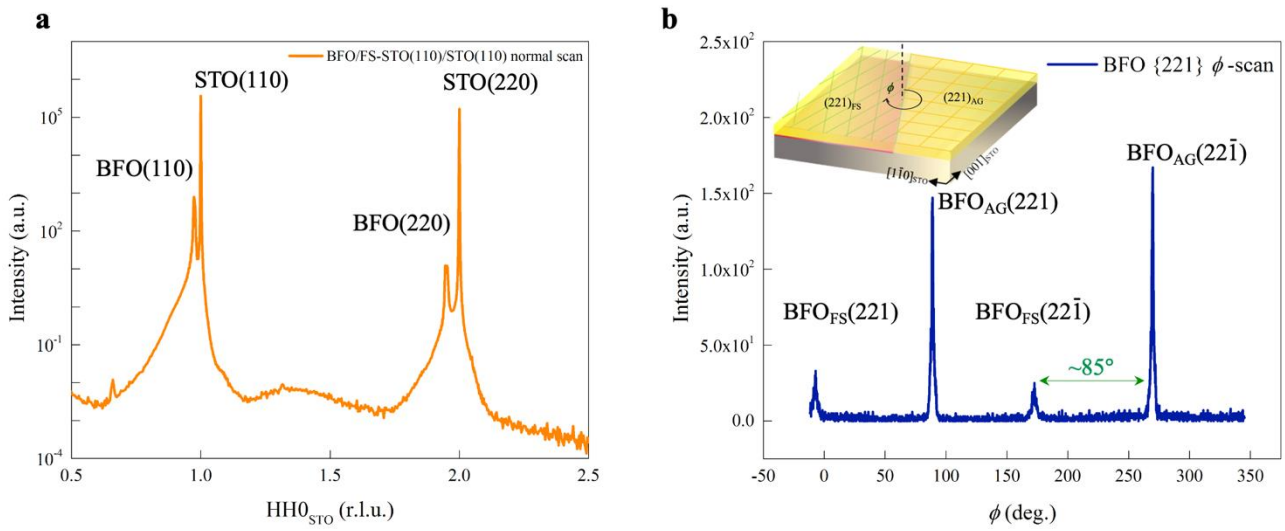
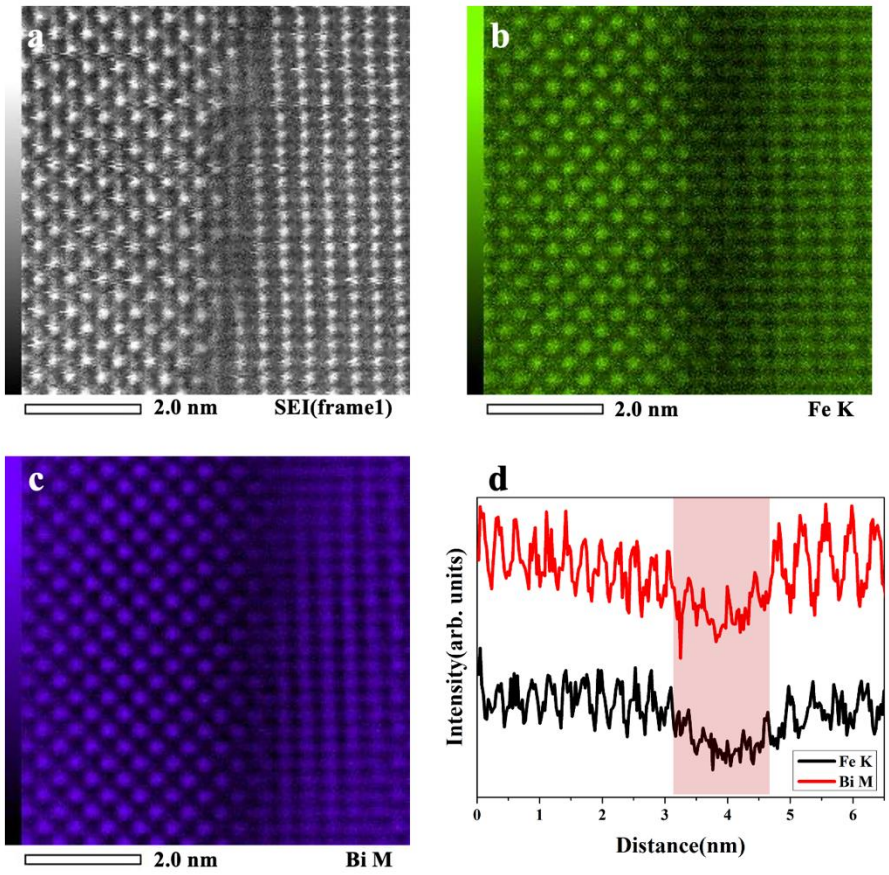
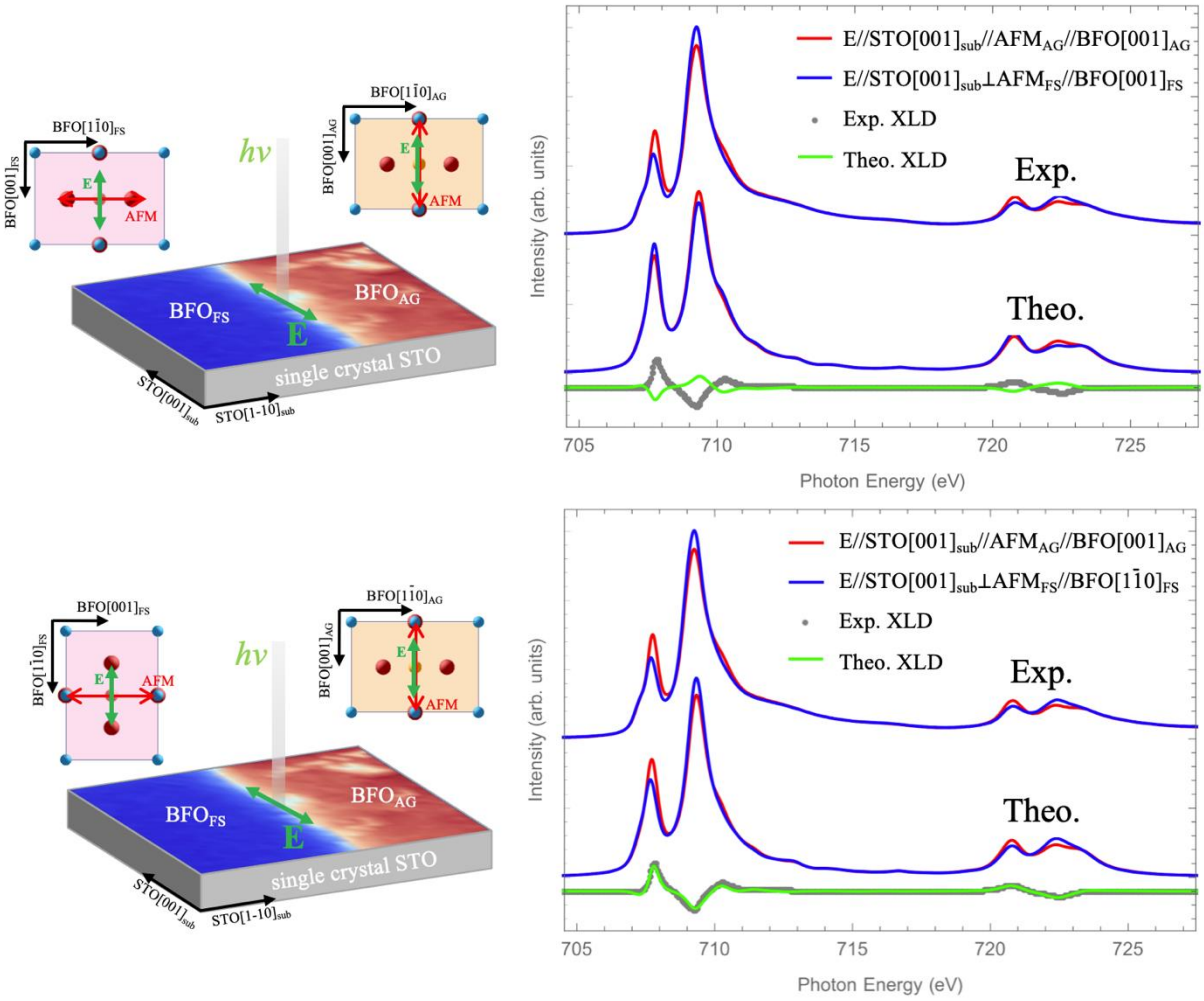


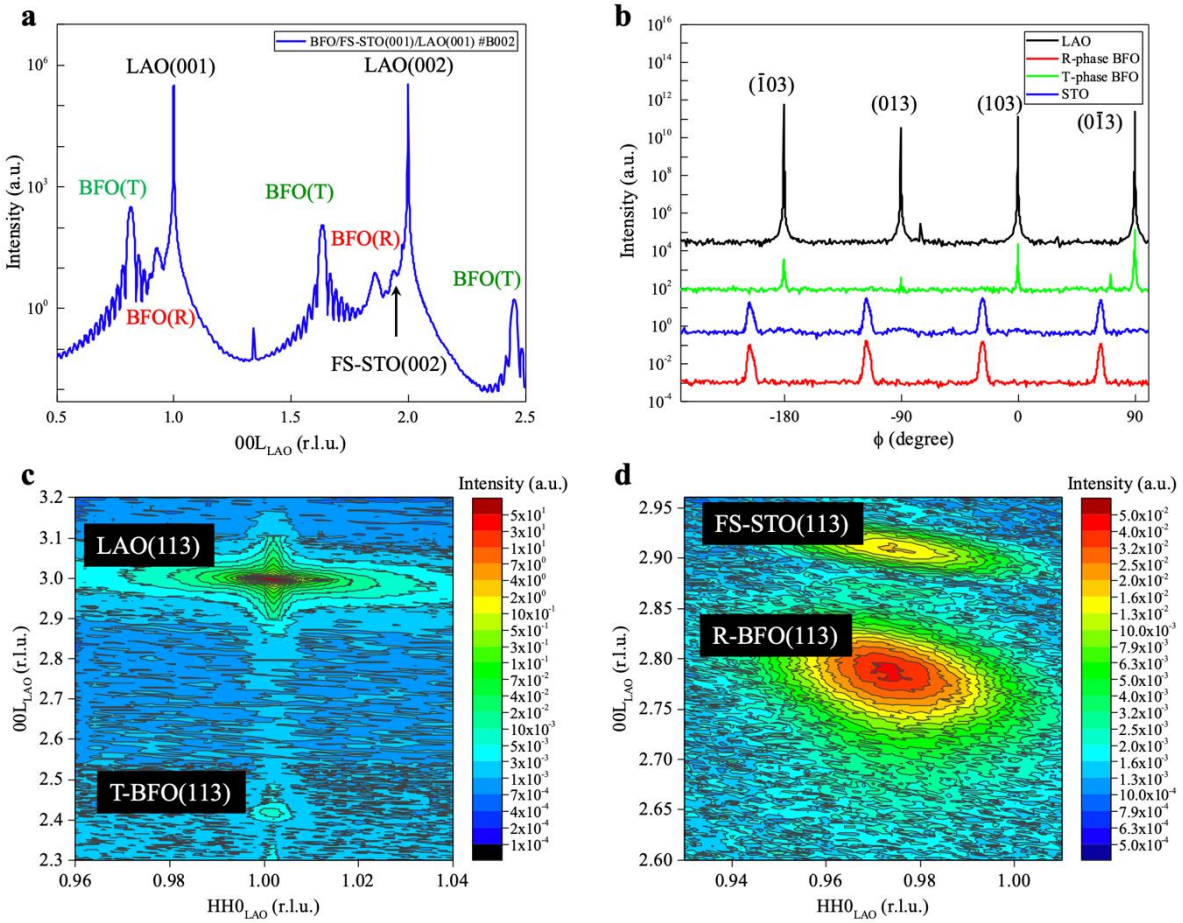
Extended Data Fig. 1 | X-ray Diffraction (XRD) of the Heteroepitaxy. a XRD normal scan of twist BFO homostructure on (110)-oriented STO substrate indicates pure (110)-oriented BFO peaks without any secondary phase. **b** XRD in-plane φ-scan of STO {221} and BFO {221}. The detection of 4 peaks in STO {221} planes indicates the contributions from both freestanding and single crystal STO substrate areas. In the present sample, the twist angle between single crystal STO substrate and freestanding STO is ~85°.



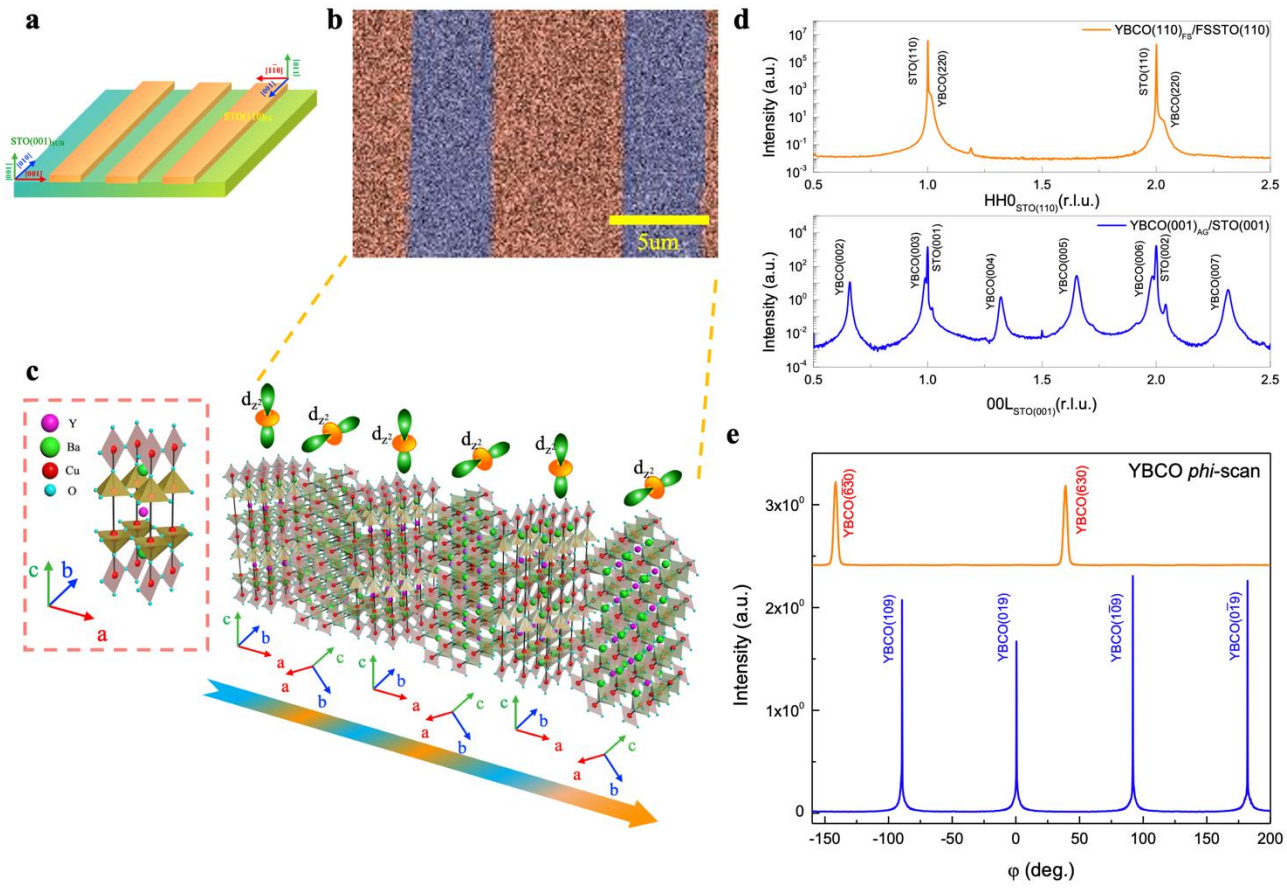
Extended Data Fig. 2 | Atomic resolved EDS map. a HAADF-STEM image of the interface region in the $BFO_{AG}/BFO_{FS.}$ **b** and **c** Atomic resolved Fe and Bi EDS map. **d** Line profiles of Fe and Bi extracted from the mapping.



Extended Data Fig. 3 | Fe $L_{2,3}$-edge X-ray absorption and linear dichroism spectra of BFO_{AG} and BFO_{FS} with E//STO[001]$_{SUB}$. The red and blue lines in the upper right and lower right panels represent the absorption spectra of BFO_{AG} and BFO_{FS}, respectively; while the green lines and gray dots show the experimental and simulated dichroism spectra, respectively. For the calculated spectra in the upper panel, only the 90° rotation of antiferromagnetic axis from BFO_{AG} to BFO_{FS} was considered, as illustrated in the left schematic. On the contrary, for the calculated spectra in the lower panel, both the 90° rotation of antiferromagnetic axis and the rotation of [1$\bar{1}$0] crystallography-axis from BFO_{AG} to BFO_{FS} were considered, as illustrated in the left schematic.



Extended Data Fig. 4 | X-ray diffraction of BFO grown on STO/LAO twist template. **a** XRD normal scan along LAO (001). The characteristic peaks of R-BFO, T-BFO, FS-STO, LAO are labeled accordingly in the figure. **b** The XRD in-plane φ-scans along {103} planes of R-BFO, T-BFO, STO, and LAO indicate the well-aligned epitaxial relationship of R-BFO/FS-STO and T-BFO/LAO, respectively. **c** The RSM around {113} plane of LAO revealed the single domain feature of T-BFO and the T-BFO is fully strained by LAO substrate **d** The RSM around {113} plane of STO also indicates a strained R-BFO phase on FS-STO.



Extended Data Fig. 5 | Patterned twisted YBCO lateral homostructures. a The designer periodic arrays of twisted template composed by $(110)_{FS}$- $(001)_{SUB}$- oriented STO. Firstly, a STO twisted template was prepared with (001)-oriented STO substrate overcovered by freestanding (110)-oriented STO layer. The stripe patterns with periodic arrays were then manufactured through the combination of etching and photolithography. **b** Scanning electron microscopy image of patterned YBCO homostructure grown on the designer twisted STO template. **c** Schematic illustration of the crystal arrangement and representative dz^2 orbital configuration of the twisted YBCO lateral homostructures. **d** XRD normal scans of YBCO films grown on (110) FSSTO (orange line) and on (001) STO substrate (blue line) area. The homostructure exhibits crystal structure of (001)- and (110)-oriented YBCO on $(001)_{SUB}$- and $(110)_{FS}$-oriented STO, respectively, which is nicely consistent with the designed crystal geometry. **e** XRD in-plane phi-scan of {630} plane of (110)-oriented YBCO and {109} plane of (001)-oriented YBCO, revealing the corresponding two-fold and four-fold symmetry of YBCO grown on the designer twisted STO template.