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Supplementary: Enhanced Piezoresponse and Dielectric Properties for $Ba_{1-x}Sr_xTiO_3$ Composition Ultrathin Films by the High-Throughput Method

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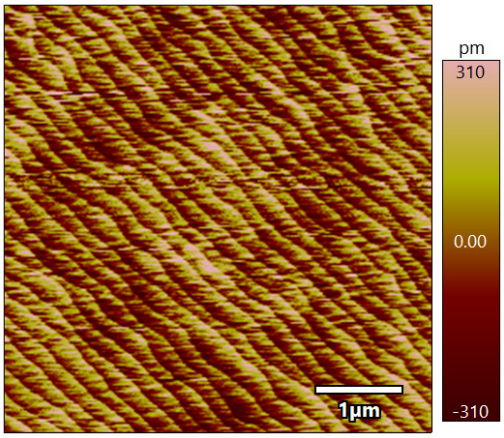

Figure S1. Surface images of Nb:STO substrate with atomic-terrace.

Citation: Zhang, N.; Wang, D.; Wang, J.; Fang, H.; He, B.; Guo, J.; Han, Y.; Zhang, P.; Shi, C.; Chen, Y.; et al. Enhanced Piezoresponse and Dielectric Properties for $Ba_{1-x}Sr_xTiO_3$ Composition Ultrathin Films by the High-Throughput Method. *Coatings* **2021**, *11*, 1491. https://doi.org/10.3390/coatings11121491

Received: 3 November 2021
Accepted:29 November 2021
Published: 3 December 2021

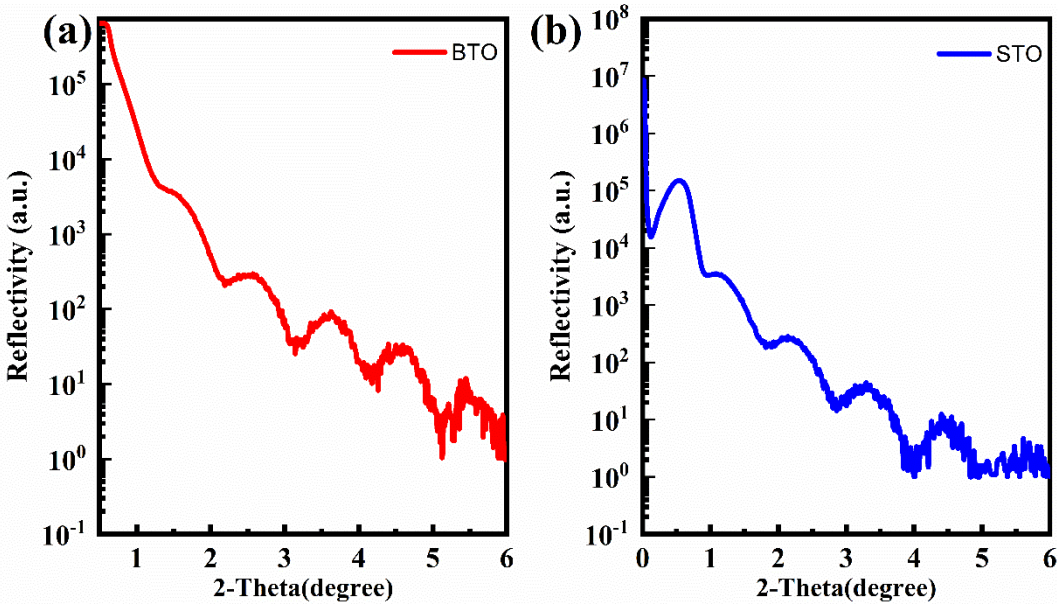

Figure S2. X-ray reflectometry measurements for BTO (**a**) and STO (**b**).

Figure S2 shows XRR patterns for BTO film and STO film which use a commercial software to simulate and calculates reflectivity curves from the first principles based on a physical model of the film stack. Then we can obtain the thickness value of film. The thickness of each layer is controlled by the number of laser pulses. Thus BTO and STO growth rates were confirmed at about 27 and 29 pulses per unit.

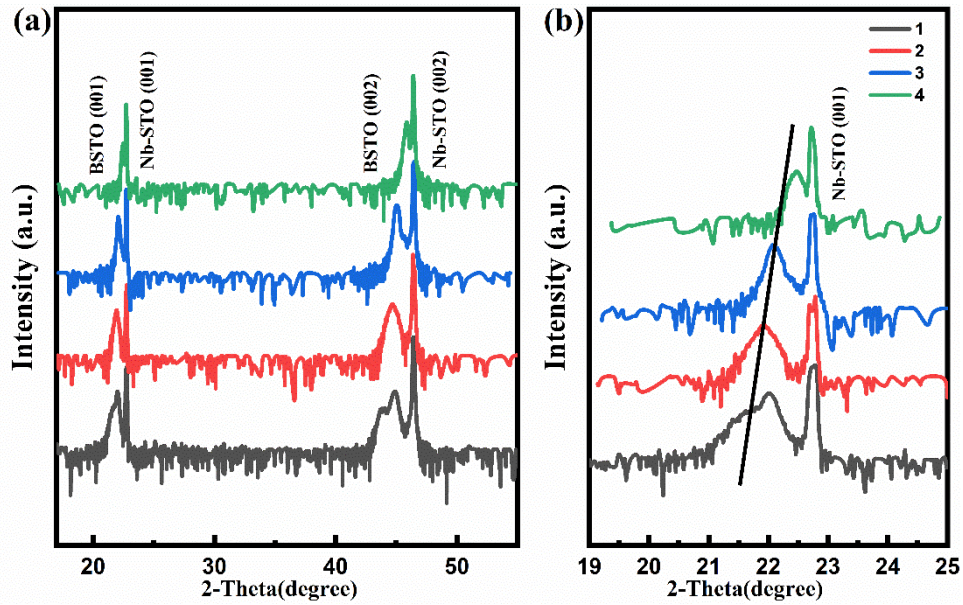

Figure S3. (**a**) XRD measurements of the BSTO thin film (80 nm) of four representing positions with increasing Sr content. (**b**) XRD pattern around (001) reflection (From Part 1 to 4, Sr content increases).

The XRD patterns of BSTO films on the Nb–STO substrate are shown in Figure S3. As Sr increasing, the BSTO (001) peak shifts closer to Nb–STO (001) peak as shown in Figure S3 (b). The shift is consistent to the varying composition. Compare to bulk BTO value, the c lattice constants decreased. The shift is consistent to the varying composition, indicating that the BSTO composition spread thin film can successfully be fabricated by LMBE with Mask.