Supplementary Information

Broadly Tunable Graphene Plasmons Using an Ion-gel Top Gate with Low Control Voltage

Hai Hu,^a Feng Zhai,^b Debo Hu,^a Zhenjun Li,^a Bing Bai,^a Xiaoxia Yang,^{*a} and Qing Dai^{*a}

a. National Center for Nanoscience and technology, Beijing 100190, China.

b. Department of Physics, Zhejiang Normal University, Jinhua 321004, China.

[†]Corresponding E-mail: <u>daiq@nanoctr.cn</u>, <u>yangxx@nanoctr.cn</u>;



Figure S1. Raman spectrum of a CVD-grown graphene sheet on 300 nm SiO_2 / Si substrate. Inset: Optical micrograph of the graphene sheet.



Figure S2. (a) AFM image of a graphene nanoribbon array with ribbon width of ~65 nm. (b) Linescan profile of the image shown in (a) corresponding to the black and red line, respectively. The graphene nanoribbons have a uniform thickness of about 2 nm.



Figure S3. (a) Resistance as a function of V_{bg} at different fixed V_{tg} . (b) The linear relation between V_{tg} and V_{bg} that results at graphene CNPs. Red circles are experimental data and they can be fitted with the black line.

When graphene channel is simultaneously controlled by the top-gate and back-gate, the shift in the back-gate voltage at CNP ($V_{bg,CNP}$) should by linearly dependent on the change of applied top-gate voltage (V_{tg}). Figure S3 a shows the electrical resistance of graphene as a function of back-gate voltage (V_{bg}) at different fixed V_{tg} . We extract the values of $V_{bg,CNP}$ and plot them as function of related V_{tg} , which exhibits a linear relation, as shown in figure S3b. The slope of the $V_{bg,CNP}$ - V_{tg} line is -0.005, which is determined by the ratio between the capacitances (unit area) of ion-gel dielectric top-gate and SiO₂

dielectric back-gate, that is, $\frac{C_{tg}}{C_{bg}} = -\frac{\Delta V_{bg,CNP}}{\Delta V_{tg}}$. Substituting the SiO₂ capacitance (0.0121)

 μ Fcm⁻²) and the slope into this formula, we can derive the top-gate capacitance as 2.42 μ Fcm⁻². Based on the calculated SiO₂ back-gate and ion-gel top-gate capacitance, we can get:

$$V_{tg} - V_{CNP} = 1.283 * 10^{-7} \sqrt{n} + 7.146 * 10^{-14} n$$
(1)

$$V_{bg} - V_{CNP} = 1.283 * 10^{-7} \sqrt{n} + 1.324 * 10^{-11} n$$
⁽²⁾

$$V_{tg} - V_{CNP} = E_F + 4.05 E_F^{2}$$
(3)

$$V_{bg} - V_{CNP} = E_F + 810 E_F^{2}$$
(4)

Where n is in units of 10^{13} cm⁻² and $E_{\rm F}$ is in units of eV. ε is the average of dielectric environment and for graphene sandwiched between two dielectric slabs with ε_1 and ε_2 , $\varepsilon = 1/2 \times (\varepsilon_1 + \varepsilon_2)$. Therefore, the resonance frequency are still different, while there are fixed wave vector and Fermi level of graphene nanoribbons.



Figure S4. The extinction spectra taken from the same graphene nanoribbon array with (blue line) and without (red line) ion-gel film. The E_F of both cases are controlled to be the same by the same back gate. The measurements are both repeated 5 times and (dotted lines) the solid lines are the averages of the individual measurements.

Figure S4 shows extinction spectra of a same graphene nanoribbons with and without spin-coating a thin film of ion-gel. In both cases, the Fermi levels are fixted to be 0.44 eV by SiO₂ back gate and all measurements are repeated 5 times. We can obviously see that the GP frequency (1452 cm⁻¹) in the blue line (spin-coated with ion-gel) is 274 cm⁻¹ less than that in the red line (without ion-gel film). This difference comes from the plasmonic frequency scales like $\varepsilon^{-1/2}$, and the environmental dielectric constant ε_{eff} of graphene changed. For air/graphene/SiO₂ structure, ε_{eff} is 2.45 while ε_{eff} equal to 4.67 for ion-gel/graphene/SiO₂.



Figure S5. Calculated relation between plasmon resonance frequencies on Fermi levels controlled by SiO_2 back gate. The experimental resonance frequencies of PP and GP peaks extracted from Figure 2b in the main text are also plotted.



Figure S6. (a) Comparison between extinction spectra with the same resonance frequencies taken from the same graphene/SiO₂ device before (red dots) and after (blue dots) coating with an ion-gel thin film. The solid lines are the fitting curves in Fano model to extract the linewidth of resonance peaks. (b) The lifetime of the GP peaks extracted from a same graphene device with and without coating ion-gel dielectric as a function of resonance frequencies.

In figure S6a, we plot two extinction spectra taken from the graphene/SiO₂ device before and after ion-gel coating, which are chosen to have similar resonance frequencies. The linewidth Γ , corresponding to the damping rate of plasmon, of the device with iongel is obviously larger than that without ion-gel. We extracted the linewidth Γ of resonance peaks of all the extinction spectra obtained by using Fano model and then calculated their plasmon lifetime T via $T = 2\hbar/\Gamma$. In order to study the effect of ion-gel, we systematically compare the graphene plasmon lifetimes of graphene/SiO₂ device before (red lines) and after the coating of ion-gel top gate (blue lines), as shown in Figure S6b.