# **Supporting Information**

## Self-assembly growth of multiferroic topological

### nanoislands array

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**Figure S1. First principal calculation for the energy of R-BFO and T-BFO. a**, Relative energy of R-BFO and T-BFO as a function of the in-plane lattice parameter. It is assumed that the in-plane lattice parameters of the strained films are the same as the substrate. **b**, Energy of R-BFO/LSMO and T-BFO/LSMO with 2-layer and 3-layer BFO. Both of the data points show the lower energy in T-BFO/LSMO model. Insets show the calculation models, where the in-plane lattice constant of LSMO is set to be the same as that of LAO.



**Figure S2. Surface energy of LSMO/LAO (001) system as a function of increasing wetting layer thickness.** Here, LAO and LSMO represent the fully relaxed surface systems respectively. 1 u.c.-, 2 u.c.-, and 3 u.c.-LSMO noting below the x-axis represent these surface systems of 1 u.c., 2 u.c. and 3 u.c. LSMO on the LAO substrate. And the clamped LSMO denotes a LSMO surface structure with its lattice parameters equaling to that of LAO substrate. This calculation results show that the surface energy of LSMO/LAO (001)-oriented surface system first decreases and then increases with increasing LSMO thickness, further hints that the LSMO as a wetting layer is able to induce the change of growth mode from 2D to 3D.



**Figure S3. RSM images for LSMO(20 nm)/LAO heterostructure.** RSM images from the (103) LAO substrate and wetting layer with thickness of about 20 nm, showing that the wetting layer of LSMO appears little relaxation and keeps almost the same in-plane lattice constant with the substrate.



**Figure S4.** Atomic model of the dislocation array on BFO nanoisland from the top view. The red circles represent the intersections of dislocation lines. The yellow and blue balls represent R-BFO nano-island and substrate (LSMO/LAO) respectively.



Figure S5. Cross-sectional STEM image of BFO nanoisland. The perfect arrangement of atoms showing no defects in the nanoisland.



**Figure S6. Dislocation array on the BFO nanoislands.** Low magnification top-view STEM image, indicating dislocations occur in each R-BFO nanoislands.

Figure S7a displays the island density as a function of wetting layer thickness (please see detailed analysis in Note 2). One can see that the island density first increases and then decrease with the increase of wetting layer thickness, well agreement with the experimental result (red curve line in Figure 2b). Figure S7b shows the island density as a function of the growth temperature for different kinetic coefficient. Interestingly, when  $\alpha = 1$ , the increase of island density is very slow within the temperature range from 620 °C to 700 °C. Nonetheless, when  $\alpha > 1$ , especial for > 5, the island density seems to appear rapid increase at the place of larger than 650 °C, consistent with the experimental result at the growth temperature range from ~600 °C to 700 °C (red curve line in Figure 3b). This result indicates that  $\alpha$  should be larger than 1.



**Figure S7. Density evolution of BFO nanoislands**. Thermal dynamics study of the relationship of nanoisland density as a function of wetting layer thickness (**a**) and growth temperature (**b**), respectively.



**Figure S8. BTO nanoislands grown on LSMO (~2 nm) buffered LAO. a, b,** Topography of BTO (~20 nm) in a large scale and a magnified area. **c**, XRD pattern of BTO, showing no impurity phase. The growth conditions are similar with that of BFO nanoislands, but with higher growth temperature (780 °C).

#### Note 1

To reveal the mechanism of the growth mode transformed from 2D to 3D, we calculated the Gibbs system free energy (abbr.  $\Delta$ G) per atom in the following three cases: 3D R-phase, 2D R-phase and 2D T-phase BFO, respectively. According to the classical thermal theory [1, 2], the Gibbs free energy can be expressed by this formula as follows:

$$\Delta G = E_{\text{bulk}} + E_{\text{surface}} + E_{\text{interface}}$$
(1)

Here, E<sub>bulk</sub>, E<sub>surface</sub> and E<sub>interface</sub> represent film or island bulk system formation energy, surface energy and interface energy between film/island and substrate, respectively. Therefore, the Gibbs free energy per atom for 2D T-phase BFO film and 2D R-phase BFO film are defined as:

$$\Delta G = \frac{1}{N} [ghR^2 + (\gamma_{int} + \gamma_f - \gamma_{sub})R^2] \qquad (2)$$

Where N, g, h and R represent atom number, formation energy per unit volume, film thickness, in-plane unit length, respectively, as illustrated by Figure N1a.  $ghR^2$  corresponds to the formation energy of BFO film,  $\gamma_{int}$ ,  $\gamma_f$  and  $\gamma_{sub}$  denote the corresponding interface energy, film surface energy and substrate surface energy.

Notably, the growth of 3-D R-phase involved the process of nucleation. The geometry structure of R-phase island is a typical trapezoid four prism, as demonstrated in our previous work [3]. Therefore, we conjecture the geometry structure of the R-phase nucleation is a typical trapezoid four prism, as shown by Figure N1b. The corresponding Gibbs energy can be written by this equation:

$$\Delta G = \frac{g}{6} (R^3 - r^3) + r^2 \gamma_{top} + \sqrt{2} \gamma_{side} (R^2 - r^2) + (\gamma_{interface} - \gamma_{sub}) R^2$$
(3)

Thus, the Gibbs energy of 3D R-phase island per atom can be expressed by the formula:

$$\Delta G_{3D-R} = \frac{1}{N} \left[ \frac{g}{6} (R^3 - r^3) + r^2 \gamma_{top} + \sqrt{2} \gamma_{side} (R^2 - r^2) + (\gamma_{interface} - \gamma_{sub}) R^2 \right]$$
(4)

Solving the minimum for the equation (3), we can obtain the critic energy of nucleation by this formula:

$$\Delta G^{*}_{3D \ R-phase} = -\frac{g}{12} (R^{*3} - r^{*3}) = \frac{16}{3g^{2}} \left[ \left( \sqrt{2} \gamma_{side} + \gamma_{interface} - \gamma_{sub} \right)^{3} - \right]$$

$$\left(\sqrt{2}\gamma_{side} - \gamma_{top}\right)^3] \quad (5)$$

Here, g, R\* and r\* denote the formation energy per unit volume, critical lengths of bottom and top sides, respectively.  $\gamma_{side}$ ,  $\gamma_{top}$ ,  $\gamma_{sub}$  and  $\gamma_{interface}$  represent the side and top surface energies of 3D R-phase island, surface energy of LSMO/LAO system and the interface energy of R-BFO/LMSO/LAO system, respectively. By calculating for these parameters (formation energy, surface energy and interface energy), the relative energy of 3D R-BFO as a function of wetting layer thickness can be drawn, as shown in Figure 5.



Figure N1. A schematic of geometry structure on the 2D-film (a) and 3D-island (b). Here, r, R and h denote the top side, bottom side length and height, respectively.

#### Note 2

To further explain the phenomenon of tailoring the density of R-BFO nano-island by the wetting layer thickness and growth temperature, as shown in Figure 2b and 3b, we analyze the function relationship between island density and wetting layer thickness and growth temperature. According to classical nucleation theory [4, 5], island density can be expressed by this formula:

$$\mathbf{n} = n_0 e^{-\frac{\Delta G^*}{KT}} \quad (6)$$

Here, n,  $n_0 \Delta G^*$ , K and T represent island density, coefficient, nucleation barrier, Boltzmann constant and growth temperature, respectively. According to the law of the equation (6), R-BFO islands will appear on the LAO substrate without the wetting layer and at the growth temperature below 650 °C. Simultaneously, there will not display a sharp increase. However, the island density experimentally behaves an unexpected result, as shown in Figures 2b and 3b. This result implies that the island density (n) should contain a stimuli term related with the growth temperature. According to the classical nucleation theory [4-5], the corresponding expression can be written by this formula:

$$n = n_0 e^{\frac{-\Delta G^*}{KT}} e^{\alpha \frac{KT - E_d}{KT}} \quad (7)$$

Here,  $\alpha$  and  $E_d$  denote a kinetic coefficient and barrier energy per atom from 2D to 3D mode.

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