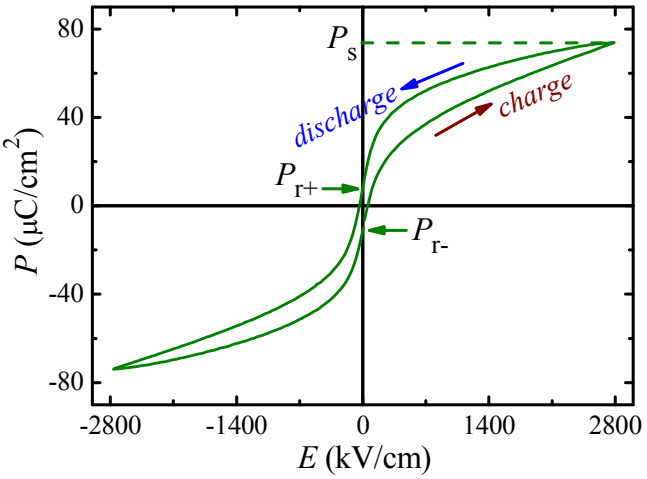

Figure S2. Schematic illustration of a polarization (*P*-*E*) hysteresis loop. P_s, P_{r+} and P_{r-} are the saturation polarization at maximum applied field E_{max}, positive remanent polarization and negative remanent polarization, respectively.

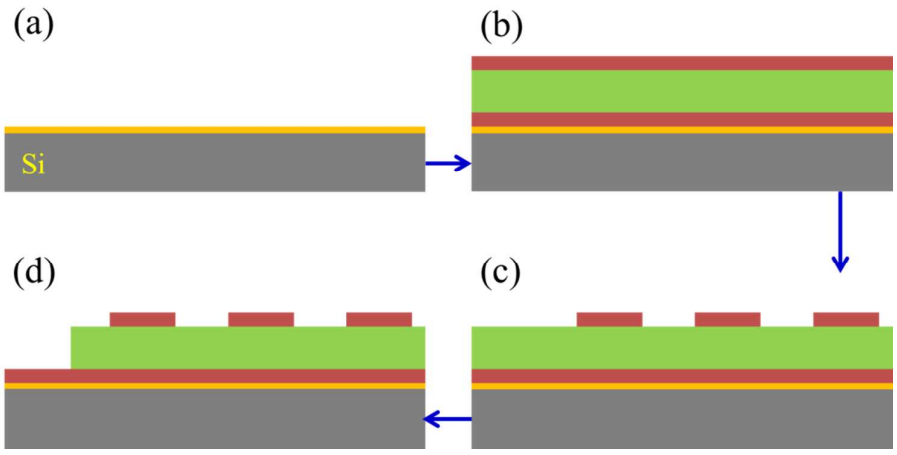

Figure S3. Schematic of the fabrication of thin films and capacitors: (a) fabrication of nanosheet ($Ca_2Nb_3O_{10}$ 'CNOns' or $Ti_{0.87}O_2$ 'TiOns') on Si using Langmuir-Blodgett technique, (b) deposition of ferroelectric stack $SrRuO_3/Pb_{0.9}La_{0.1}(Zr_{0.52}Ti_{0.48})O_3/SrRuO_3$ (SRO/PLZT/SRO) on nanosheet/Si using pulsed laser deposition, (c) argon-ion beam etching of SRO top electrode and (d) wet-chemical etching of PLZT film.

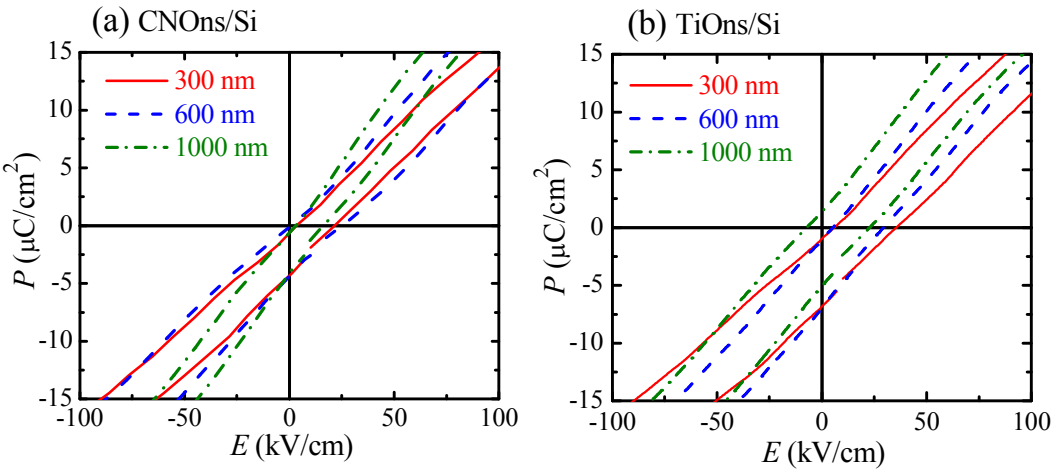

Figure S4. Zoon-in view, in the range of -100 to +100 kV cm^{-1}, of the *P-E* loops measured at an applied electric field of 1000 kV cm^{-1} of PLZT films with various thicknesses on (a) CNOns/Si and (b) TiOns/Si.

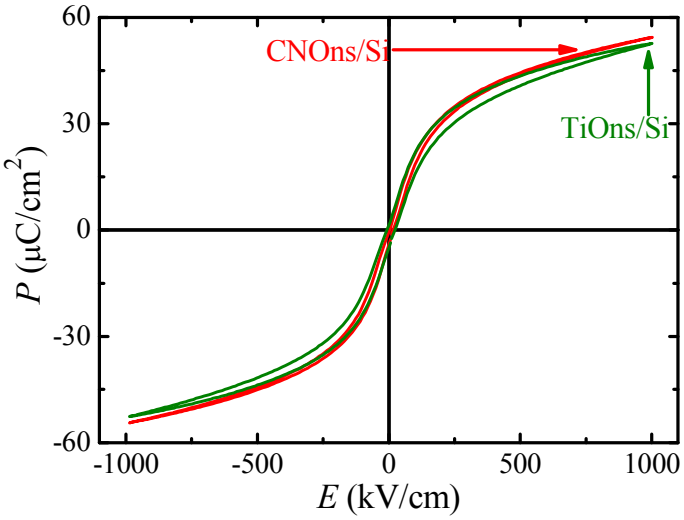

Figure S5. Comparison of the *P-E* loops of 1000-nm-thick PLZT films on CNOns/Si and TiOns/Si, measured at 1000 kV cm^{-1}.

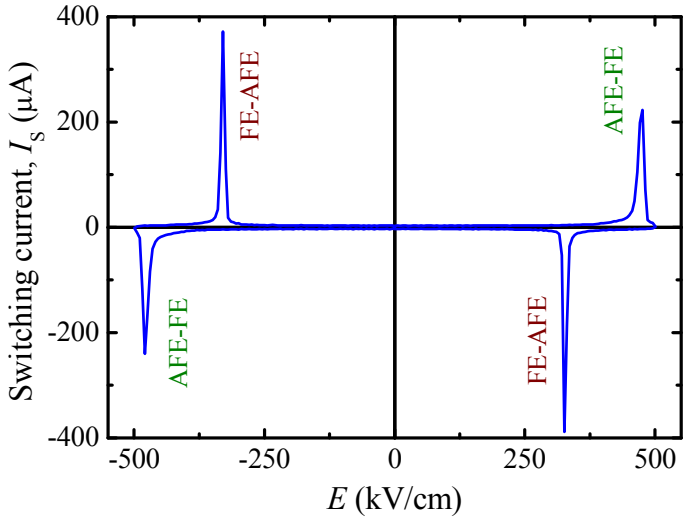

Figure S6. Switching current–electric field of antiferroelectric $PbZrO_3$ thin film (corresponding *P-E* loop is shown in Figure S1d) grown on SRO/CNOns/Si substrate.

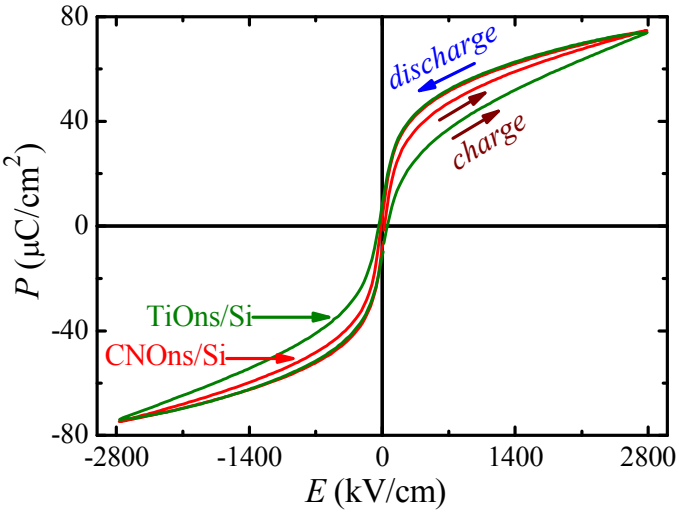

Figure S7. Comparison of the *P-E* loops of 1000-nm-thick PLZT films on CNOns/Si and TiOns/Si, measured at 2800 kV cm^{-1}.

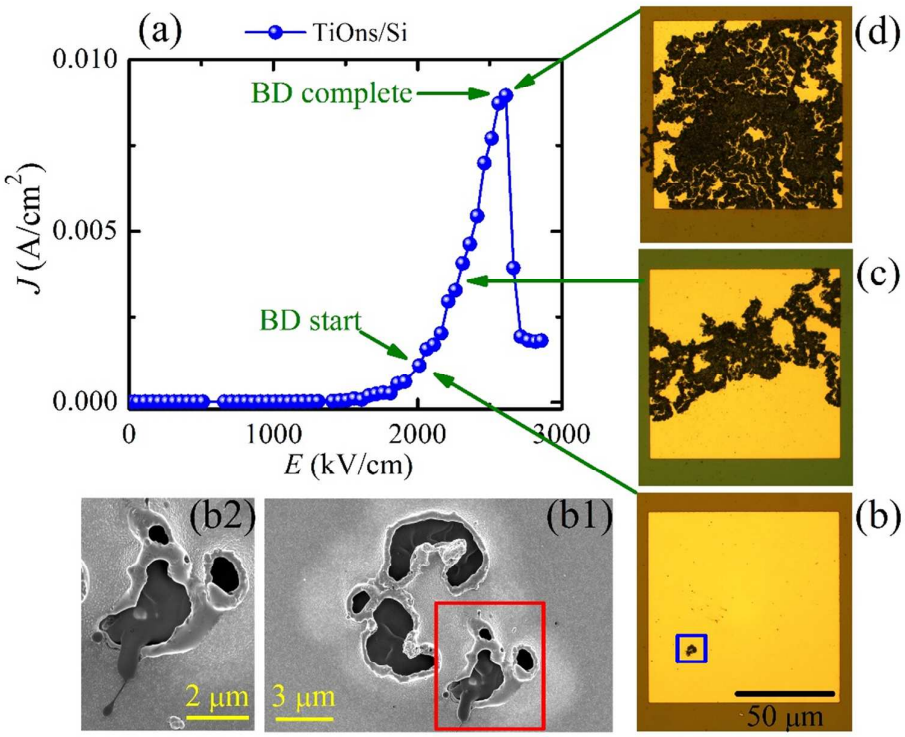

Figure S8. (a) Leakage current density–electric field (*J-E*) of 1000-nm-thick PLZT film grown on SRO buffered TiOns/Si. Microscope images of (b) breakdown spot or staring point of breakdown process at about 2085 kV cm^{-1}, (c) with increasing applied electric field and (d) when the capacitor or top electrode is fully broken. SEM images of (b1) zoom-in the blue-box line in (b) and (b2) zoom-in the red-box line in (b1).