

Supporting Information

Superior characteristics of graphene field effect transistor enclosed by chemical-vapor-deposition-grown hexagonal boron nitride

Muhammad Waqas Iqbal¹, Muhammd Zahir Iqbal¹, Xiaozhan Jin², Jonghwa Eom^{1*} and
Chanyong Hwang^{2*}

¹Department of Physics and Graphene Research Institute, Sejong University, Seoul 143-747, Korea

*²Center for Nanometrology, Korea Research Institute of Standards and Science, Daejeon 305-
340, Korea*

*e-mail: eom@sejong.ac.kr, cyhwang@kriss.re.kr

Figure S1 is a schematic diagram of the simplified transfer method of the graphene/h-BN film. First, the polymethyl methacrylate (PMMA) was spin-coated on the CVD-grown graphene film on Cu foil, after which the foil was etched out by soaking in ammonium persulfate (APS) solution for 24 h. The graphene film on PMMA was transferred onto as-grown h-BN film on a Cu foil, and then the Cu foil was etched out by APS solution. Finally, the h-BN/graphene/PMMA film was transferred onto a Si/SiO₂ substrate. After the PMMA film was removed by soaking in acetone, the CVD-grown graphene on CVD-grown h-BN film was obtained on the Si substrate with a 300-nm-thick SiO₂ capping layer.

We have measured the electronic transport properties of graphene field effect transistor by sweeping the back gate voltage. The results are shown in Figure S2. Figure S2a shows typical hysteresis curve observed in pristine CVD-grown graphene on SiO₂ substrate, where the back gate voltage was swept continuously from -60 to +60 V and from +60 V to -60 V. We note that charge neutrality point (CNP) moves towards negative (positive) back gate voltage (V_{bg}) for the gate voltage sweep from -60 V (+60 V) to +60 V (-60 V). The hysteresis in CNP is 10 V for graphene on SiO₂ substrate, which is due to charge impurities in SiO₂ substrate. Similar transport measurement has been done for graphene on CVD-grown h-BN and graphene sandwiched between CVD-grown h-BN (h-BN/graphene/h-BN) as shown in Figures S2b and c. The hysteresis in CNP for graphene on CVD h-BN is around 3 V, whereas almost no hysteresis has been observed for graphene sandwiched between CVD h-BN. The hysteresis indicates that a number of charge impurities exist at SiO₂ substrate, whereas very few charge impurities are present at CVD-grown h-BN.

The effect of deep ultraviolet light (DUV) illumination in air on graphene has been examined without flowing O₂ gas. The Dirac point shift for graphene on different substrates after DUV exposure to air is shown in Figure S3. The Dirac point was observed at 18 V for

graphene on SiO₂, whereas the Dirac point of graphene on h-BN substrate was observed to have moved to 7 V after 30 min of DUV illumination. The slight doping effect after DUV illumination for graphene on h-BN was attributed to the gas molecules on the top side.¹ As air contains O₂ molecules at 21% volume ratio, a doping effect by O₂ molecules is still expected. DUV provides enough energy for oxygen ions to adsorb onto the surface of graphene.² The p-type doping effect in graphene on Si/SiO₂ substrates originates from the interaction of graphene with O₂ bound to the silicon SiO₂, and is more facilitated in the presence of O₂ gas during DUV illumination.^{3,4} As the h-BN surface is free from O₂ molecules, the doping effect by DUV illumination is less effective for graphene on h-BN, as shown in Figure S3b. The inert, low density of charged impurities and flat nature of the h-BN substrate offer great advantages for the control of the doping level in graphene up to a desired level while preserving the high mobility.

We have also examined the Raman spectra to identify a doping effect in graphene resulting from O₂ gas flow during DUV exposure on different substrates. The Raman spectra of pristine CVD-grown graphene on SiO₂ and h-BN and of CVD-grown graphene exposed to DUV in O₂ gas flow are shown in Figure S4. The shift in the G and 2D peaks after 30 min of DUV illumination in O₂ gas flow was stronger in the SiO₂-supported sample than in h-BN. A blue-shift in the G- and 2D-peak positions was recorded after DUV illumination in O₂ gas flow for 30 min on both substrates. It is already known that the blue-shift in G- and 2D-peak positions indicates p-type doping of graphene. The p-type doping effect was also confirmed by the decrease in the FWHM of the G peak in graphene.⁵⁻⁸ The shift in the G- and 2D-peak positions for graphene on h-BN after 30 min of DUV illumination in O₂ gas flow is shown in Figure S4b. This kind of doping in graphene on h-BN by using gaseous molecules has the advantage of maintaining the doping effect at the desired level without disturbing the original

honeycomb structure of graphene, while preserving the high mobility.

We have used atomic force microscopy (AFM) to confirm the thickness and morphology of CVD-grown h-BN sample. Figure S5a represents surface topology of CVD-grown h-BN by using AFM. All scans were taken in tapping mode under ambient conditions, where scan area was $20 \times 20 \mu\text{m}^2$. The line profile of CVD-grown h-BN sample is shown in Figure S5b. The thickness measured by AFM is 4.3 nm, which indicates 6 layers of CVD-grown h-BN.

We have also used AFM to confirm the surface morphology of CVD-grown graphene on CVD-grown h-BN sample before and after DUV+O₂ treatment. Figure S6 describes AFM images of CVD-grown graphene on CVD-grown h-BN. Scan was taken in tapping mode under ambient conditions, where scan area was kept $20 \times 20 \mu\text{m}^2$. AFM images were taken almost same position before and after DUV+O₂ treatment. AFM images do not show much difference in surface morphology after DUV+O₂ treatment. The purpose of DUV+O₂ treatment was p-type doping of graphene rather than cleaning the surface.

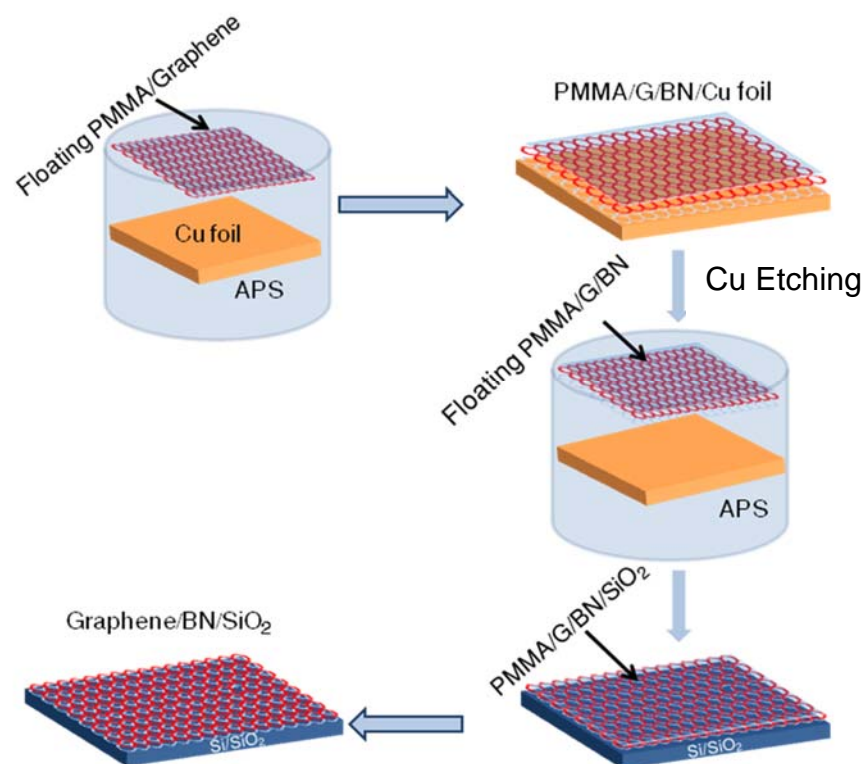


Figure S1. Schematic diagram for the simplified transfer method of graphene/h-BN film.

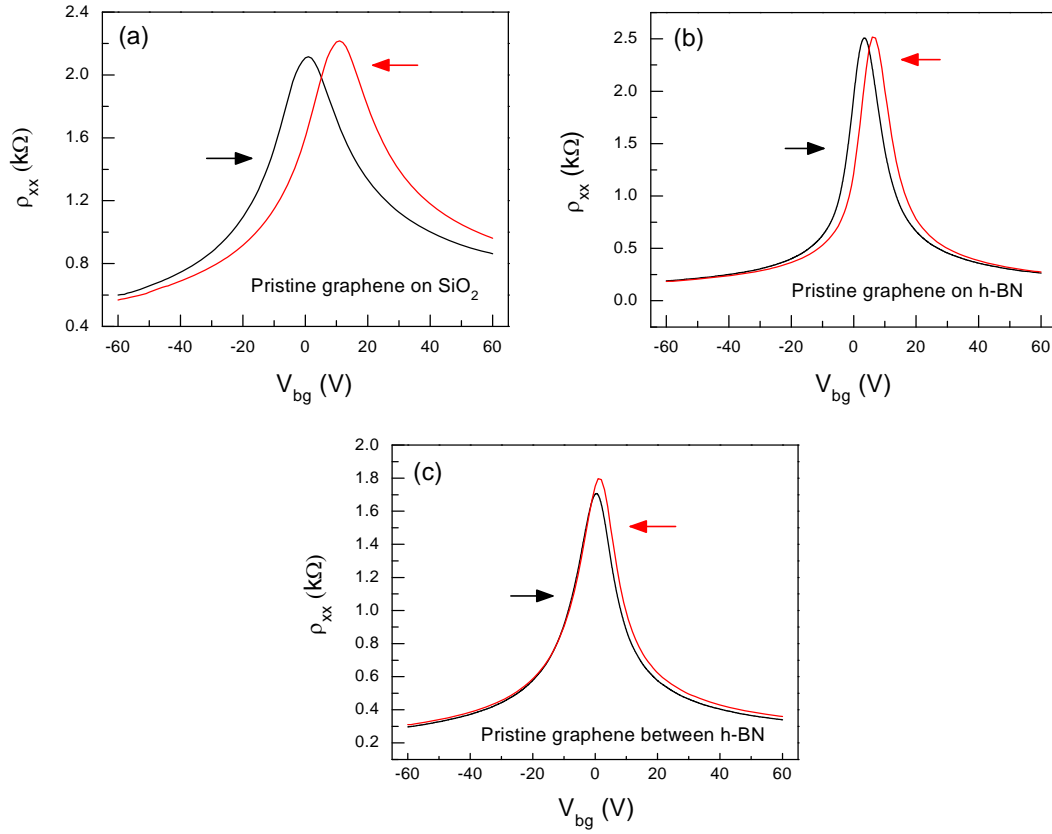


Figure S2. (a) Transport measurement of pristine CVD-grown graphene on SiO_2 substrate, where the back gate voltage was swept continuously from -60 to $+60$ V and from $+60$ V to -60 V. (b) Transport measurement of pristine CVD-grown graphene on CVD h-BN substrate. (c) Transport measurement of pristine CVD-grown graphene sandwiched between CVD h-BN substrate. The measurement was performed in vacuum at room temperature.

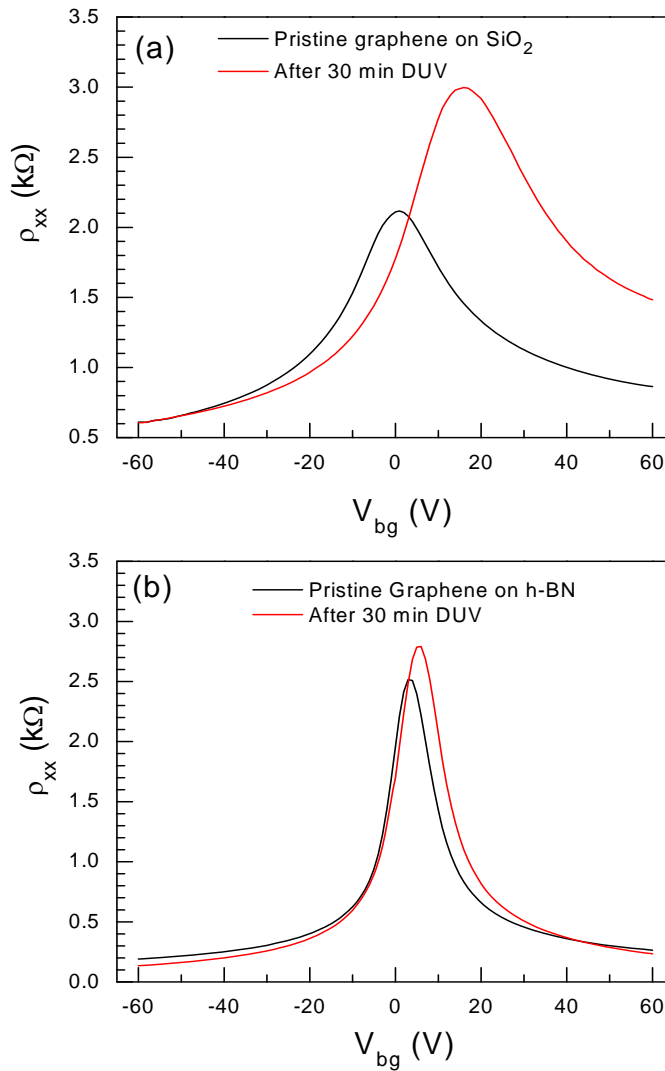


Figure S3. Gate-voltage-dependent resistivity of the graphene devices (a) on SiO_2 after 30 min of DUV exposure in air without oxygen gas flow, where the Dirac point was observed at +18 V on the SiO_2 substrate, and (b) on h-BN after 30 min of DUV exposure in air, where the Dirac point was observed at +7 V on the h-BN substrate. A slight doping effect was observed after DUV illumination for the graphene layer supported on h-BN as a result of the gas molecules on the top.

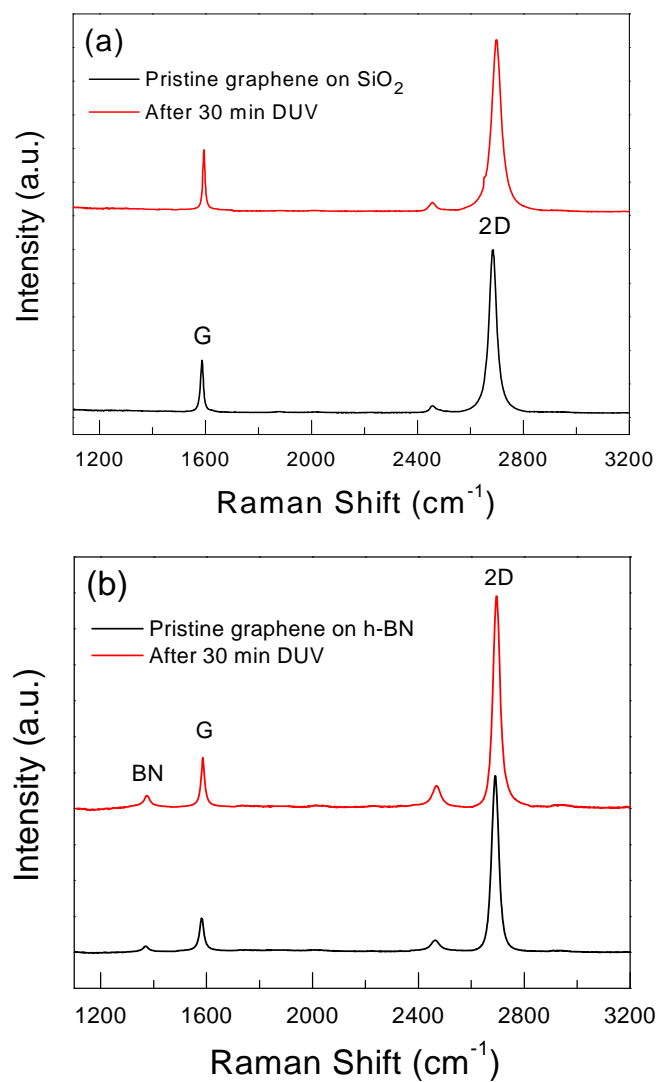


Figure S4. Raman spectra of graphene devices after 30 min of DUV illumination in O₂ gas flow (a) on SiO₂, where a blue-shift was observed in the G- and 2D-peak positions under these conditions, and (b) on h-BN.

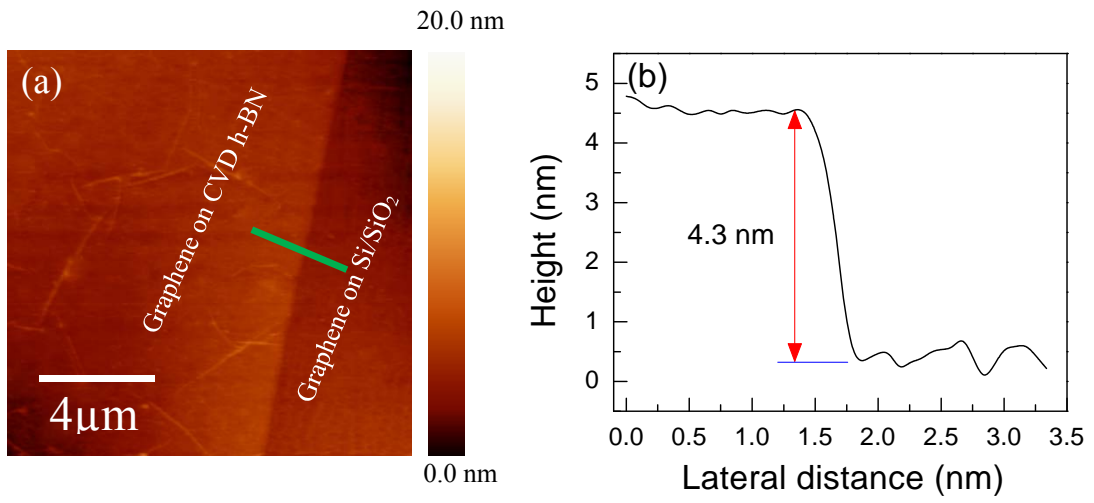


Figure S5. (a) AFM image of CVD-grown graphene on CVD-grown h-BN. (b) Line profile of CVD-grown graphene on CVD-grown h-BN. The thickness of CVD h-BN is 4.3 nm which indicates 6 layers of CVD-grown h-BN.

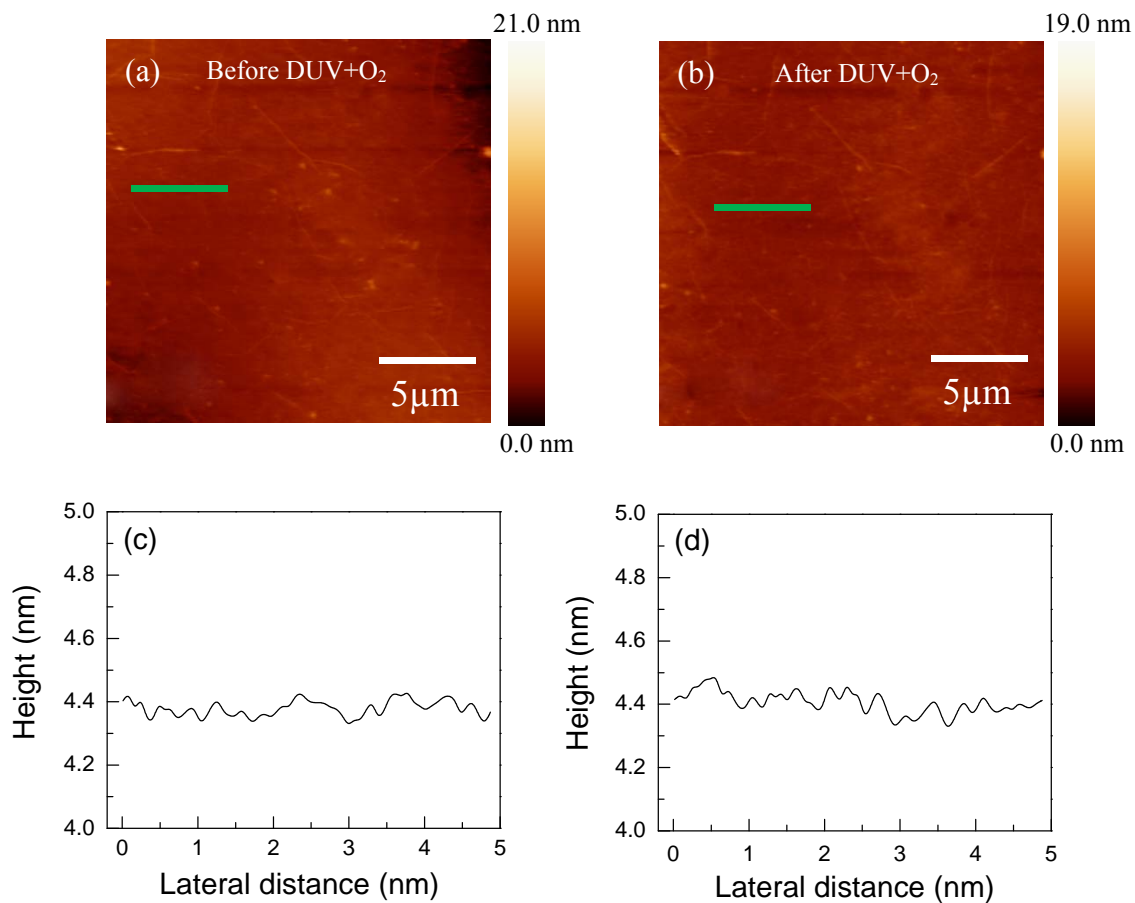


Figure S6. (a) AFM image CVD-grown graphene on CVD-grown h-BN sample before DUV+O₂ treatment. (b) AFM image CVD-grown graphene on CVD-grown h-BN sample after DUV+O₂ treatment. (c) Line profile of CVD-grown graphene on CVD-grown h-BN before DUV+O₂. (d) Line profile of CVD-grown graphene on CVD-grown h-BN after DUV+O₂.

References

1. L. Wang, Z. Chen, C. R. Dean, T. Taniguchi, K. Watanabe, L. E. Brus and J. Hone, *ACS Nano*, 2012, 6, 9314-9319.
2. M. Z. Iqbal, S. Siddique, M. W. Iqbal and J. Eom, *J Mater Chem C*, 2013, 1, 3078-3083.
3. L. Yu, X. L. Pan, X. M. Cao, P. Hu and X. H. Bao, *J Catal*, 2011, 282, 183-190.
4. M. Batzill, *Surf Sci Rep*, 2012, 67, 83-115.
5. M. W. Iqbal, A. K. Singh, M. Z. Iqbal and J. Eom, *J Phys-Condens Mat*, 2012, 24, 335301.
6. J. Yan, Y. B. Zhang, P. Kim and A. Pinczuk, *Phys Rev Lett*, 2007, 98, 166802.
7. A. Das, S. Pisana, B. Chakraborty, S. Piscanec, S. K. Saha, U. V. Waghmare, K. S. Novoselov, H. R. Krishnamurthy, A. K. Geim, A. C. Ferrari and A. K. Sood, *Nat Nanotechnol*, 2008, 3, 210-215.
8. M. Kalbac, A. Reina-Cecco, H. Farhat, J. Kong, L. Kavan and M. S. Dresselhaus, *ACS Nano*, 2010, 4, 6055-6063.