

## Electronic Supplementary Information (ESI)

### Three dimensionally-ordered 2D MoS<sub>2</sub> vertical layers integrated on flexible substrates with stretch-tunable functionality and improved sensing capability

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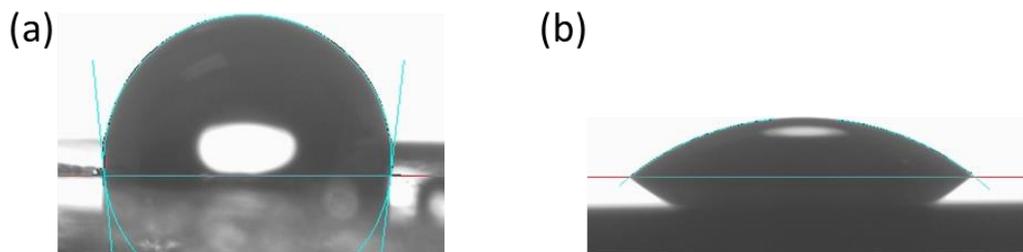
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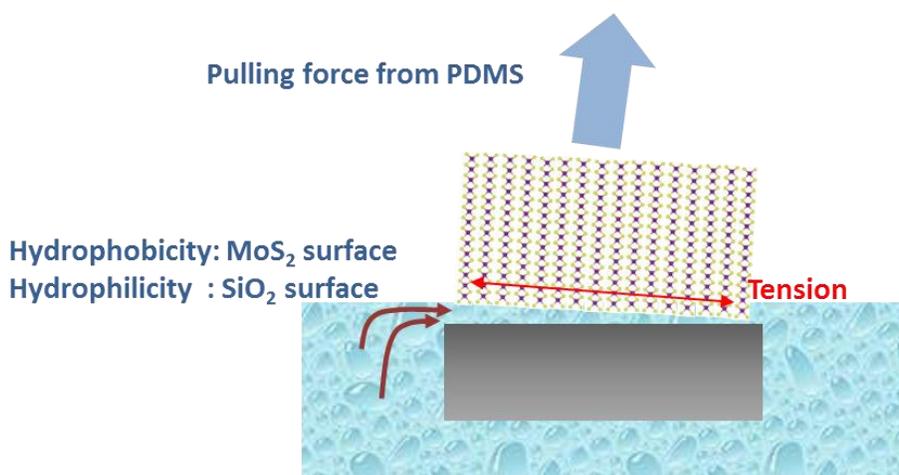
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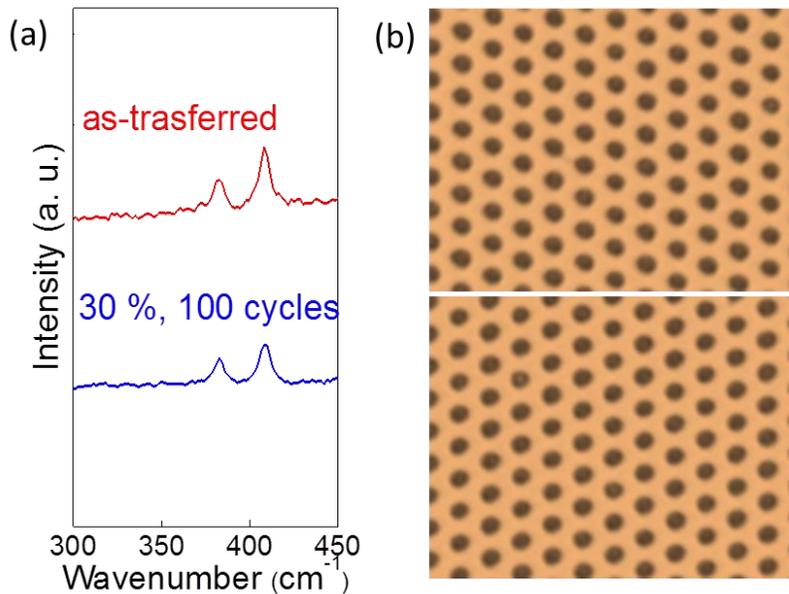
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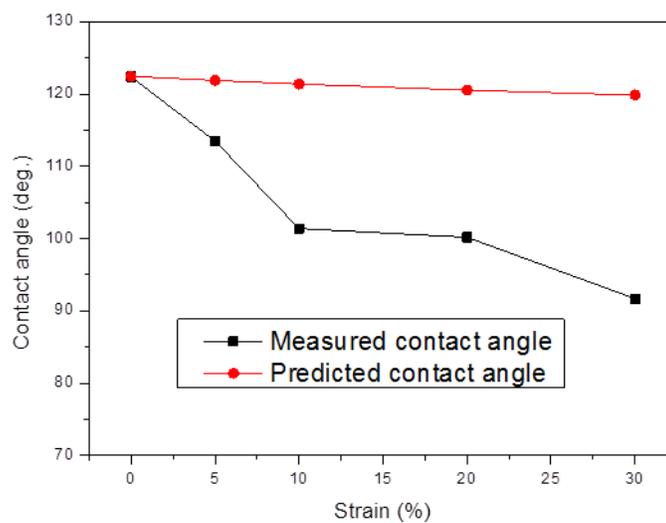
**Figure S1.** Water contact angle measurements of (a) 2D MoS<sub>2</sub> layers with vertically-aligned layers, and (b) SiO<sub>2</sub> substrate revealing hydrophobicity and hydrophilicity, respectively.



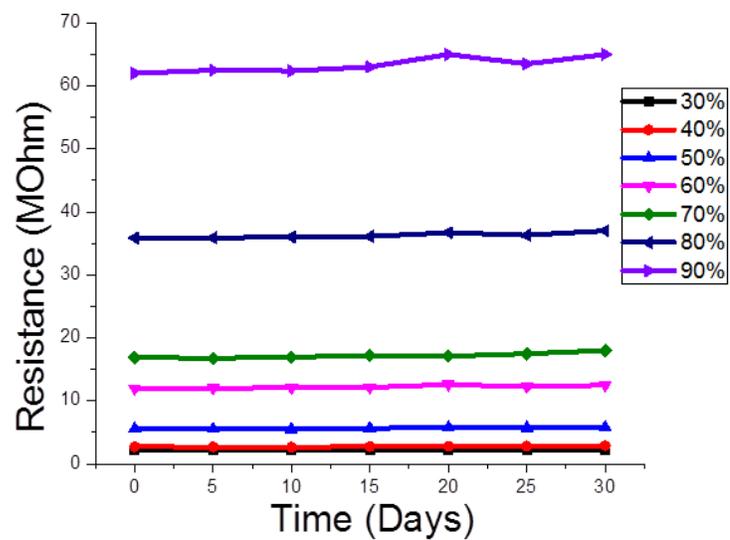
**Figure S2.** Schematic illustration to demonstrate the facile separation of vertically-aligned 2D layers assisted by the water wettability difference of SiO<sub>2</sub> vs. MoS<sub>2</sub> and the pullinf force exerted by PDMS.



**Figure S3.** (a) Raman profiles of 2D vertical MoS<sub>2</sub> layers/PDMS before (red) and after (blue) repeated tensile stretch. (b) Optical microscopy images of the sample before (top) and after (bottom) tensile stretch.



**Figure S4.** Comparison of water contact angle values obtained from experimental measurements (black dots) vs Wenzel equation prediction (red dots).



**Figure S5.** Long-term stability of vertically-aligned 2D MoS<sub>2</sub> layers for humidity sensing.