Supporting information for

Free-Floating Synthetic Nanosheets by Atomic Layer Deposition

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1. Experimental details

1.1. Sacrificial Polymer layer. Poly (acrylic acid) (PAA, Mw = 1,800) and poly (methyl methacrylate) (PMMA, Mw = 16,000 and 350,000) were purchased from Sigma-Aldrich. Poly (vinyl alcohol) (PVA, Mw = 16,000 with 98% hydrolysis and 95,000 with 95% hydrolysis) was purchased from Acros Organics. All polymers were used without further purification. The solvent for PVA and PAA was deionized (DI) water. PMMA was dissolved in toluene (anhydrous 99.8%) or tetrahydrofuran (THF, anhydrous 99.9%) purchased from Sigma-Aldrich. After dissolved in the solvents with 2 wt% concentration, they were spin-coated onto a Si/SiO_2 substrate for 30 s at 3000 rpm.

1.2. Atomic Layer Deposition. Atomic layer deposition was performed in a custom hot wall viscous flow tube reactor where each precursor was dosed alternatively at an operating pressure of 2 Torr. Nitrogen gas as the carrier and purge gas was purified before introducing into the reactor with an inert gas filter (Gatekeeper®, Entegris Inc.). Precursors for Al_2O_3 and ZnO were Trimethylaluminum (TMA, 98%) and diethylzinc (DEZ, 95%) respectively. Titanium tetrachloride ($TiCl_4$, 99%) was the precursor for TiO_2 ALD. All precursors were purchased from

Strem Chemicals, Inc. Deionized (DI) water is the co-reactant for all ALD processes. ALD sequences are consisted of [TMA/N_2/H_2O/N_2] = [1/30/1/60 s] and [DEZ/N_2/H_2O/N_2] = [2/50/2/50 s] for Al_2O_3 and ZnO films, respectively. Partial pressure for TMA and DEZ was 0.1–0.2 Torr and deposition temperature was 90 °C. TiO_2 film was deposited with the sequence of [$TiCl_4$/N_2/H_2O/N_2] = [1/40/1/40 s]. Partial pressure of $TiCl_4$ was 0.01–0.02 Torr and deposition temperature was 100 °C. For each run, the polymer-coated silicon substrate with total surface area of 4 cm^2 was loaded into the ALD reactor. As a control, and to provide independent analysis of the deposited film thickness, ALD layers were simultaneously deposited on clean oxidized silicon wafer pieces.

1.3. Nanosheet separation. In order to facilitate the dissolution of the polymer, the ALD film was cut with a razor blade. Then the substrate with ALD film on the polymer layer was dipped in the solvent with the temperature of 70 °C. The solvents used were toluene for PMMA and DI water for PVA and PAA. After several minutes, the nanosheets began to separate from the substrate and visibly appear in the solvent solution. For further analysis, they were collected on a clean Si/SiO_2 substrate and annealed at 450–500 °C for 1 h in air to remove residual polymer.

1.4. Characterization. ALD film thickness was measured with ellipsometry (Alpha-SE Ellipsometer, JA Woollam Co., Inc.) using a reference Si/SiO_2 substrate. Thickness of nanosheets was measured from atomic force microscopy (AFM, DI 3000) image after annealed at 450 °C for 1 h to remove residual polymers. Optical microscopic images were captured with an Olympus BX60 microscope.