

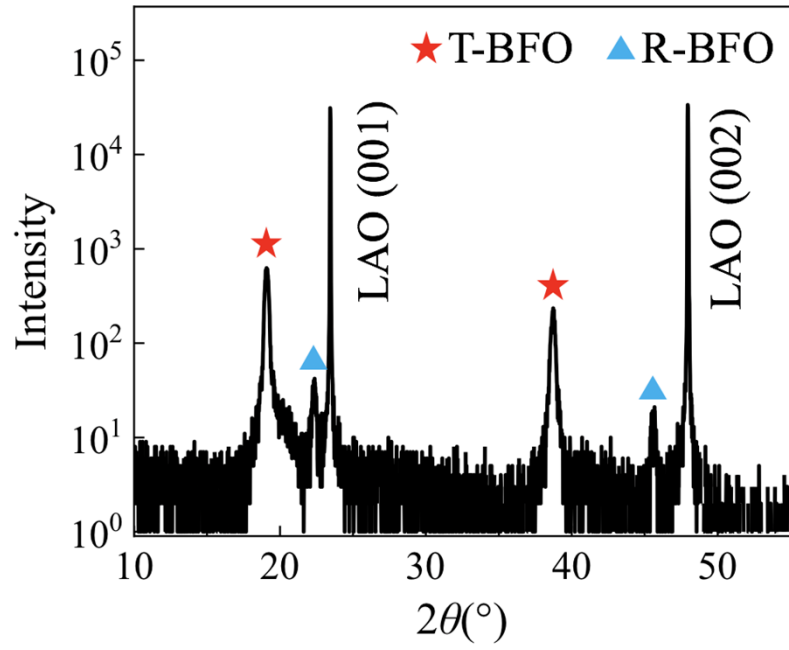
## Supporting Information

### Self-assembly growth strategy for highly ordered ferroelectric nanoisland array

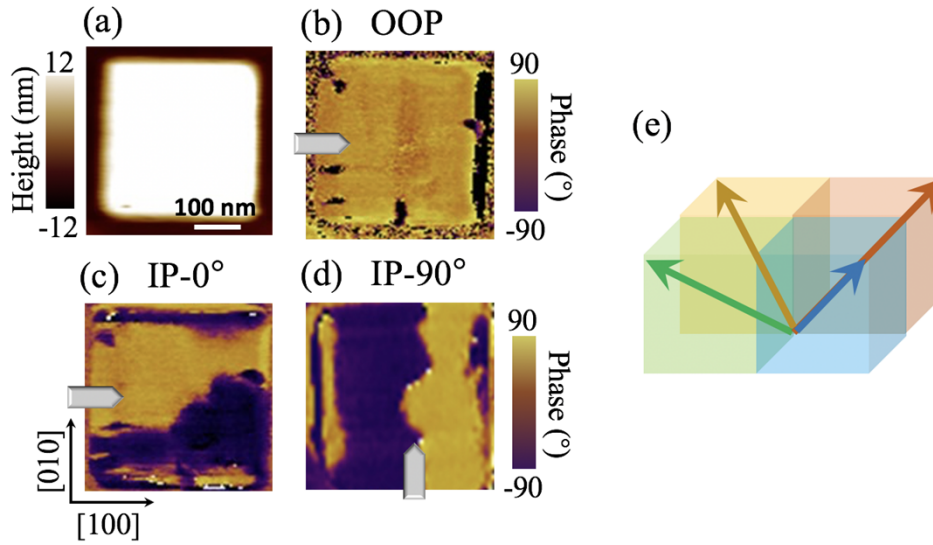
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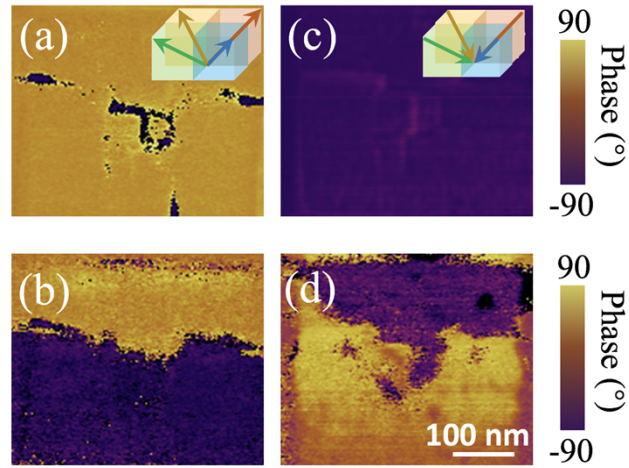
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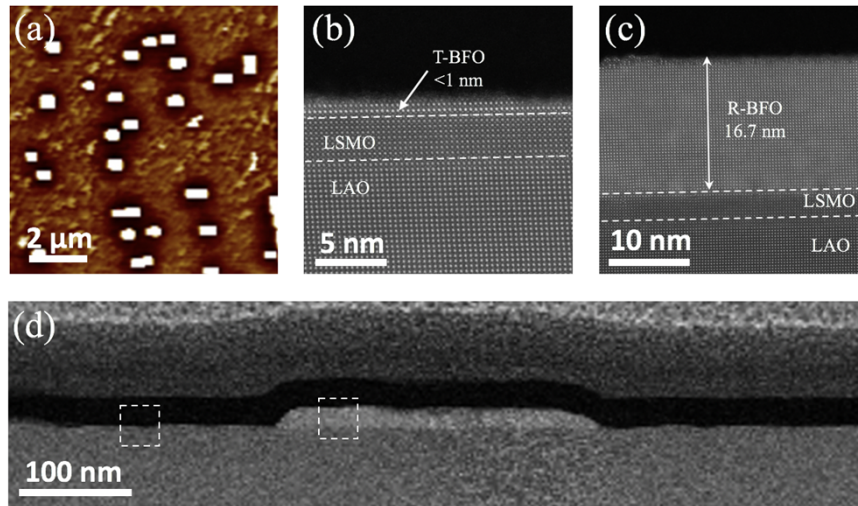
**Fig. S1.**  $\theta$ - $2\theta$  diffraction pattern of BFO/LSMO film grown on patterned LAO substrate.



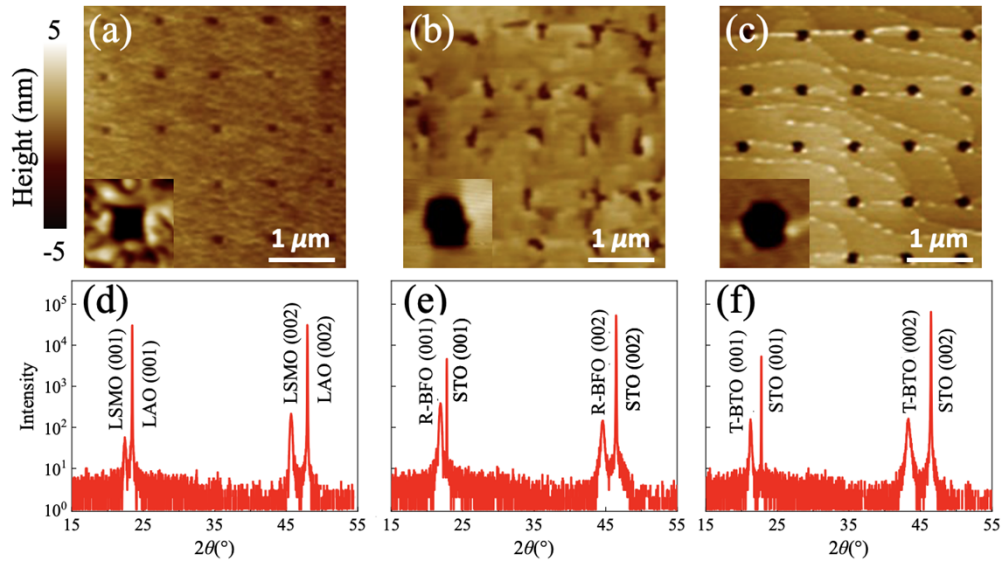
**Fig. S2.** Ferroelectric domain structures of a single BFO nanoisland grown on the nanocavity. (a) AFM image. (b) OOP phase image. (c) IP-0° phase image. (d) IP-90° phase image. The gray arrows in PFM images represent the cantilever of the scanning probe. IP-0° and IP-90° phase images were measured with the cantilever parallel and perpendicular to [100] direction of LAO substrate, respectively. (e) 3D schematic of ferroelectric domain structure reconstructed by PFM images, which is a center-divergent quad-domain configuration. The arrows represent the polarization vectors.



**Fig. S3.** Switching characterization of BFO nanoislands on nanocavities. Pristine (a) OOP and (b) IP-0° phase images of a BFO nanoisland. (c) OOP and d) IP-0° phase images of the same BFO nanoisland after switched by +7.3 V bias applied on AFM tip (sample was grounded). Insets are 3D schematics of ferroelectric domain structures before and after switching, with arrows representing the polarization vectors.



**Fig. S4.** Structure of BFO nanoislands grown on LSMO/LAO heterostructure at the early growth stage (300 pulses). (a) AFM image of BFO nanoislands. STEM images of (b) the T-phase matrix and (c) the R-phase nanoisland, corresponding to the two white dashed boxes labeled in (d) the low-magnification STEM image.



**Fig. S5.** AFM images and  $\theta$ - $2\theta$  diffraction pattern of perovskite oxides grown on patterned substrates. (a, d) LSMO/LAO (001), (b, e) BFO/LSMO/STO (001) and (c, f) BTO/STO (001). Insets are close-up images of a single nanostructure ( $200 \times 200$  nm).