Supporting information for

# Piezoelectric and Dielectric Properties of Multilayered $\mathrm{BaTiO}_{3} /(\mathrm{Ba}, \mathrm{Ca}) \mathrm{TiO}_{3} / \mathrm{CaTiO}_{3}$ Thin Films 

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Table S1 The bulk lattice constant of every composition layer and substrate.

| Compositions | $\mathrm{a}(\AA)$ | $\mathrm{b}(\AA)$ | $\mathrm{c}(\AA)$ |
| :--- | :---: | :---: | :---: |
| $\mathrm{BaTiO}_{3}$ | 3.997 | 3.997 | 4.037 |
| $\mathrm{CaTiO}_{3}$ | 5.387 | 5.439 | 7.646 |
| $($ Orthohombic $)$ | $(3.809$ in cubic $)$ | $(3.846$ in cubic $)$ | $(3.823$ in cubic $)$ |
| $\left(\mathrm{Ba}_{0.85} \mathrm{Ca}_{0.15}\right) \mathrm{TiO}_{3}$ | 3.972 | 3.972 | 4.019 |
| $\left(\mathrm{Ba}_{0.75} \mathrm{Ca}_{0.25}\right) \mathrm{TiO}_{3}$ | 3.959 | 3.959 | 4.003 |
| $\mathrm{Nb}: \mathrm{SrTiO}_{3}$ | 3.905 | 3.905 | 3.905 |

According to Bragg equation:

$$
\begin{equation*}
2 \mathrm{~d}_{\mathrm{hicl}} \sin \theta-\mathrm{K} \lambda \tag{S1}
\end{equation*}
$$

The $\mathrm{d}_{002}$ of peak1, Peak2, and Peak3 can be calculated, and $\mathrm{d}_{001}\left(\mathrm{~d}_{001}=2 \mathrm{~d}_{002}\right)$ corresponds to the out of plane constant $\mathrm{c}_{\text {measured }}$. Based on the unchanged volume of each layer, the in plane constant $\mathrm{a}_{\text {film }}$ can be calculated from equation S 2 :

$$
\begin{equation*}
a_{\text {buik }} \times b_{\text {bulk }} \times c_{\text {buik }}=a_{\text {film }} \times b_{\text {film }} \times c_{\text {measured }} \tag{S2}
\end{equation*}
$$

Where the $a_{\text {bulk }}, b_{\text {bulk }}$ and $c_{\text {bulk }}$ of each layer can be obtained from Table S 1 , the Cmeasured is the calculated $d_{001}$, thus we can get the in plane constant of each layer $\mathrm{a}_{\text {film }}=b_{\text {film }}$.


Figure S1. Comparison of SS-PFM (a) and conventional PFM (b) of amplitude-voltage butterfly loops and phase-voltage hysteresis loops for $\mathrm{CT}^{(1)} / \mathrm{BCT}^{(15} 5^{(1)} / \mathrm{BT}^{(1)}$ sample.


Figure S2. Distribution of amplitude in Figure 3 and Figure 4 for (a) $\mathrm{S} 1: \mathrm{CT}^{(1)} / \mathrm{BCT}^{(1)}{ }^{(1)} / \mathrm{BT}^{(1)}$ (b)S2: $\mathrm{CT}^{(1)} / \mathrm{BCT}^{(15} 5^{(1)} / \mathrm{BT}^{(2)}$ (c) $\mathrm{S} 3: \mathrm{CT}^{(1)} / \mathrm{BCT}^{2} 5^{(1)} / \mathrm{BT}^{(1)}(\mathrm{d}) \mathrm{S} 4: \mathrm{CT}^{(1)} / \mathrm{BCT}^{2} 5^{(1)} / \mathrm{BT}^{(2)}$.


Figure S3. Distribution (a) of amplitude (b) and phase contrast in Figure 5.

