Supporting Information

Negative-Pressure Enhanced Ferroelectricity and Piezoelectricity in *Lead-free* BaTiO₃ Ferroelectric Nanocomposite Films

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Figure S1. (a) Left panel: cross-sectional high-resolution STEM image along the interface between B (BTO) and S (Sm_2O_3). The BTO (dark contrast phase due to lower Z number) and the Sm_2O_3 (bright contrast phase due to higher Z number) are phase-separated. The inset shows the fast Fourier transformed image. Right panel: the corresponding Fourier-filtered image without periodic misfit dislocations (the dashed red lines guide the interfaces). (b) The selected area diffraction (SAD) image. (c)

Plain-view STEM image. A distinct Sm_2O_3 (bright contrast phase due to higher Z number, highlighted by red dash circles) nanopillar in BTO (dark contrast phase due to lower Z number) matrix structure of the thin film with a clear phase separation. The inset is a schematic diagram of VAN structure. (d) The corresponding fast Fourier transformed (FFT) filtered image of Figure c showing without periodic misfit dislocations along the interface between BTO and Sm_2O_3 highlighted by red dash circles. The inset is a schematic diagram of VAN structure.



Figure S2. Selected area diffraction (SAD) pattern of $BaTiO_3$ in $BaTiO_3:Sm_2O_3$ nanocomposite films. The separated diffraction dots might result from the distortion of the $BaTiO_3$ lattices, which proves the potential monoclinic structure of $BaTiO_3$.



Figure S3. X-ray reciprocal space map (RSM) of (420) Bragg reflections of Nb:SrTiO₃ and BaTiO₃ at Phi=218.3° for plain BaTiO₃ 100 nm film.



Figure S4. Zoom-in view of temperature dependence of lattice parameter d_{222} in 500nm-thick BaTiO₃:Sm₂O₃ nanocomposite films for showing two distinct regimes separated by a kink in the lattice parameter clearly.



Figure S5. (a) Ferroelectric hysteresis loop and (b) Piezoelectric coefficient and phase hysteresis loops for plain BaTiO₃ 100 nm film measured at 25 °C.



Figure S6. Room-temperature butterfly loops obtained before and after the Simple Harmonic Oscillator (SHO) fitting.