Supporting Information

Electrolyte-gated graphene field-effect transistors for detecting pH and protein adsorption

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1. Confirmation of single-layer graphene

Many single- and multi-layer graphene films can be observed on a 285 nm SiO_2/Si substrate created from natural graphite by mechanical exfoliation, as shown in Figure S1. The number of graphene layers can be confirmed by Raman spectroscopy. In Raman spectroscopy, a He-Ne laser at 632.8 nm was used for the excitation light, and was focused on the sample surface with a diameter of about 1 μm by a microscope objective lens.

Figure S2 compares the 632.8 nm Raman spectra of 1 and 2 layer(s) graphene and highly oriented pyrolytic graphite (HOPG). Two strong peaks with a G band at ~1580 cm^{-1} and a 2D band at ~2650 cm^{-1} could be observed. A single peak can be observed for the single-layer graphene, while a much broader peak, shifted to a high-frequency, could be observed for the bilayer graphene. As shown in Figure S2, a single peak in the 2D band in the Raman spectrum is direct evidence of single-layer graphene.[1] In this work, we used only single-layer graphene for the devices.

2. Ionic conductivity

Figure S4 shows leak current as a function of top-gate voltage at pH 4.0, 5.8 and 7.8. The leak current slightly increased with increasing the top-gate voltage. The absolute value of the leak current was less than 10 nA for all measurement in this work, which was 0.001 times smaller than the drain current. And this characteristics is almost independent on the pH. Therefore, it can be considered that the leak current was independent for the drain current change by increasing pH and BSA adsorption in this measurement conditions.

3. pH dependence of the carrier mobility

Figure S4 shows the electron and hole field-effect mobility [$\mu = (1/C_g)(\partial\sigma/\partial V_g)$] plotted as a function of pH value. The electron [hole] mobility slightly decreasing [increasing] with increasing the pH value. Resent research showed that the mobility was not changed by NO_2 exposure [2] and changing pH [3] (for epitaxial graphene), and the mobility was drastically changed after potassium deposition [4] and NaF doping.[5] These results indicate that increased solution pH have some effects on scattering to carriers on single-layer graphene obtained by mechanical exfoliation of the natural graphite.

4. Hole doping by BSA adsorption

Figure S5 shows G-V_{TGS} characteristics before and after BSA adsorption. The field-effect mobility was estimated to be 4,400 and 3,700 cm^2/Vs for before and after BSA adsorption, respectively. As a result, the hole concentration ($p = \sigma/e\mu$) was increased from 7.8×10^{11} to 9.5×10^{11} cm^{-2} by the 300-nM BSA adsorption.

Reference

[1] Ferrari, A. C.; Meyer, J. C.; Scardaci, V.; Casiraghi, C.; Lazzeri, M.; Mauri, F.; Piscanee, S.; Jiang, D.; Novoselov, K. S.; Roth, S.; Geim. A. K. *Phys. Rev. Lett.* **2006**, 97, 187401.

[2] Schedin, F.; Geim, A. K.; Morozov, S. V.; Hill, E. W.; Blake, P.; Katsnelson, M. I.; Novoselov, K. S. *Nat. Mater.* **2007**, 6, 652.

[3] Ang, P. K.; Chen, W.; Wee, A. T. S.; Loh, K. P. *J. Am. Chem. Soc.* **2008**, 130, 14392.

[4] Chen, J. H.; Jang, C.; Adam, S.; Fuhrer, M. S.; Williams, E. D.; Ishigami, M. *Nat. Phys.* **2008**, 4, 377.

[5] Chen, F.; Xia, J.; Tao, N. *Nano Lett.* **2009**, 9, 1621.

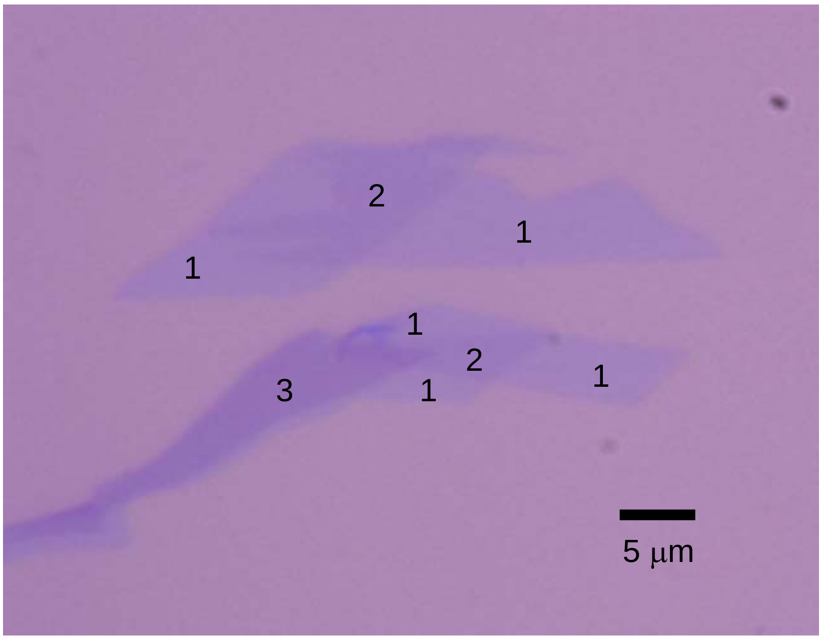

Figure S1: Optical micrograph of the single- and multi-layer graphene on a 285 nm SiO_2/Si substrate.

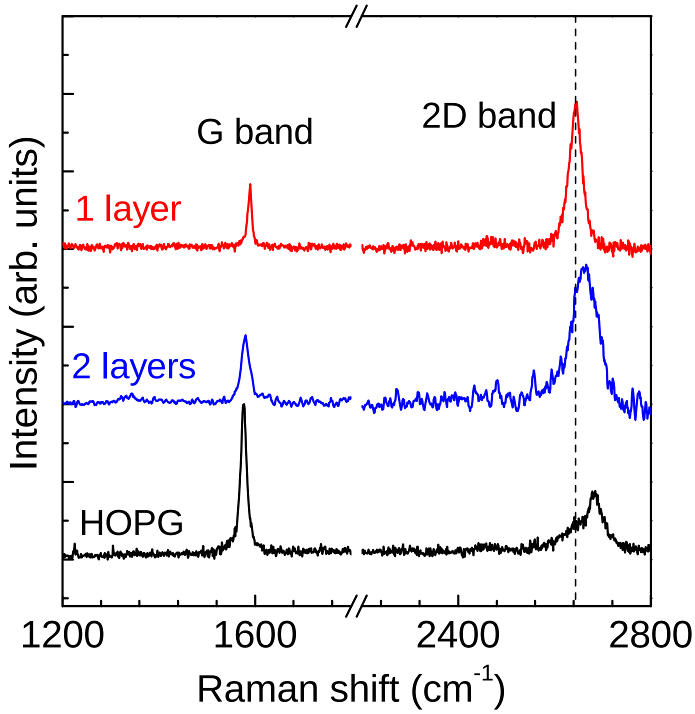

Figure S2: Raman spectra of G- and 2D-bands for 1 and 2 layer(s) graphene and HOPG.

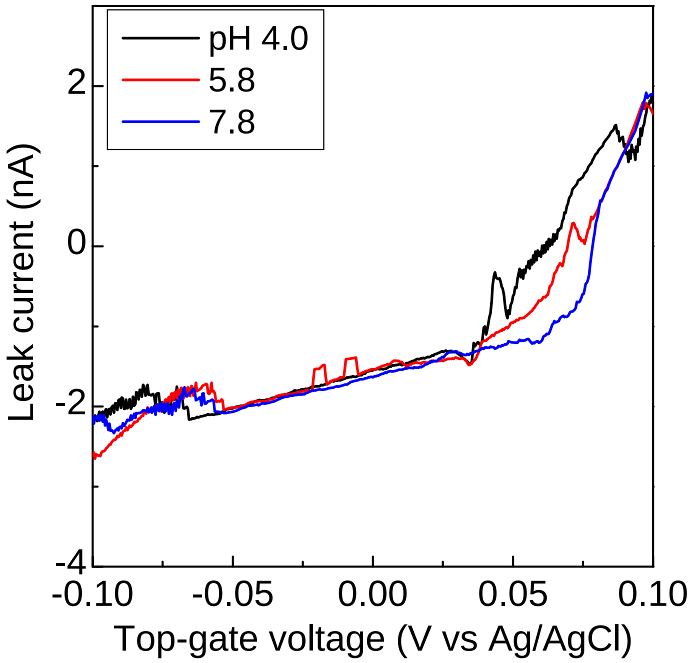

Figure S3: Leak current plotted as a function of the top-gate voltage.

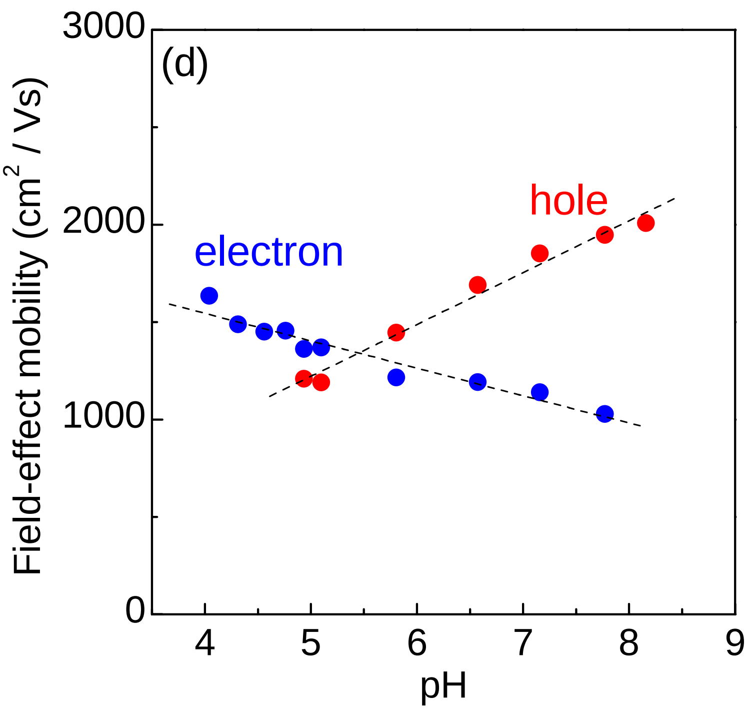

Figure S4: Field-effect mobility of electron and hole as a function of solution pH. Dashed lines indicate the linear fit to the data points.

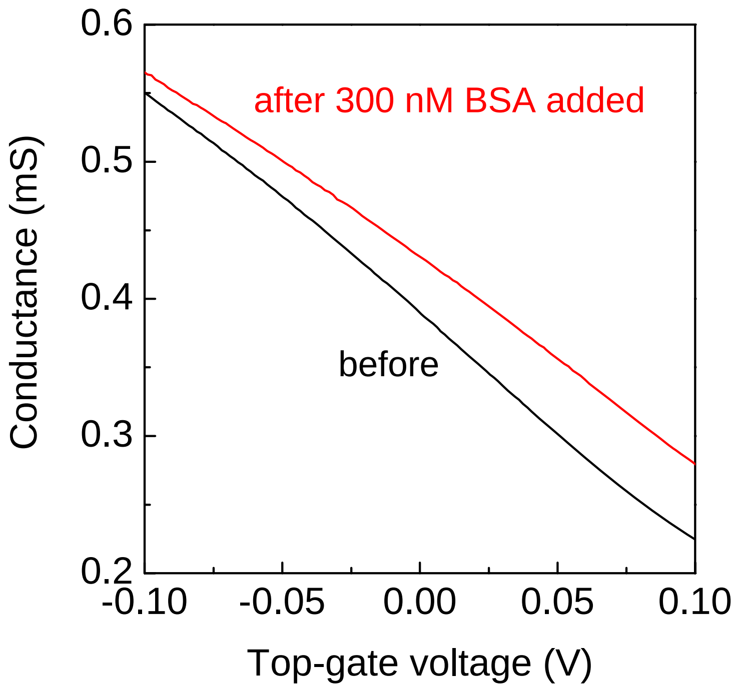

Figure S5: G-V_{TGS} characteristics before and after BSA adsorption.