

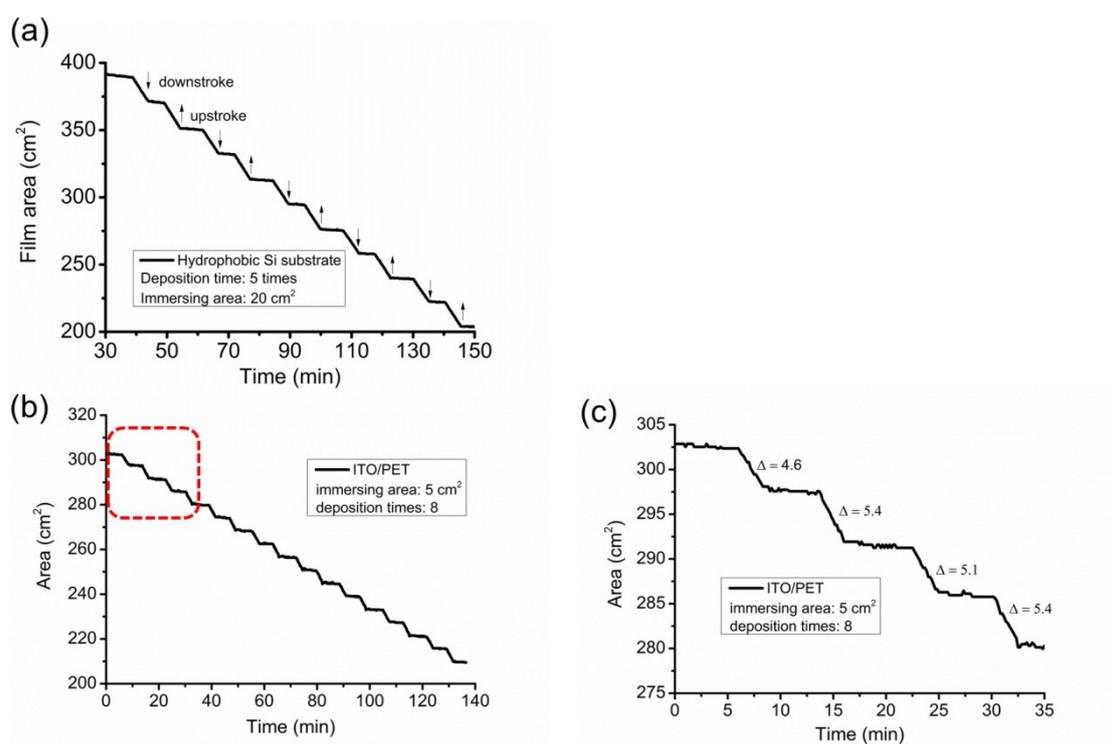
## Supplementary Information

### Highly Oriented Poly(vinylidene fluoride-co-trifluoroethylene) Ultrathin Films with Improved Ferroelectricity

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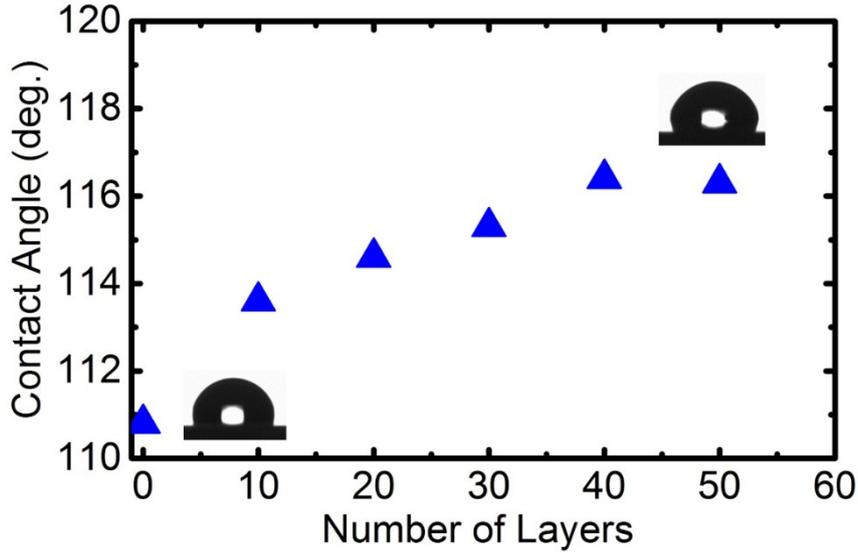
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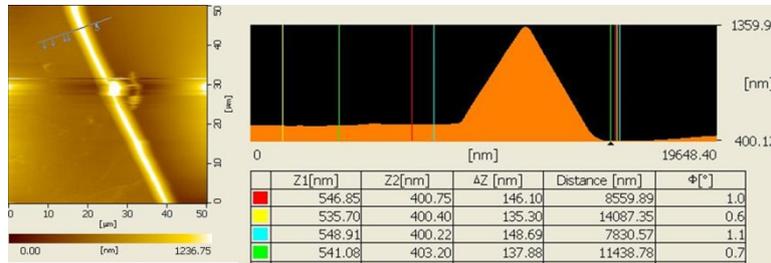


**Fig. S1** Deposition curves of P(VDF-TrFE)/pDDA LB nanofilms onto (a) a hydrophobic silicon substrate for five-time deposition (10 layers) and (b) an ITO-coated PET substrate for eight-time deposition (16 layers), and (c) the first two times of the deposition on the ITO-coated PET substrate.

The uniform decrease for each deposition cycle indicates a regular deposition procedure onto different substrates. The transfer ratio is the quotient of the reduced film area during each deposition cycle divided by the immersing surface area of the substrate into water subphase. Therefore, the transfer ratios on both the PET substrate and the silicon substrate are unity for both the downstroke and upstroke.



**Fig. S2** Development of water contact angle with different P(VDF-TrFE)/pDDA LB nanofilm thicknesses.



**Fig. S3** AFM images of 40-layer P(VDF-TrFE)/pDDA LB nanofilms (50:1) on a hydrophobic silicon substrate.

**Table S1.** Ferroelectric parameters of five-layer P(VDF-TrFE)/pDDA LB nanofilms measured at 10 Hz.

<i>Voltage</i> (V)	5	10	15
$P_r$ ( $\mu\text{C cm}^{-2}$ )	0.18	2.7	5.0
$E_c$ ( $\text{MV m}^{-1}$ )	18	142	328

**Table S2.** Ferroelectric parameters of a five-layer P(VDF-TrFE)/pDDA LB nanofilms measured at 15 V.

<i>Freq.</i> (Hz)	<b>10</b>	<b>100</b>
<i>P<sub>r</sub></i> ( $\mu\text{C cm}^{-2}$ )	5.0	2.8
<i>E<sub>c</sub></i> (MV m <sup>-1</sup> )	328	240