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Ultra-Thin Films of Solution-Exfoliated Hexagonal Boron Nitride by Langmuir Deposition

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Figure S1. SEM image of bulk hBN flakes on Si substrate. The scale bar is 2 $\mu\text{m}.$



Figure S2. Microscopy of multi-layer hBN films. SEM images of (a) D1, (b) D2, and (c) D3 films of as-prepared hBN films on Si substrate. Analyzed SEM images of (d) D1 and (e) D2 samples using image J software. Red area shows the Si substrate, (f) D1-film pixel distribution as a function of pixel brightness, along greyscale values of 0-255. Exposed Si substrate has a sharp narrow peak at lower brightness level as a result of the charging effect. The scale bar is 1 µm and it is effective for SEM images in (b)- (e).



Figure S3. (a) Optical microscopy image of the sparse coatings of the hBN film deposited on Si substrate from hBN dispersion exfoliated in DCE and ethanol. The scale bar is 50 $\mu m.$



Figure S4. AFM images of exfoliated hBN sheets resulted from (a) Method A, (b) Method B and (c) Method C prepared by spin coating on Si substrates. The scale bar is 500 nm.



Figure S5. Raman spectra of bulk hBN powder and LB film prepared from exfoliated hBN by Method C with legends indicated in the graph. Silicon substrates were used in this measurement.



Figure S6. Photograph of unsuccessful hBN LB film. The patchy texture visible in the image illustrates the voids in the film between densely aggregated (in 2D) hBN islands even after compression to the extent that subphase water flowed over the edge of Teflon trough which is noted in the photo.



Figure S7. Optical band gap energy evaluated by Tauc analysis on the ellipsometry data.



Figure S8. Device structure schematic of the investigated hBN capacitor, including a highly doped silicon substrate, a bottom contact, and a patterned top contact.

As shown in Table S1, a clear equivalence between boron and nitrogen was obtained by analyzing EDS mapping results. The presence of carbon and oxygen is expected considering that carbon is a byproduct of this measurement technique, and oxygen is likely a result of operating at low vacuum.

Table S1. Composition of hBN film (D4) obtained using EDS mapping.				
	Element	At. %		
	Boron	42.63		
	Nitrogen	42.90		
	Carbon	11.96		
	Oxygen	2.52		
	Total	100.00		

Table S2. Height distribution statistics and surface parameters of hBN film calculated from AFM images of D1, D2, and D3 samples.

	D1	D2	D3
Maximum (nm)	170.2	232.9	266.2
Ave value (nm)	24.8	41.2	59.4
Median (nm)	21.0	35.6	57.0
Ra (Sa) (nm)	9.8	17.5	20.8
Rms (Sq) (nm)	14.4	24.0	27.1
Skew	2.6	1.8	1.059