

Electronic Supplementary Material

Polyelectrolyte multilayer electrostatic gating of graphene field-effect transistors

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Supplementary Material 1: SEM images of graphene, PAH/graphene and PSS/graphene

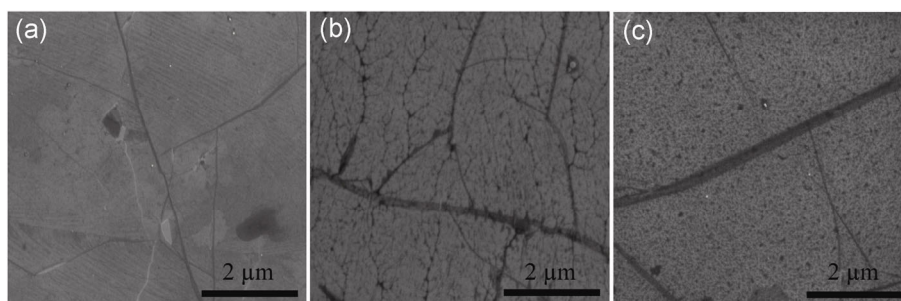


Figure S1 (a) SEM image of the graphene surface. (b) SEM image of graphene coated by PAH. (c) SEM image of graphene coated by PSS.

Supplementary Material 2: Raman spectra of graphene, PAH/graphene and PSS/graphene

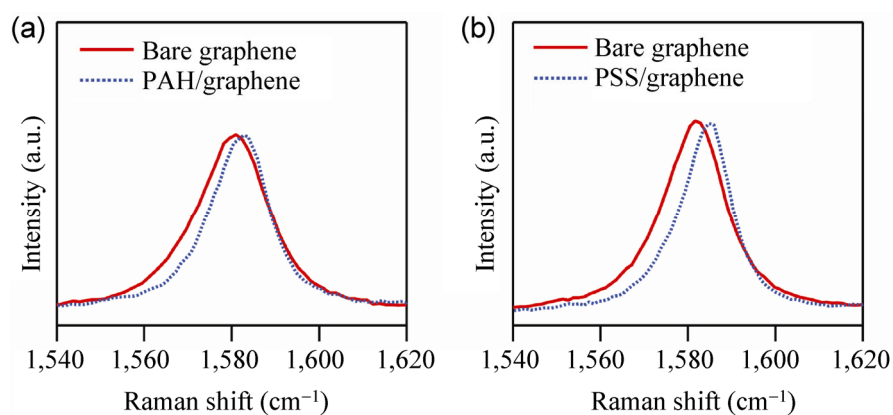


Figure S2 Raman spectra of G peak position and FWHM values for single layer graphene before and after coating polyelectrolyte film (a) PAH and (b) PSS.

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To show that PAH and PSS on graphene surface act as positive and negative potential gating, Raman spectroscopy was employed to monitor the changes of G-band and its full width at half maximum (FWHM) value before and after deposition of the PEM. We first measured the Raman spectrum of clean graphene that was transfer printed onto SiO₂ by PDMS. Then the polyelectrolyte films PAH or PSS were deposited on the graphene surface. Figures S2(a) and S2(b) present the Raman spectra of the G band before and after adsorbing the polyelectrolyte film PAH or PSS. After PAH coating, the average position of G band has a blue shift about 3.1 cm⁻¹ while the FWHM value of G band decreases by 3.2 cm⁻¹. This blue shift and FWHM reduction are caused by the positively charged PAH and due to stiffening of G band for non-adiabatic Kohn-anomaly [S1, S2]. For the PSS coating, the G band is shifted to right about 4.2 cm⁻¹ while the FWHM value reduces by 3.4 cm⁻¹. These changes are generated by the negatively charged PSS film due to stiffening of the G band according to the adiabatic Born-Oppenheimer approximation [S1]. The observations of the blue shift of the G band and the reduction of FWHM values for absorbing positively charged PAH and negatively charged PSS on graphene surface are both in agreement with previously obtained results for electrostatic gating [S2–S4].

Supplementary Material 3: FTIR spectra of graphene, PAH/graphene and PSS/graphene

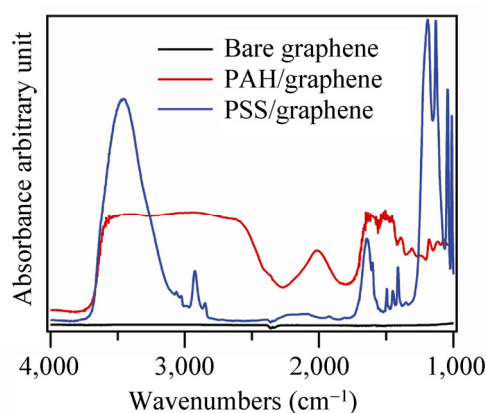


Figure S3 FTIR spectra of bare graphene, PAH/graphene and PSS/graphene films on calcium fluoride substrates.

In order to confirm the expected composition of the polyelectrolyte multilayer, FTIR was employed on samples made for this project. Figure S3 shows the FTIR spectra of bare graphene, PAH/graphene and PSS/graphene films on calcium fluoride substrates. The bare graphene shows no adsorption in the FTIR spectrum. For the PAH/graphene film, the band around 3,330 cm⁻¹ is attributed to the NH₃⁺ group. In the PSS/graphene film, the band around 1,190 cm⁻¹ is due to the SO₃⁻ group. The FTIR spectra confirm the presence of PAH and PSS polyelectrolyte films on the graphene surface, as expected.

Supplementary Material 4: High magnification view of Figs. 4(a) and 4(b)

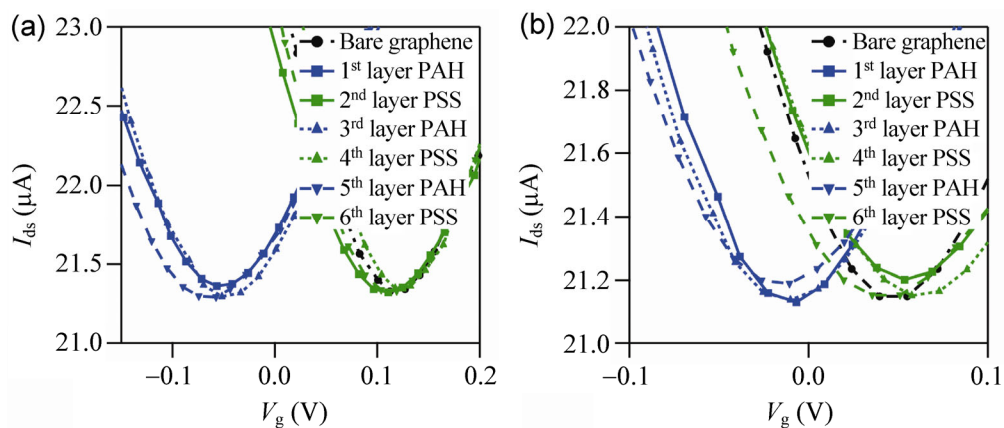


Figure S4 Transfer characteristics of graphene FET devices as a function of PAH/PSS multilayers measured in (a) 1 mM NaCl and (b) 100 mM NaCl at $V_{ds} = 0.1$ V. Transfer characteristics of the device before (black dashed and dotted line) and after polymer coating with PAH (blue lines) and PSS (green lines). 1st and 2nd layers are solid lines with squares. 3rd and 4th layers are dotted lines with triangles. 5th and 6th layers are dashed lines with reverse triangles.

Supplementary Material 5: Thickness measurement of PAH and PSS

To measure the thickness of PAH and PSS films on the surface of graphene, ellipsometry was employed. The CVD-grown graphene was transferred onto a Si wafer using PMMA transfer. After PMMA was cleaned by acetone, the graphene sample was annealed in H_2/Ar (50%/50%) at 400 °C for 1 h. Then the PAH and PSS films were deposited on the graphene surface by the same process described in the main article. The average thickness is about 1.2 nm for both PAH and PSS films on the surface of graphene.

References

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