Supplementary Material

Uniform arrays of center-type topological domains in epitaxial ferroelectric thin films

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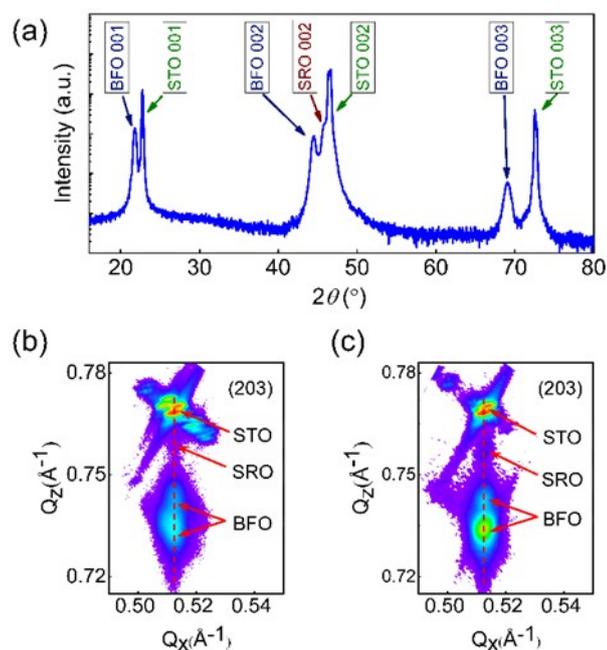
Fig. S1 Typical (a) θ-2θ X-ray diffraction spectrum for a 'mosaic-like' $BiFeO_3$ film. Reciprocal space mapping around the (203) reflection of (b) the 'mosaic-like' and (c) 'stripe-like' $BiFeO_3$ film.
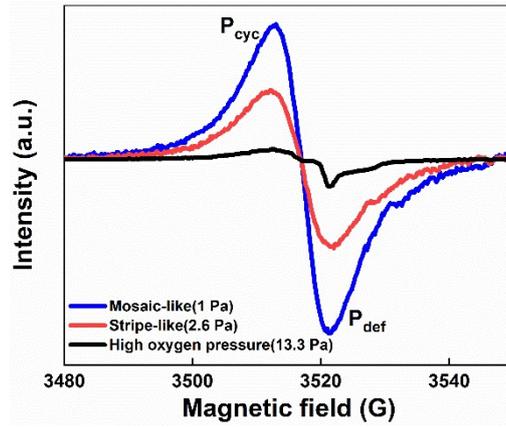
Fig. S2 EPR spectra of 'mosaic-like' (1.0 Pa), 'stripe-like' (2.6 Pa) and high oxygen pressure (13.3 Pa) BFO films.
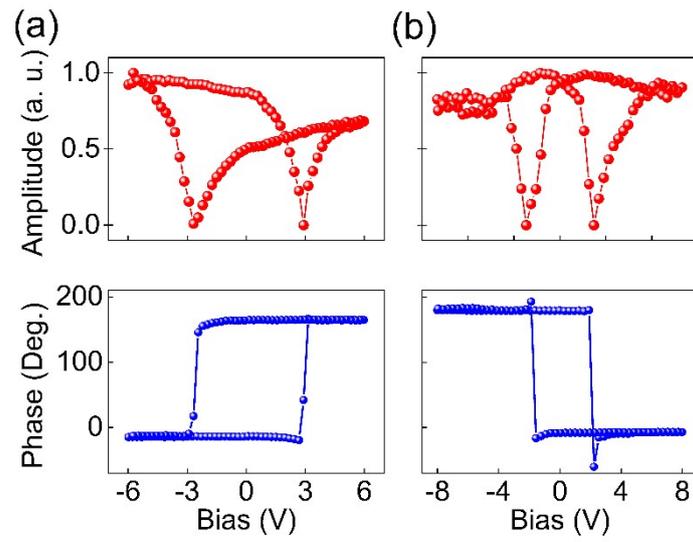
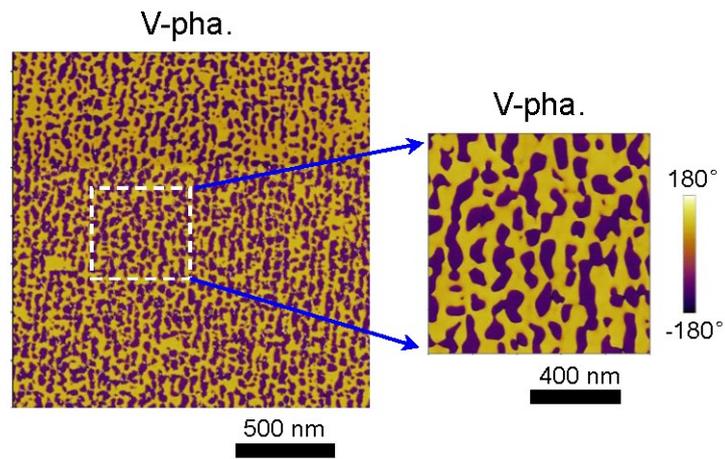**Fig. S3** PFM hysteresis for (a) the 'mosaic-like' and (b) 'stripe-like' BFO film.

Fig. S4 VPFM phase images of stripe domains spontaneously exist in the 'stripe-like' (2.6 Pa) film.
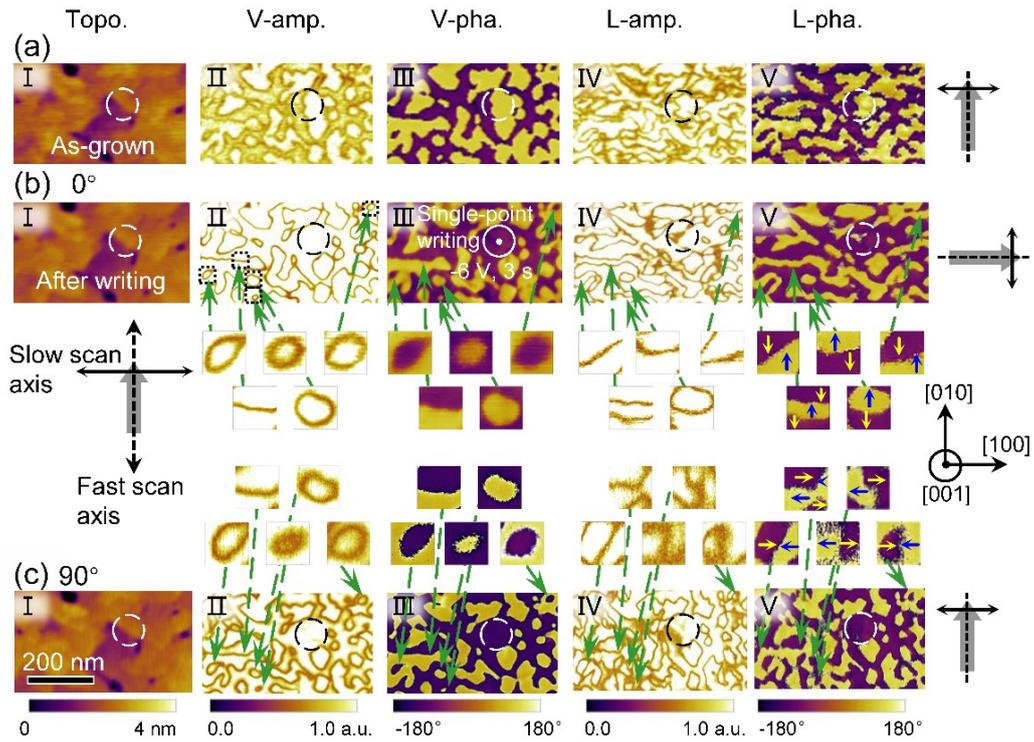
Fig. S5 Vectorial PFM images of center-domains spontaneously exist in 'mosaic-like' film. (a) The topography, VPFM amplitude and phase images for as-grown state of the film. (b-c) The topography, VPFM amplitude and phase images, LPFM amplitude and phase images for two different sample rotation angles (b) 0°and (c)90°. The inset zoom-in local area PFM images in the gap between (b), (c) illustrate five frequently observed domain structures in the film, and three of them are shown in Fig. 2 in the main text.
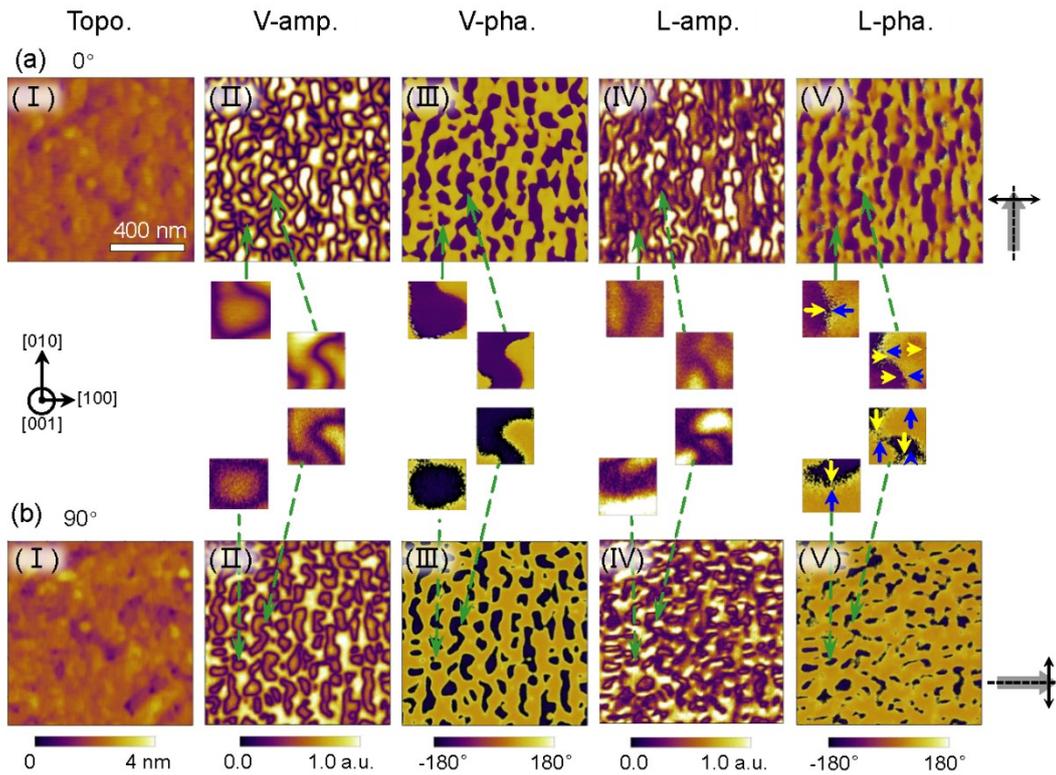
Fig. S6 Vectorial PFM images of center-domains spontaneously exist in 'stripe-like' film. The topography, VPFM amplitude and phase images, LPFM amplitude and phase images for two different sample rotation angles (a) 0°and (b) 90°. The inset zoom-in local area PFM images in the gap between (a), (b) illustrate two frequently observed domain structures in the film.
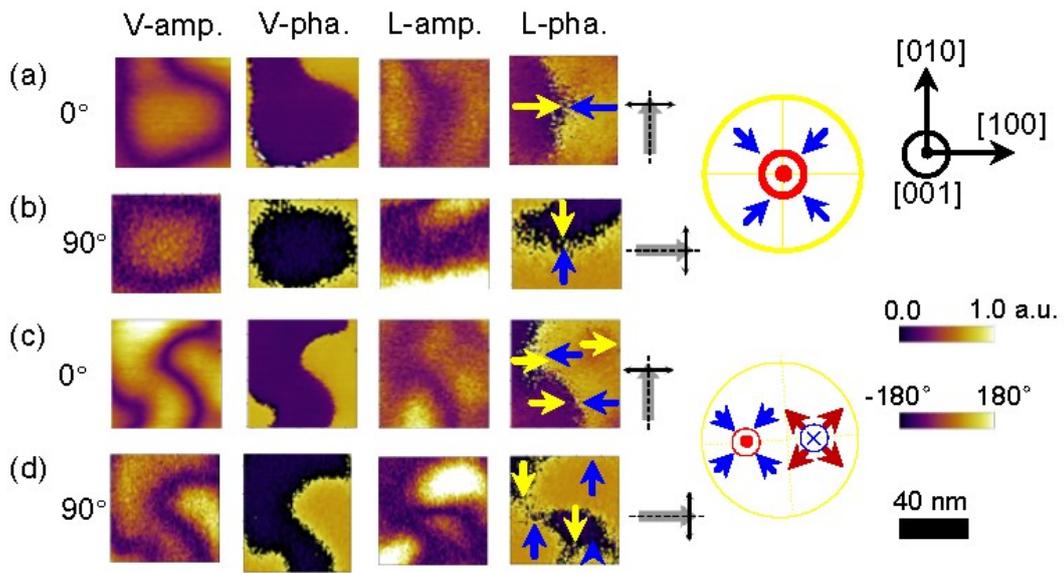
Fig. S7 For "stripe-like" film, (a) 0° and (b) 90° vectorial PFM images of the center-convergent type domain, with upward out-of-plane polarization; (c) 0° and (d) 90° vector PFM images of the double-center type domain, with upward and downward out-of-plane polarization. The cantilever orientation indicates its fast scan axis and slow scan axis during the LPFM measurements.
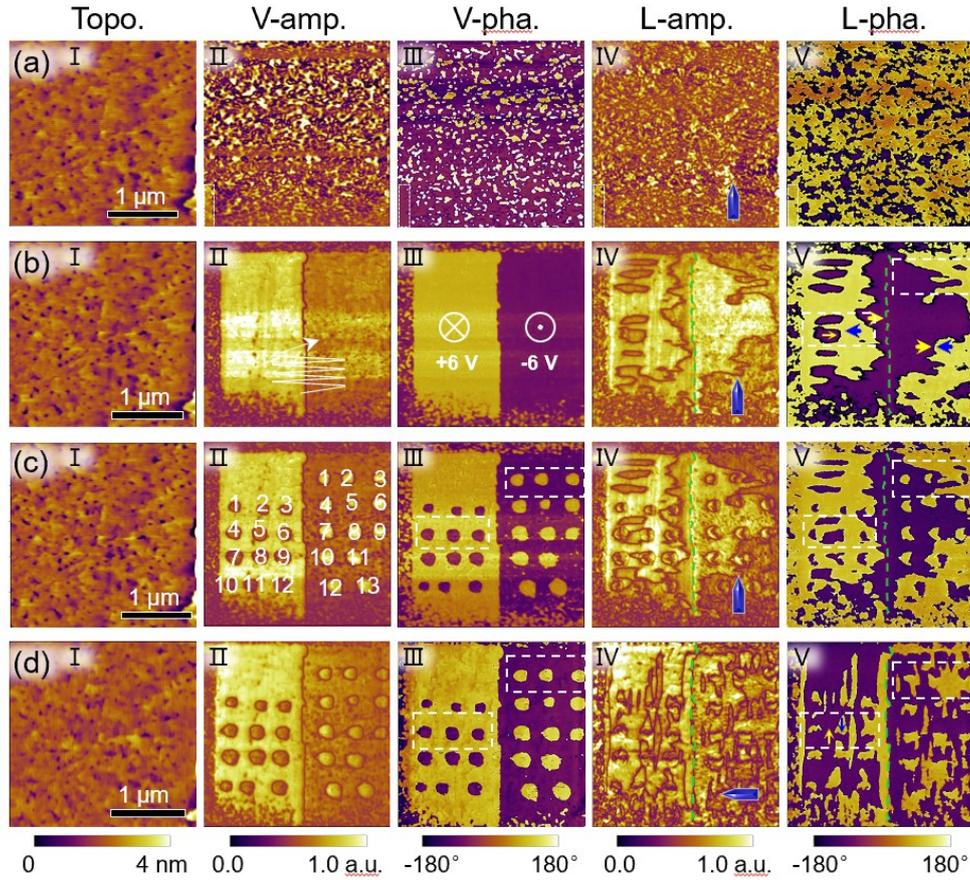
Fig. S8 Vectorial PFM images of center-domains created by area and single-point writing for 'mosaic-like' film. (a-d) The topography, VPFM amplitude and phase images, LPFM amplitude and phase images, for three different states: (a) as-grown, (b) after area writing, and after single-point writing with (c) 0° and (d) 90° sample rotation angles. The white zigzag line with arrow in (b) indicates the scanning direction of the probe. The green dashed lines in (b), (c) and (d) correspond to the written boundary with positive and negative bias. The rectangular region surrounded by white dashed lines in (b), (c) and (d) correspond to the selected single-point writing results of Fig. 4 in the main text. The numbers in (c) V-amp. image is note for the single-point writing, left and right arrays correspond to negative and positive pulse, respectively. The detail parameter of writing is given in Table 1.

Table 1. Pulse width and amplitude of single-point writing with number mark in V-amp. image [Fig. S8 (c II)].

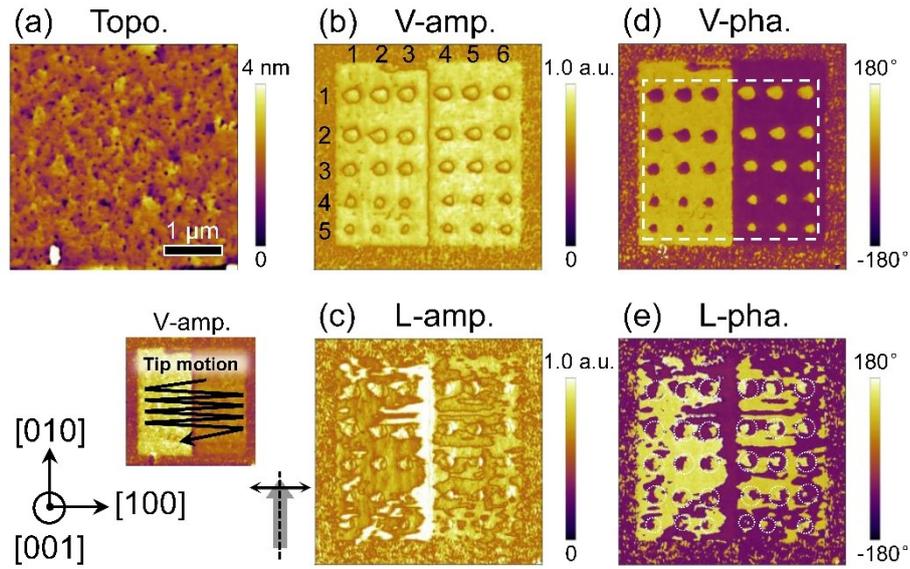| 1	2	3		1		2	3
6 s, -6 V	6 s, -6 V	6 s, -6 V		3 s, +8 V		3 s, +8 V	3 s, +8 V
4	5	6		4		5	6
3 s, -8 V	3 s, -8 V	3 s, -8 V		6 s, +8 V		3 s, +8 V	6 s, +8 V
7	8	9		7		8	9
12 s, -8 V	10 s, -8 V	10 s, -8 V		10 s, +8 V		10 s, +8 V	3 s, +8 V
10	11	12		10		11	
1 s, -8 V	3 s, -8 V	6 s, -8 V		15 s, +8 V		15 s, +8 V	
				12		13	
				20 s, +8 V		20 s, +8 V	

Fig. S9 Vectorial PFM images of center-domains created by area and single-point writing for 'mosaic-like' film with opposite tip motion during area writing process in **Fig. S8**. (a) The topography, VPFM (b) amplitude and (d) phase images, LPFM (c) amplitude and (e) phase images. The pseudocubic axis orientations of the film is also represented. The locations of single-point writing are labeled using numbers in (b) V-amp. image, left and right arrays correspond to negative and positive pulse, respectively. The detail parameter of writing is given in Table 2.

Table 2. Pulse width and amplitude of single-point writing with number mark in V-amp. image [Fig. S9 (b)].

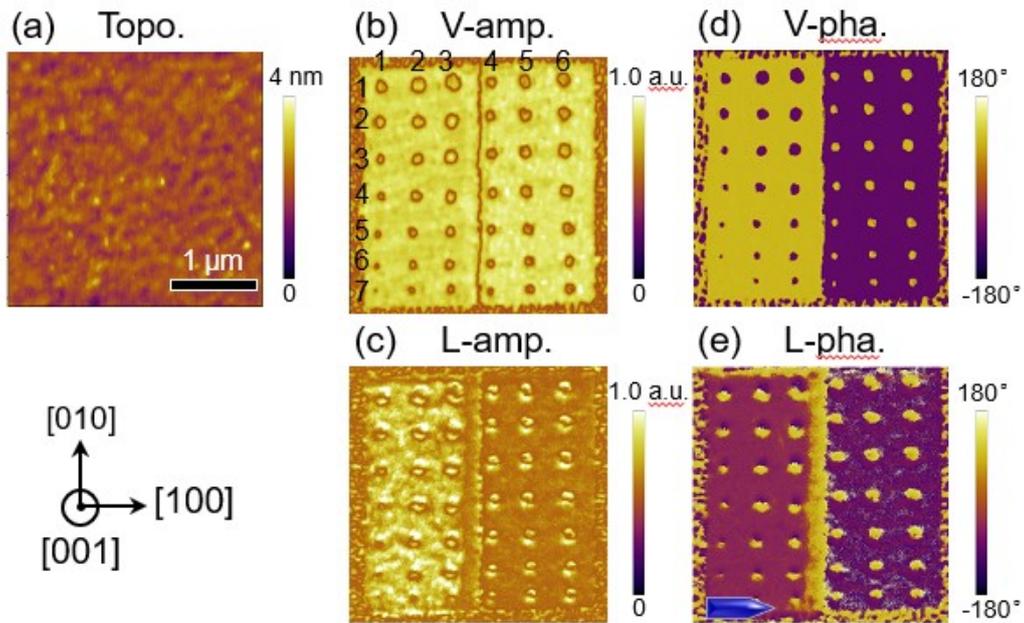|	1	2	3	4	5	6
1	10 s, -8 V	10 s, -8 V	10 s, -8 V	10 s, +8 V	10 s, +8 V	10 s, +8 V
2	6 s, -8 V	6 s, -8 V	6 s, -8 V	6 s, +8 V	6 s, +8 V	6 s, +8 V
3	3 s, -8V	3 s, -8V	3 s, -8V	3 s, +8V	3 s, +8V	3 s, +8V
4	1 s, -8 V	1 s, -8 V	1 s, -8 V	1 s, +8 V	1 s, +8 V	1 s, +8 V
5	0.3 s, -8 V	0.3 s, -8 V	0.3 s, -8 V	0.3 s, +8 V	0.3 s, +8 V	0.3 s, +8 V

Fig. S10 Vectorial PFM images of center-domains created by area and single-point writing for 'stripe-like' film. (a) The topography, VPFM (b) amplitude and (d) phase images, LPFM (c) amplitude and (e) phase images. The pseudocubic axis orientations of the film is also represented.

Table 3. Pulse width and amplitude of single-point writing with number note in V-amp. image [Fig. S10 (b)].

	1	2	3	4	5	6
1	25 s, -5 V	25 s, -7 V	25 s, -9 V	25 s, +5 V	25 s, +7 V	25 s, +9 V
2	18 s, -5 V	18 s, -7V	18 s, -9 V	18 s, +5V	18 s, +7V	18 s, +9V
3	12 s, -5V	12 s, -7 V	12 s, -9 V	12 s, +5V	12 s, +7V	12 s, +9V
4	7 s, -5 V	7 s, -7 V	7 s, -9 V	7 s, +5 V	7 s, +7 V	7 s, +9 V
5	3 s, -5 V	3 s, -7 V	3 s, -9 V	3 s, +5 V	3 s, +7 V	3 s, +9 V
6	1 s, -5 V	1 s, -7 V	1 s, -9 V	1 s, +5 V	1 s, +7 V	1 s, +9 V
7	0.3 s, -5 V	0.3 s,-7V	0.3 s, -9V	0.3 s, +5V	0.3 s, +7 V	0.3 s, +9 V