Supplementary Materials

Surface-Step-Terrace Tuned Second-Order Nonlinear Optical Coefficients of Epitaxial Ferroelectric $BaTiO_3$ Films

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The AFM and PFM images were characterized by commercially available AFM platform (Asylum Research (MFP-3D-infinity) and Bruker (Dimension Icon AFM)), and the crystalline structure were characterized via X-ray Diffraction (Rigaku-D/Max 2500 and BEDE D1 system). The cross-sectional TEM image were carried out by high-resolution transmission electron microscopy (TEM, JEM 2100F).

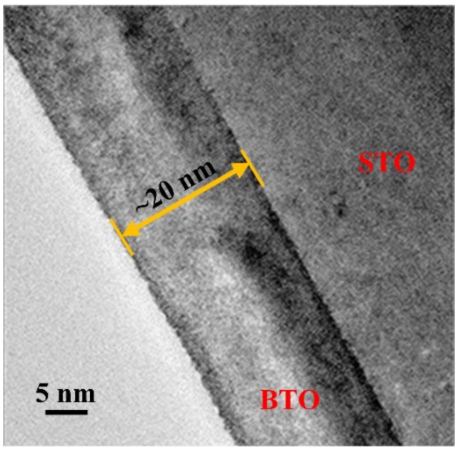

FIG. S1 Cross-section TEM image of BTO film grown on 0° miscut substrate.

Cross-sectional TEM image of BTO film grown on 0° miscut substrate are shown in FIG. S1. It can be found that the thickness of BTO film is about 20 nm.

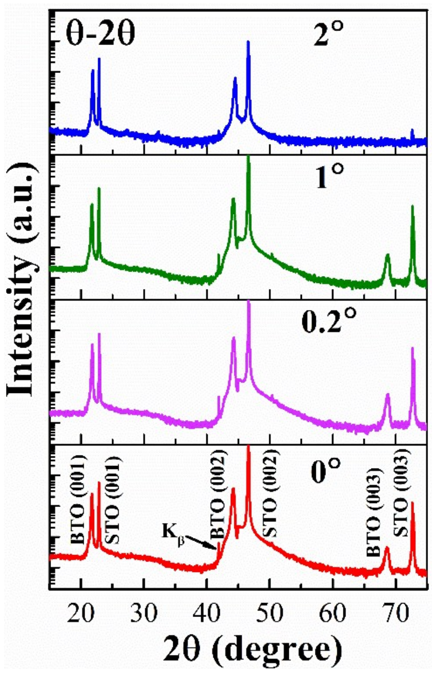

FIG. S2 Typical θ-2θ XRD patterns of BTO films grown on various (001) STO miscut substrates.

The typical θ-2θ XRD patterns of BTO films grown on various (001) STO miscut substrates are shown in FIG. S2. The peak at 41.8° corresponds to the STO substrate (002) K_β reflection.

The sharp (00*l*) diffraction peaks suggest that the films have good single-crystal quality and excellent epitaxial nature, and the phase structure of the films is tetragonal.

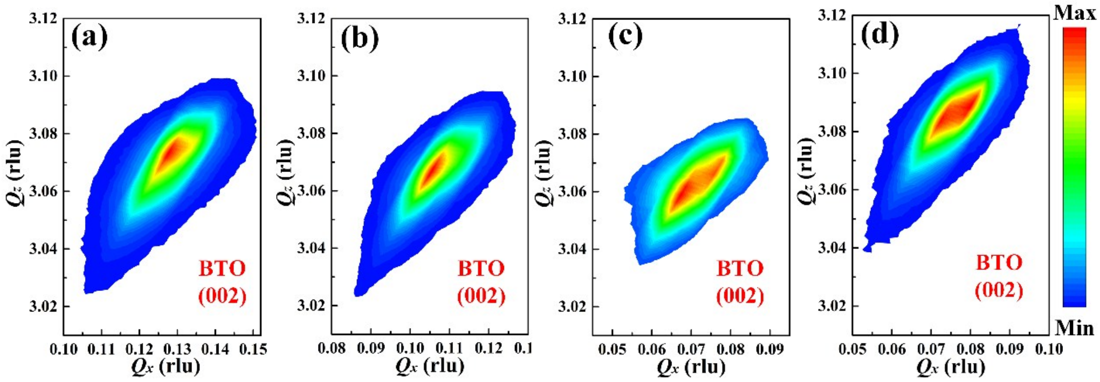

FIG. S3 RSM patterns of (002) reflections from BTO film grown on (a) 0°, (b) 0.2°, (c) 1° and (d) 2° miscut substrates.

The RSM patterns of (002) reflections from BTO film grown on 0°, 0.2°, 1°, and 2° miscut substrates are shown in FIG. S3. As can be seen, the BTO films grown on different miscut angles present different value of Q_z, which indicates different lattice parameter *c* in the four BTO samples. Therefore, the BTO samples exhibit different strain states induced by surface-step-terrace.

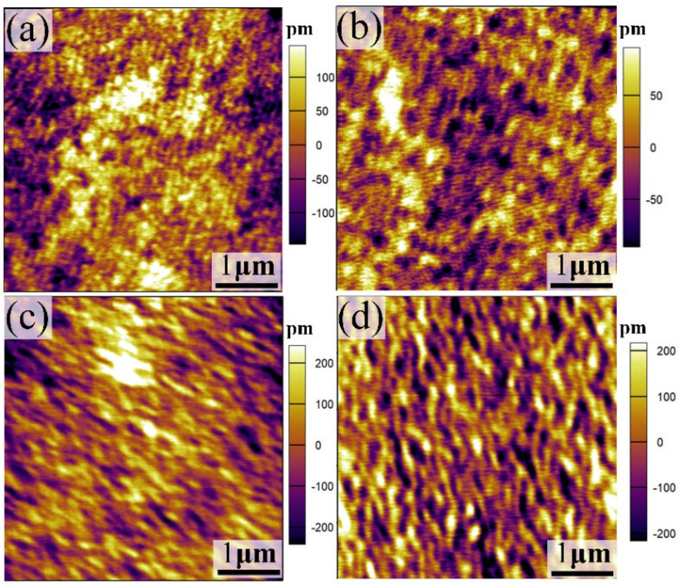

FIG. S4 Atomic force microscope images of BTO films grown on (a) 0°, (b) 0.2°, (c) 1° and (d) 2° miscut substrates.

The morphology of BTO surfaces was studied by atomic force microscope. The results of 5 μm×5 μm area scan are shown in FIG. S4(a)-(d). As can be seen, the roughness of BTO films is less than 0.5 nm and the terraces from the substrate are visible, which indicates a layer-by-layer mode of the film growth.

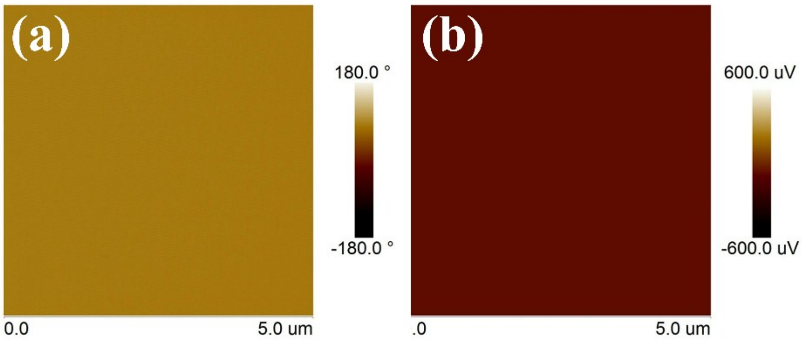

FIG. S5 Out-of-plane and in-plane PFM image of BTO film grown on 2° miscut substrate.

The out-of-plane (OP) and in-plane (IP) PFM images of BTO film grown on 2° miscut substrate are shown in FIG. S5(a) and (b), respectively. The uniform contrast of OP PFM image indicates the polarization components along *c* direction are the same. No signal above the noise level can be obtained from the IP PFM image, which means that there is no IP component of the polarization.[1] Therefore, we can conclude that the domain structure of BTO film grown on 2° miscut substrate is monodomain. The PFM results from other samples show similar phenomenon.

Reference:

1 Y. H. Chu, L. W. Martin, M. B. Holcomb and R Ramesh, *Mater. Today*, 2007, **10**, 16-23.