Supplementary Information for

Thickness-dependent phase boundary in Sm-doped BiFeO\textsubscript{3} piezoelectric thin films on Pt/Ti/SiO\textsubscript{2}/Si substrates

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The Raman mapping results of the selected samples are shown in Fig. S1(a)-(f) in a mixing phase appearance. Raman profiles of two selected points from $R$-rich region and $O$-rich region are depicted in Fig. S1(g) and S1(h), respectively. It can be seen that both $R$ and $O$ phases exist in the two regions in spite of different phase intensity.

Surface morphology images of BS10F-60nm to BS10F-420nm samples are shown in Fig. S2(a)-(g). All the seven topographic images are subjected to the same scale bar. No thickness dependent change for roughness or grain size was found according to AFM images.

X-ray photoelectron spectroscopy (XPS, ESCALAB 250Xi, Thermo Scientific, USA) was conducted on BS10F-300nm sample to verify the element information of thin films. The full spectrum and individual element’s spectra are presented in Fig. 3(a)-(d). XPS study clearly confirms the presence of different elements in the prepared Sm-doped BiFeO$_3$ system.

Fig. S4(a)-(c) display the XRD and Raman profiles of the four selected samples whose piezoelectric response loops were compared in Fig. 5. From step scanning of (200)$_{pc}$ peak and Raman spectra, it can be concluded that both $R$ and $O$ phases exist in BS11F-420nm sample according to the discussion in the main text. The different (200)$_{pc}$ peak shape of BS11F-420nm sample between Fig. 1(b) and Fig. S4(b) is resulted from different X-ray sources, of which one is synchrotron X-ray and the other is Cu-$K_\alpha$ containing both $K_{\alpha1}$ and $K_{\alpha2}$. 
Fig. S1. (a)-(f) Raman maps (phase-mixed mode) collected from (a) BS10F-420nm, (b) BS11F-420nm, (c) BS12F-420nm, (d) BS10F-120nm, (e) BS11F-120nm and (f) BS12F-120nm. Raman spectrum of (g) R-rich region and (f) O-rich region. Limits were set in the Raman shift between approximately 120 to 160 cm\(^{-1}\) and 260 to 320 cm\(^{-1}\) to define contributions from the R and O phases respectively.

Fig. S2. Surface morphology images (a) BS10F-60nm, (b) BS10F-120nm, (c) BS10F-180nm, (d) BS10F-240nm, (e) BS10F-300nm, (f) BS10F-360nm and (g) BS10F-420nm.
Fig. S3. (a) XPS spectra of BS10F-300nm sample. (b) Fe 2p peaks, (c) Sm 3d peaks and (d) Bi 4f peaks.

Fig. S4. (a) XRD profiles, (b) step scannings of (200)$_{pc}$ peak and (c) Raman spectra of BS11F-420nm, BS9F-180nm, BS13F-360nm and BS15F-120nm samples.