

Supporting Information Data Submitted to Nanoscale

To further confirm the substrate-controlled BFO phase and orientation of CFO pillars and to investigate the microstructure of those heterostructures, the TEM result was performed as seen in Fig. S1. The cross-sectional TEM images clearly show the two separated constituent phases on both LAO (Fig. S1 a) and STO (Fig. S1 b) substrate, where the CFO pillars were embedded in the T-BFO and R-BFO matrix respectively. The interface of octahedral CFO pillars tilted to the LAO substrate surface in compare with on the STO substrate thus the CFO pillars emerges as triangular and rectangular shaped islands, respectively, which is consistent with the AFM images. Confirmed by The high-resolution TEM and the corresponding Fast-Fourier transform (FFT) patterns in the insets Fig S1 c and d, the two phases exhibited good epitaxial relationships: $[100]_{\text{T-BFO}}//[111]_{\text{CFO}}$ on LAO substrate and $[100]_{\text{R-BFO}}//[100]_{\text{CFO}}$ on STO substrate, respectively agree with the XRD result.

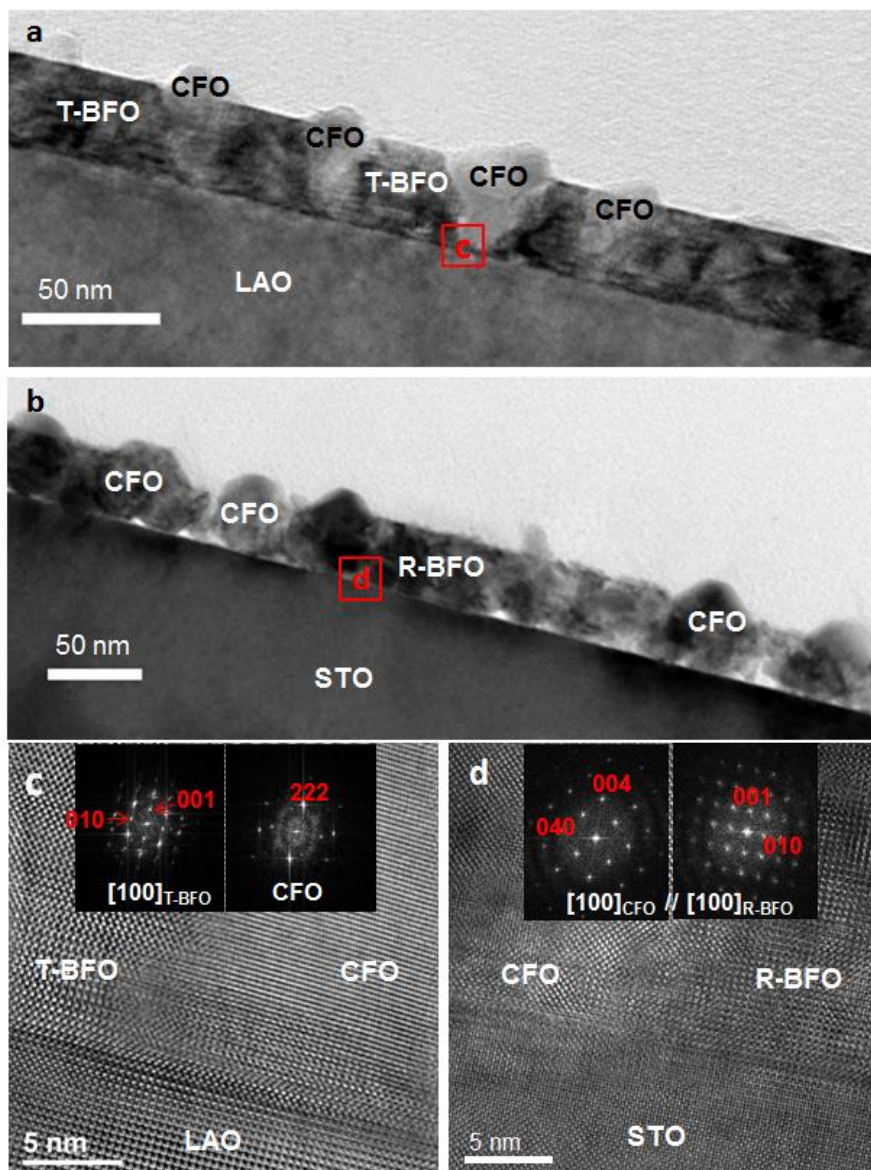


Figure S1: Cross-sectional TEM images taken along $[100]_{\text{LAO}}$ and $[100]_{\text{STO}}$ zone axes showing CFO pillars embedded in (a) T_BFO matrix and in (b) R_BFO matrix. The high-resolution images are taken from the marked areas of (c) T_BFO-CFO/LAO and (d) R_BFO-CFO/STO with the corresponding FFT patterns in the insets.