Supporting Information

Efficient Liquid Nitrogen Exfoliation of MoS₂ Ultra-Thin Nanosheets in Pure 2H Phase

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This supporting information contains 1 table and 15 figures in 12 pages.

1 Experimental

Chemicals. Molybdenum sulfide (MoS_2) powders (with flake sizes about 2 µm, purity of 99%) was purchased from Sigma-Aldrich. 3,4,9,10-perylenetetracarboxylic acid anhydride (PTCA), N-methyl-2-pyrrolidone (NMP) (99%), isopropanol (IPA) (99%), chloroform (99%), and ethanol (99%) were purchased from Sigma-Aldrich. Other chemicals are of analytical grade and used without further purification. Liquid nitrogen (L-N₂) was purchased from the company of Yehao gas.

Characterization. The morphology of the prepared MoS_2 nanosheets was characterized using a JOEL JEM-1230 transmission electron microscope (TEM) and atom force microscopy (AFM) equipped with a Dimension 3100 (Veeco, CA) in Height mode. High-resolution TEM (HRTEM) characterization was performed on a TECNAI G2 F20 S-Twin delectron microscope. Scanning transmission electron microscopy (STEM) investigations were carried out using a spherical aberration-corrected microscope (JEOL JEM-ARM 200F) with a DCOR probe corrector (CEOS GmbH) at 200 kV. The Raman studies of the films were carried out on a WITec CRM200 confocal Raman microscopy system with a laser excitation energy of 488 nm and an air cooling charge coupled device (CCD) as the detector. The scanning electron microscopy (SEM) images were obtained by Hitachi S-4800. X-ray photoelectron spectroscopy (XPS) data were collected using a PHI 3056 spectrometer with an Al anode source operated at 15 kV and an applied power of 350 W with samples mounted on indium foil. X-ray diffraction (XRD) patterns were obtained using a Rigaku miniflex II. UV-Vis absorption spectra was obtained from a Shimadzu UV-3600 spectrophotometer. The photograph was taken using a commercial Nikon camera.

2 Table Part

| Table | S1 . | Summary | of t | the | recently | reported | nonvolatile | WORM | memory | device |
|---------|-------------|---------|------|-----|----------|----------|-------------|------|--------|--------|
| perform | nanc | ce | | | | | | | | |

| Active layer ^a | ON/OFF ratio | Retention time | Reference |
|---------------------------|-------------------|-----------------------|-----------|
| PVK | 104 | 10 ⁴ s | [1] |
| BTDBPI | 104 | 3.6×10 ³ s | [2] |
| Au NPs:lignin | 5×10 ³ | 10 ³ s | [3] |
| PI(TPF-Ph-OMe) | 10 ⁵ | 3.6×10 ³ s | [4] |
| Cu _x O | 104 | 10 ⁴ s | [5] |
| CsPbBr ₃ | 104 | 1.2×10 ³ s | [6] |
| AlO _x -native | 104 | 10 ³ s | [7] |
| MoS ₂ -PCBM | 10 ² | / | [8] |
| MoS ₂ -PTCA | 10 ⁵ | 10 ⁴ s | This Work |

^a PVK is Poly(9-vinylcarbazole), BTDBPI is aromatic polyimide, Au NPs:lignin is gold nanoparticles embedded alkali lignin, PI(TPF-Ph-OMe) is poly(4,4'-(2,7-bis(3,5-dimethoxyphenyl)-9H-fluorene-9,9-diyl)dianiline), PCBM is [6,6]-phenyl-C₆₁-butyric acid methyl ester.

3 Figure parts



Figure. S1 SEM image of MoS₂-5N acquired after centrifugation at 1500 rpm.



Figure. S2 (a) SEM images and (b) the corresponding enlarged images of bulk MoS₂;

(c) SEM images and (d) the corresponding enlarged images of $p-MoS_2-5N$.



Figure. S3 TEM images of (a) MoS_2 -3N and (b) MoS_2 -10N.



Figure. S4 HRTEM image of MoS₂-5N.



Figure. S5 STEM image of MoS_2 -5N. From dark to yellow, the layer thickness of MoS_2 increases.



Figure. S6 AFM image and height profile (inset) of MoS₂-5N.



Figure. S7 Thickness (a) and lateral size (b) distributions of MoS₂-5N.



Figure. S8 TEM image (a) and corresponding lateral size distribution (b) of MoS₂-5N.



Figure. S9 AFM height profiles (a) and image (b) of MoS_2 -3N.



Figure. S10 XPS survey spectrum of MoS₂-5N.



Figure. S11 Enlarged XRD patterns of bulk MoS₂ and MoS₂-5N.



Figure. S12 Enlarged XRD patterns of different MoS₂-5N.



Figure. S13 Concentration versus exfoliation time of the liquid nitrogen exfoliation of

 $MoS_{2}. \\$



Figure. S14 I–V characteristics of the ITO/ PTCA (x wt %)-MoS₂/Al diode memory devices, where x = 0, 5, 15, 30, 50 and 100 for panels of (a), (b), (c), (d), (e) and (f), respectively.



Figure. S15 Raman spectra (a) and XRD patterns (b) of the freshly prepared PTCA (5 wt %)-2H MoS₂ composite and that after store at room temperature for 1 month.

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