Supporting Information

Tuning the pH Response of Monolayer Hexagonal Boron Nitride/Graphene Field-Effect Transistors

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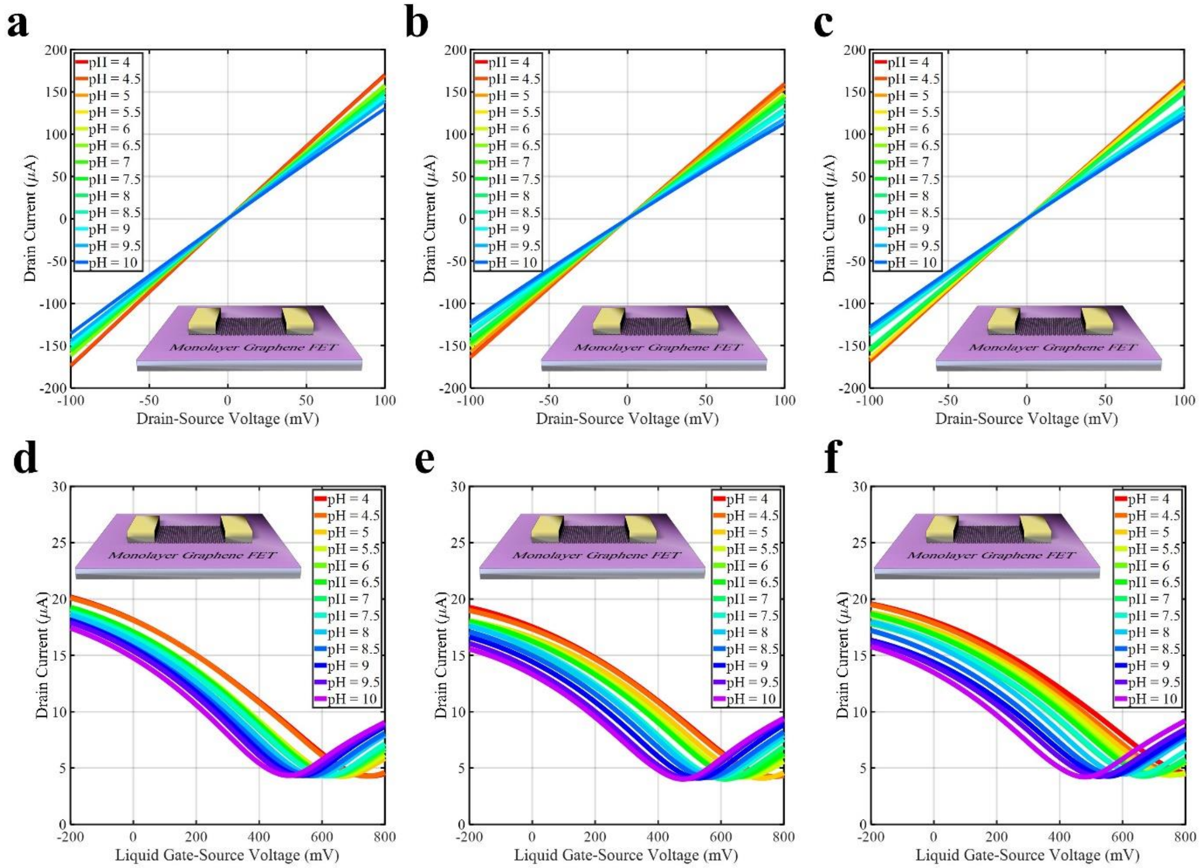

Figure S1. Raw electrical data for monolayer graphene/SiO_2 as a function of 10 mM phosphate solution pH where two-terminal (**a-c**) and liquid-gated three-terminal (**d-f**) measurements were used to extract resistances and Dirac voltages. Insets are computer generated graphics of the monolayer graphene field-effect transistor.

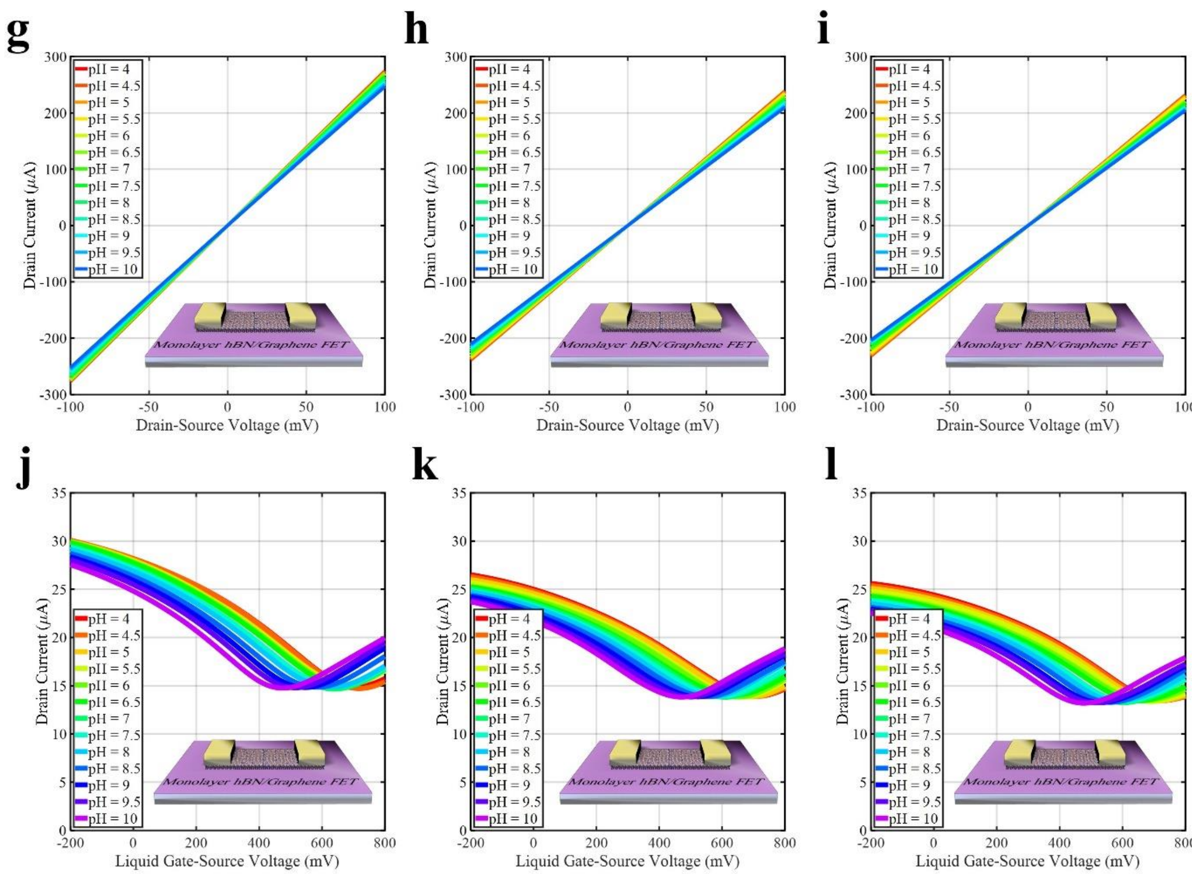

Figure S2. Raw electrical data for hBN/graphene/SiO_2 as a function of 10 mM phosphate solution pH where two-terminal (**a**-**c**) and liquid-gated three-terminal (**d**-**f**) measurements were used to extract resistances and Dirac voltages. Insets are computer generated graphics of the monolayer hBN/graphene field-effect transistor. For the three-terminal measurements, the drain-source bias was 10 mV.

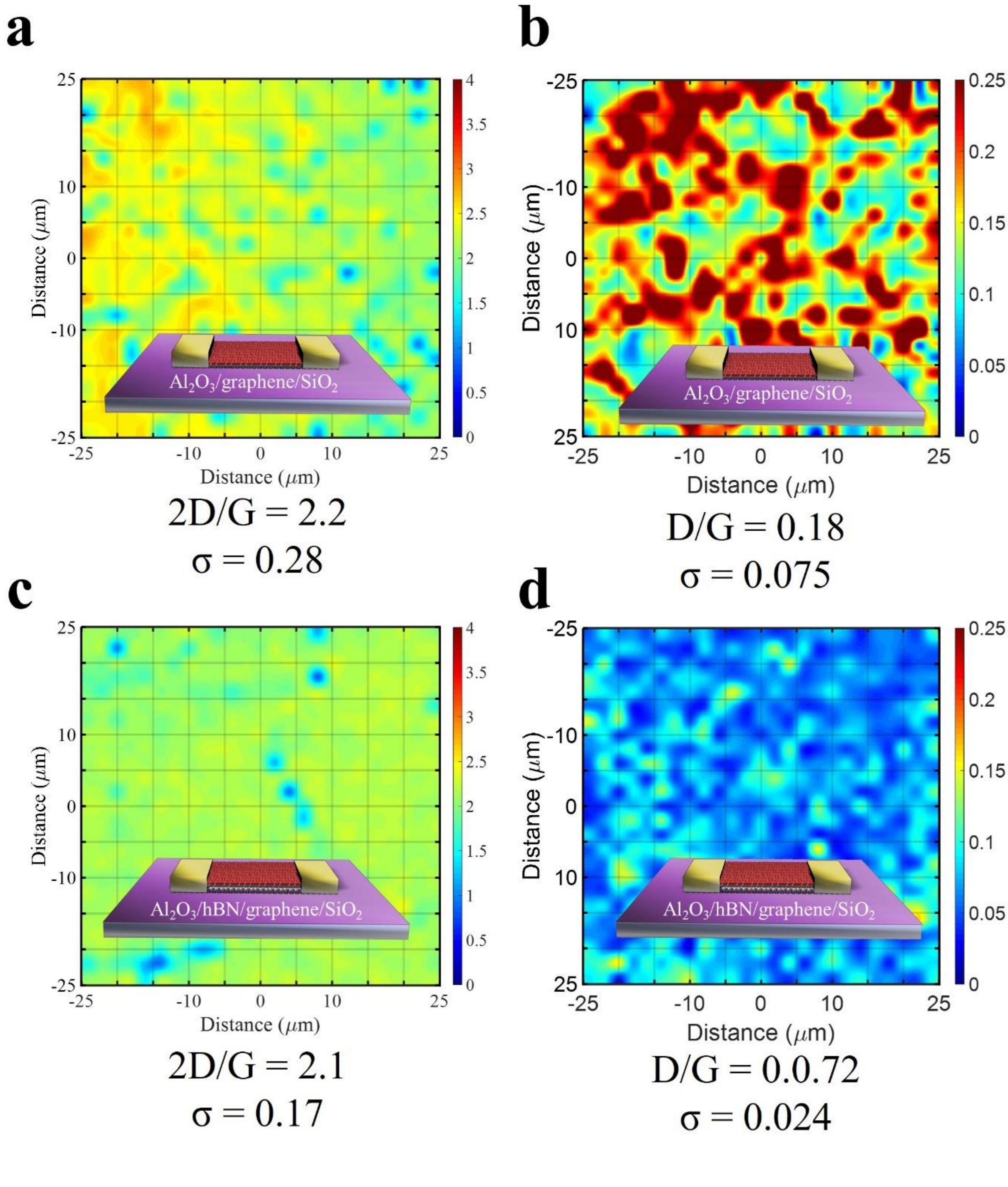

Figure S3. Raman spectral maps were used to extract the 2D/G and D/G phonon intensity ratios of 9 nm of Al_xO_y from electron beam deposition on graphene/SiO_2 (**a**, **b**) and hBN/graphene/SiO_2 (**c**, **d**). Insets are computer generated graphics of an Al_xO_y film on the 2D devices.

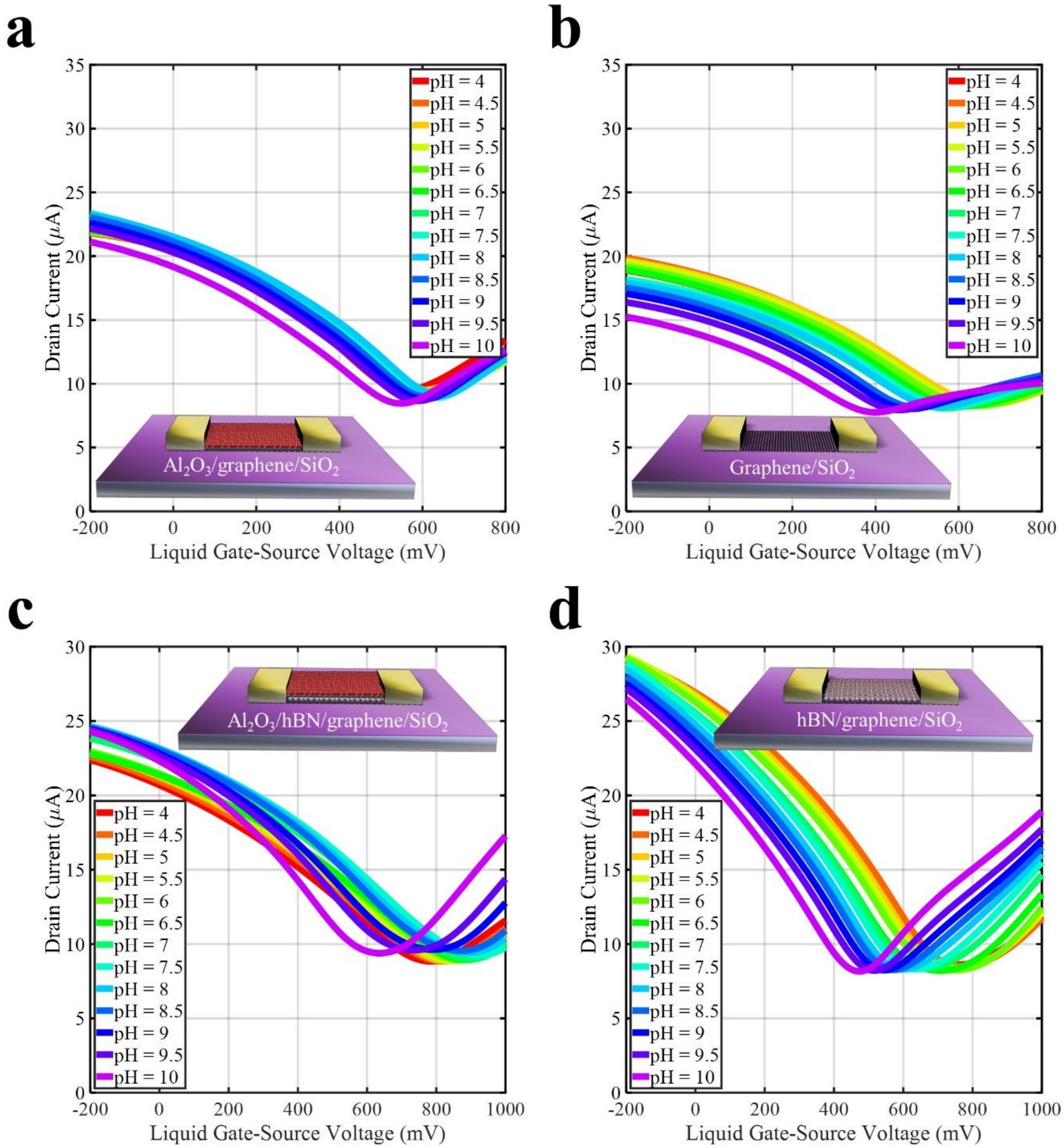

Figure S4. Raw data for liquid-gated transfer characteristics of 9 nm of Al_2O_3 from electron beam deposition before and after basic wet etching on graphene/SiO_2 (**a**, **b**) and hBN/graphene/SiO_2 (**c**, **d**). Insets are computer generated graphics of an Al_xO_y film on the 2D devices before etching, and its removal after etching. Drain-source bias was 10 mV.

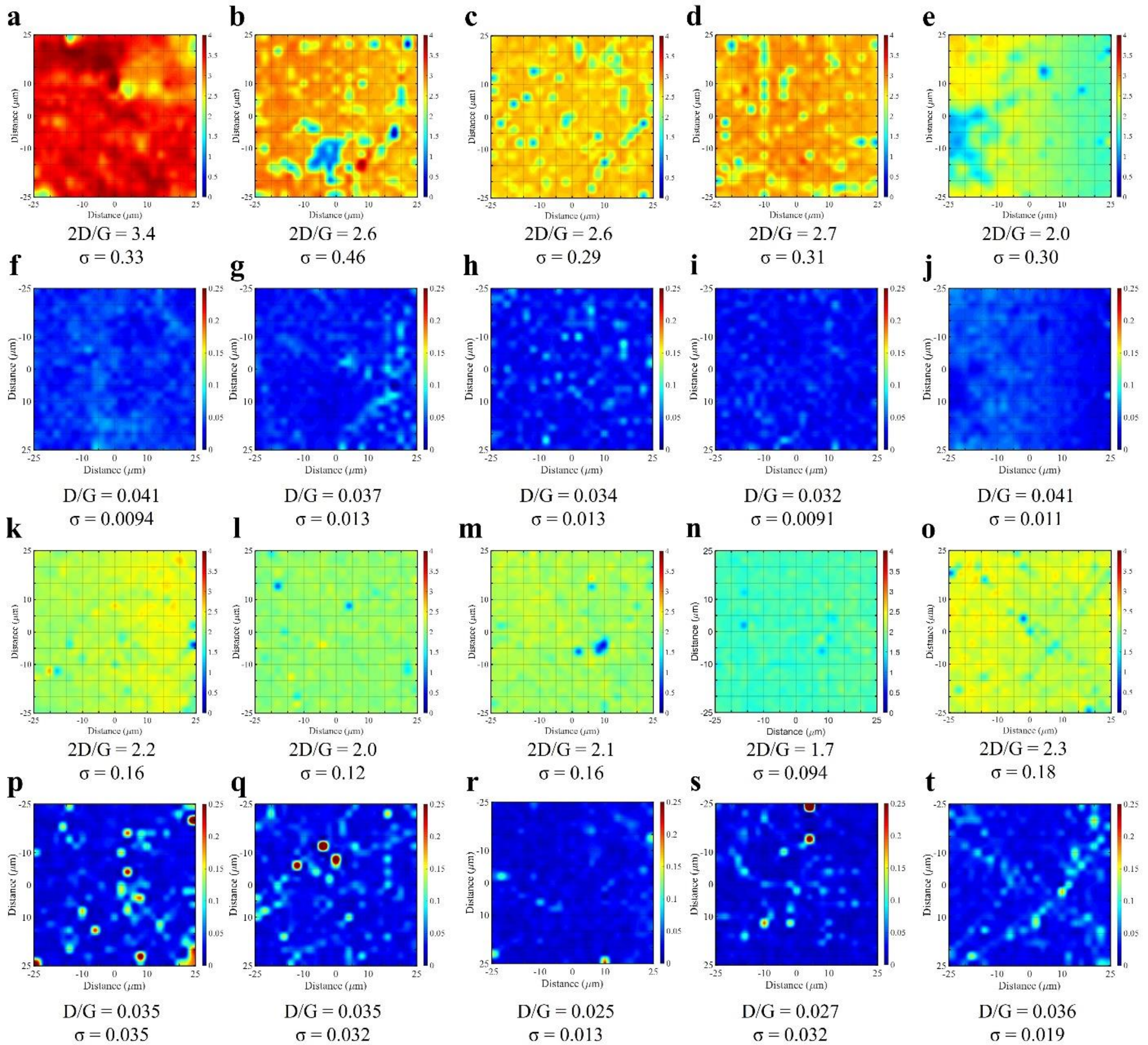

Figure S5. Raman spectral maps were used to extract the 2D/G and D/G phonon intensity ratios as a function of atomic layer deposition thickness of Al_xO_y on graphene/SiO_2 (**a-o**) and hBN/graphene/SiO_2 (**k-t**). The thicknesses of Al_xO_y used here, from left to right, are: 0, 4.3, 8.6, 12.3, and 23.0 nm. The 2D/G and D/G ratios and their standard deviations for 625 measurements over the sample surface are displayed below the Raman spectral maps.

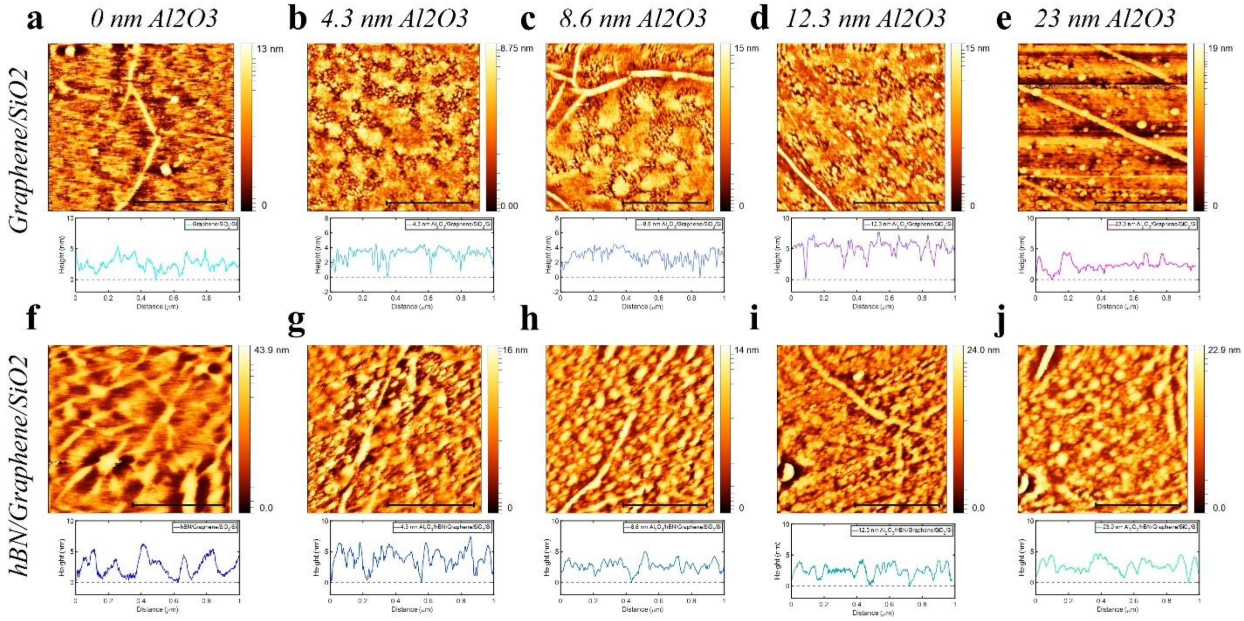

Figure S6. Atomic force microscopy as a function of the thickness of atomic layer deposition of Al_xO_y on graphene/SiO_2 (**a-e**) and hBN/graphene/SiO_2 (**f-j**). One-dimensional profiles are extracted and plotted directly below the AFM topography.

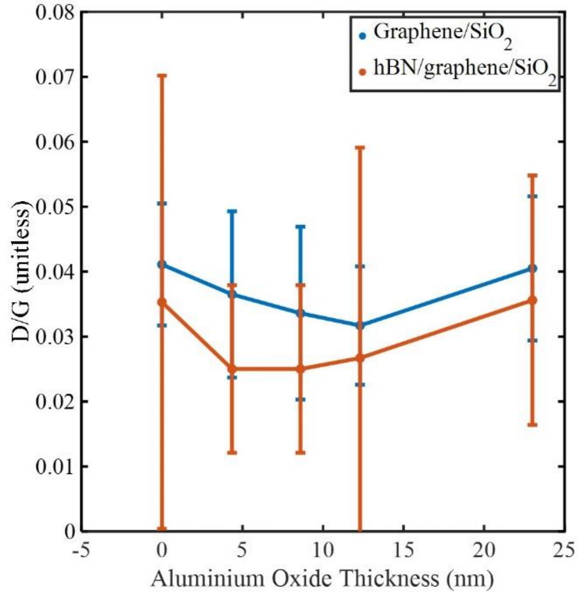

Figure S7. Average D/G ratios from the Raman spectral mapping of atomic layer deposition of Al_xO_y on either graphene/SiO_2 (blue) or hBN/graphene/SiO_2 (red). The standard deviations of the D/G ratios were used as error bars.

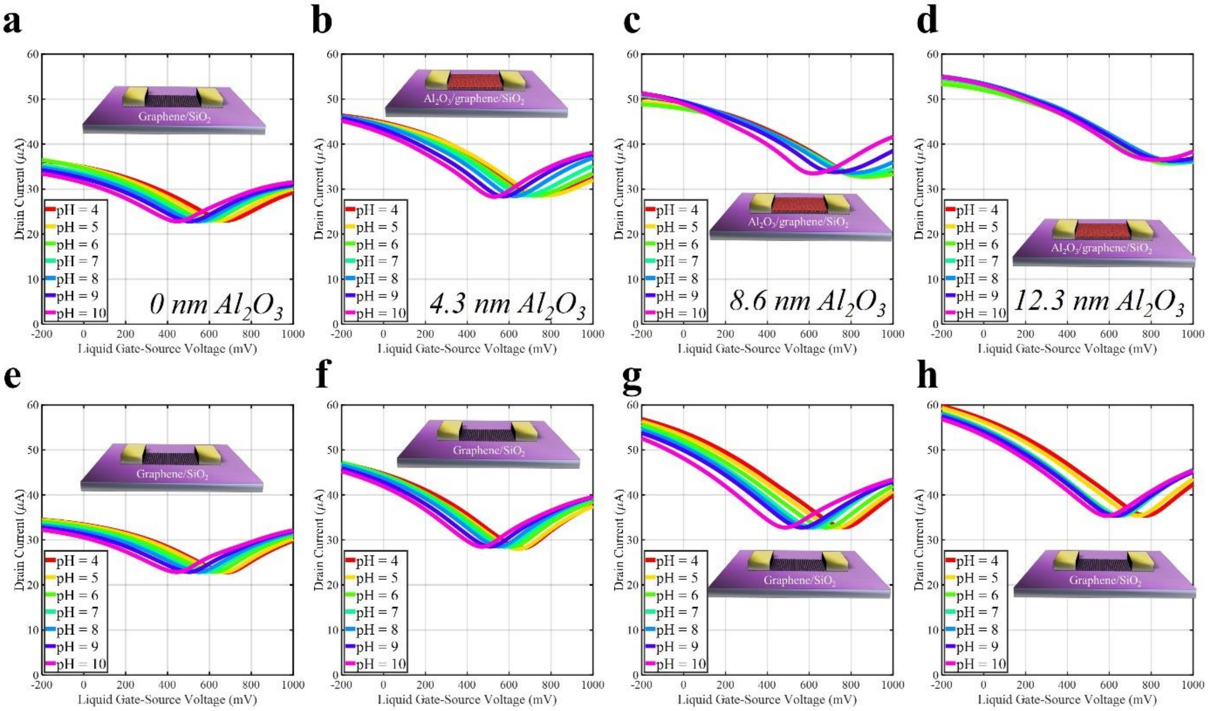

Figure S8. Raw data for liquid-gated transfer characteristics of atomic layer deposition of Al_xO_y on graphene/SiO_2. (**a-d**) are the devices three-terminal response before basic wet etching, whereas (**e-h**) are the devices after basic wet etching. Insets are computer generated graphics of the device state: with or without the Al_xO_y nanofilm. Drain-source bias was 10 mV.

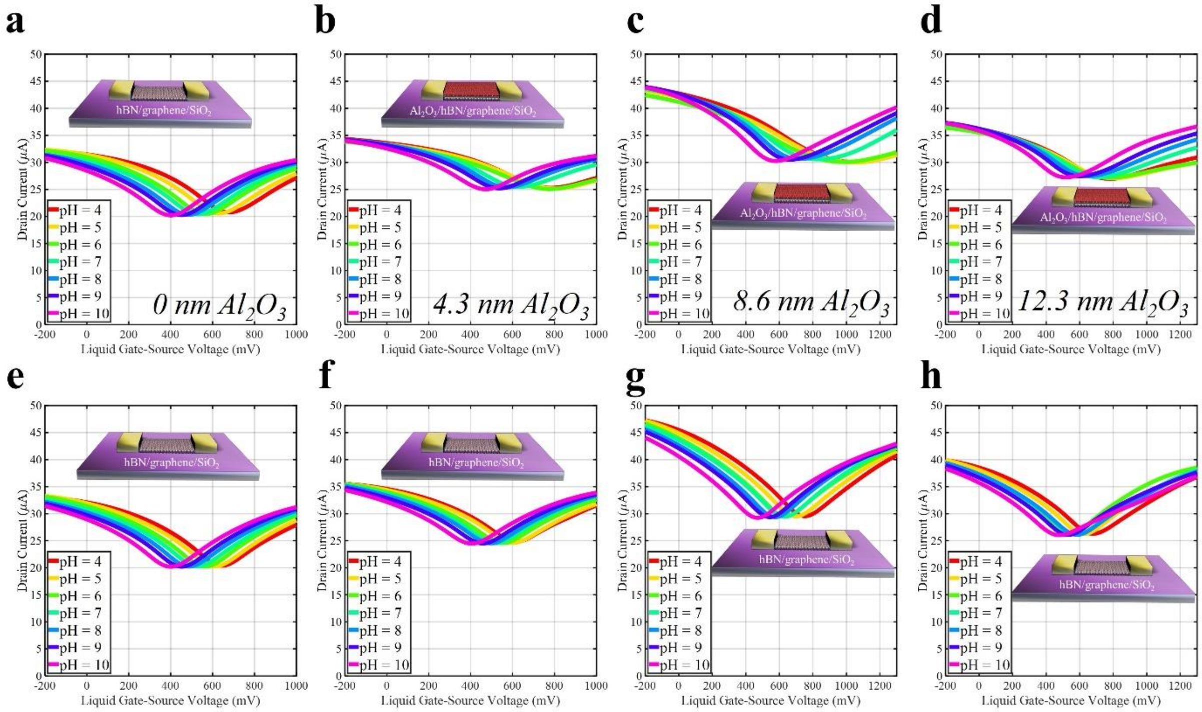

Figure S9. Raw data for liquid-gated transfer characteristics of atomic layer deposition of Al_xO_y on hBN/graphene/SiO_2. (**a-d**) are the devices three-terminal response before basic wet etching, whereas (**e-h**) are the devices after basic wet etching. Insets are computer generated graphics of the device state: with or without the Al_xO_y nanofilm. Drain-source bias was 10 mV.

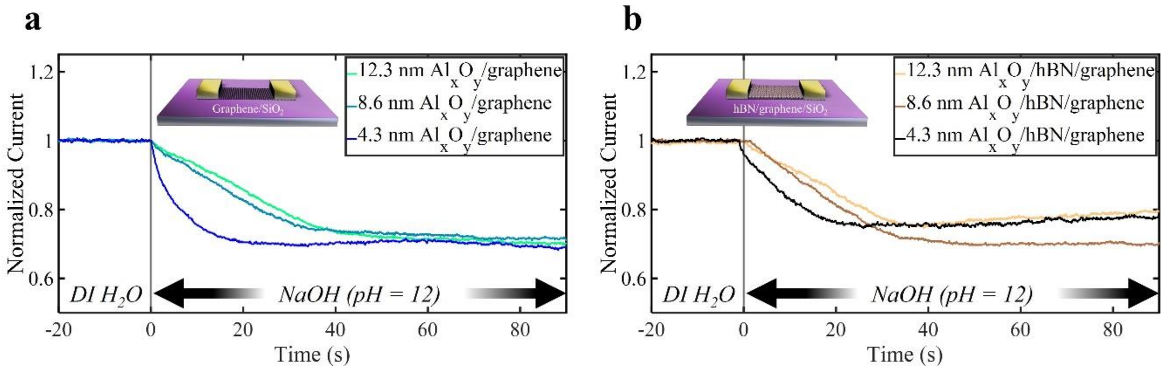

Figure S10. Chronoamperometry of various thicknesses of ALD Al_xO_y on separate (**a**) graphene/SiO_2 and (**b**) monolayer hBN/graphene/SiO_2 field effect transistors. Drain-source bias was 10 mV and current is normalized by the current at 0 seconds. Time less than 0 seconds are the devices operating under 25 μL of deionized water. At 0 seconds, 50 μL of aqueous NaOH (pH = 12) was added. The ALD thicknesses of Al_xO_y were: 4.3 nm, 8.6 nm, and 12.3 nm as measured by ellipsometry.