

Supplementary Information for

“Highly Confined Tunable Mid-Infrared Plasmonics in Graphene Nanoresonators”

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1 Approximating the Fermi Level Position

In order to determine the Fermi level position of our devices, we first measured the resistance vs. applied gate voltage dependence of the graphene sheet that contained the nanoresonators, as shown in Fig. S1. From these measurements we were able to determine the charge neutral point (CNP) for each device, which corresponds to applied gate voltage that aligns the Fermi level of the graphene with the Dirac point, leading to a peak in the resistance curve. Once the CNP was known, we used a simple capacitor model in order to approximate the position of E_F for a given gate voltage. For a 285nm SiO_2 layer, this relationship is given by $|E_F| = 0.0319\sqrt{|V_{\text{CNP}} - V_G|}$. For most devices, V_G could be varied from -100V to +200V without causing electric breakdown of the SiO_2 layer.

We found that our as-prepared samples were hole doped, and that the degree of hole doping was dependent on the etchant we used to remove the copper foil that the graphene was grown on. As shown in Fig. S1, when an Ammonium Persulfate (APS) solution (2% by wt.) was used as the etchant, the CNP typically occurred near $V_G = 50\text{V}$. In contrast, when an Iron(III) Chloride(FeCl_3) solution (40% by wt.) was used as the etchant, the CNP occurred at much higher gate biases, typically with V_G near +180V. This intrinsic hole doping allowed us to electrostatically shift the E_F from 0 to -0.52 eV.

The above analysis applies to the bare graphene surface. However, it has been recently observed by Thongrattanasiri, *et al*¹ that the simple capacitance model typically used to estimate the Fermi level position of graphene devices may change when the graphene is patterned in a nanoribbon geometry. In particular, it was predicted by those authors that the Fermi level position can deviate strongly near the nanoribbon edges, and that this deviation can affect the plasmonic

character of the graphene nanoresonators. In order to see if such effects play a role in our experiment, we compare in Fig. 2a the normalized transmission spectra of the bare graphene surface to that of the nanoresonators, with light polarized parallel to the nanoresonators. These data show the characteristic rise and drop in the normalized transmission of doped graphene on SiO₂, with the drop in transmission roughly occurring at $2E_F$, above which interband transitions are no longer Pauli blocked². It can be seen in these data that the drop in transmission of the nanoresonators occurs at similar energies to the bare graphene, and that the spectra evolve in a similar fashion.

Although Figure S2 shows some deviations between the spectra taken on the bare graphene as compared to the nanoresonators, it should be noted that the transmission spectra on the resonators can be influenced by a number of factors other than those mentioned by Ref. [1]. First, the mobility of the graphene in the nanoresonators is likely to be much lower than the mobility of the bare graphene due to the lithographic processing that is used to pattern them, and such a change in mobility can alter the normalized transmission features. Second, charge traps and edge states can be introduced on the nanoresonators as a result of the patterning, and those can lead to a constant change in the background charge density. Such edge effects may also play a role in offsetting or moderating the effects predicted in Ref. [1].

2. Plasmon Lifetime

In Fig. S3 we plot the width, $\Delta\omega_p$, of the transmission peaks for graphene nanoresonators of different size and doping levels. A naïve estimation of the plasmon lifetime from the inverse linewidth yields lifetimes of 5-50 fs based on these data, with the lifetime decreasing as the resonances move to higher frequencies. One explanation of this trend is that the graphene plasmons should be damped as the plasmon energy moves above the in-plane optical phonon energy of at 0.2eV.³ Although this effect may play a role in increasing $\Delta\omega_p$ as the resonant frequencies (ω_p) increase, it is important to note that the peak widths that we experimentally measure are due to multiple effects, many of which are expected to show an energy dependence. First, as the plasmons are confined into smaller graphene nanoresonators (which exhibit higher ω_p for the same carrier density), the edge roughness-to-width ratio of the nanoresonators becomes larger and thus the quality factor of the resonator worsens. Second, there is an

ensemble averaging effect that broadens the measured plasmon resonances due to the $\pm 2\text{nm}$ deviations in the fabricated nanoresonator widths. These small deviations in width will lead to a spread of ω_p values that will be larger for smaller lengthscale nanoresonators, and will also change as a function of doping. Finally, the permittivity of the SiO_2 is dispersive under 0.25eV , which distorts the shape of the resonance peaks and thus leads to an energy-dependent broadening effect on the transmission peaks that is unrelated to the plasmon lifetime. These effects make it difficult to estimate the true energy- and doping-dependent plasmon lifetime based on the experimentally measured $\Delta\omega_p$.

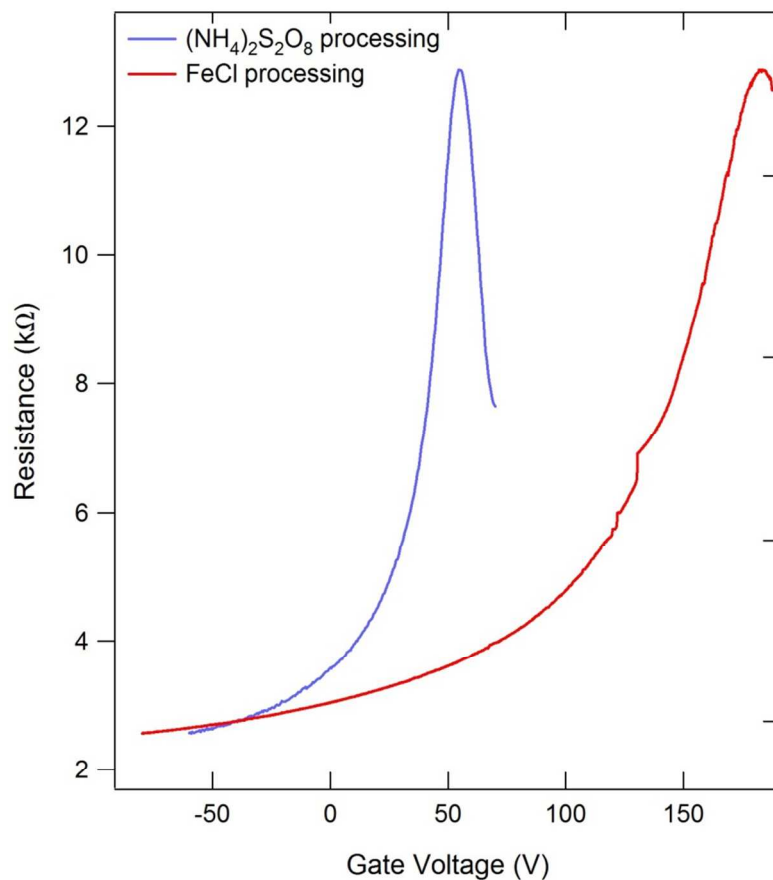


Figure S1. Resistance vs. Gate Voltage curves of graphene field effect transistor devices containing nanoresonators. The red line corresponds to a device that was prepared using Iron(III) Chloride (40% by wt.) to etch away the copper foil, while the blue line indicates a device that was prepared using Ammonium Persulfate (2% by wt.) as the etchant. Source and drain electrodes are typically separated by 0.5 – 5.0 mm, and are placed on the bare graphene, at least 200um from the nanoresonators

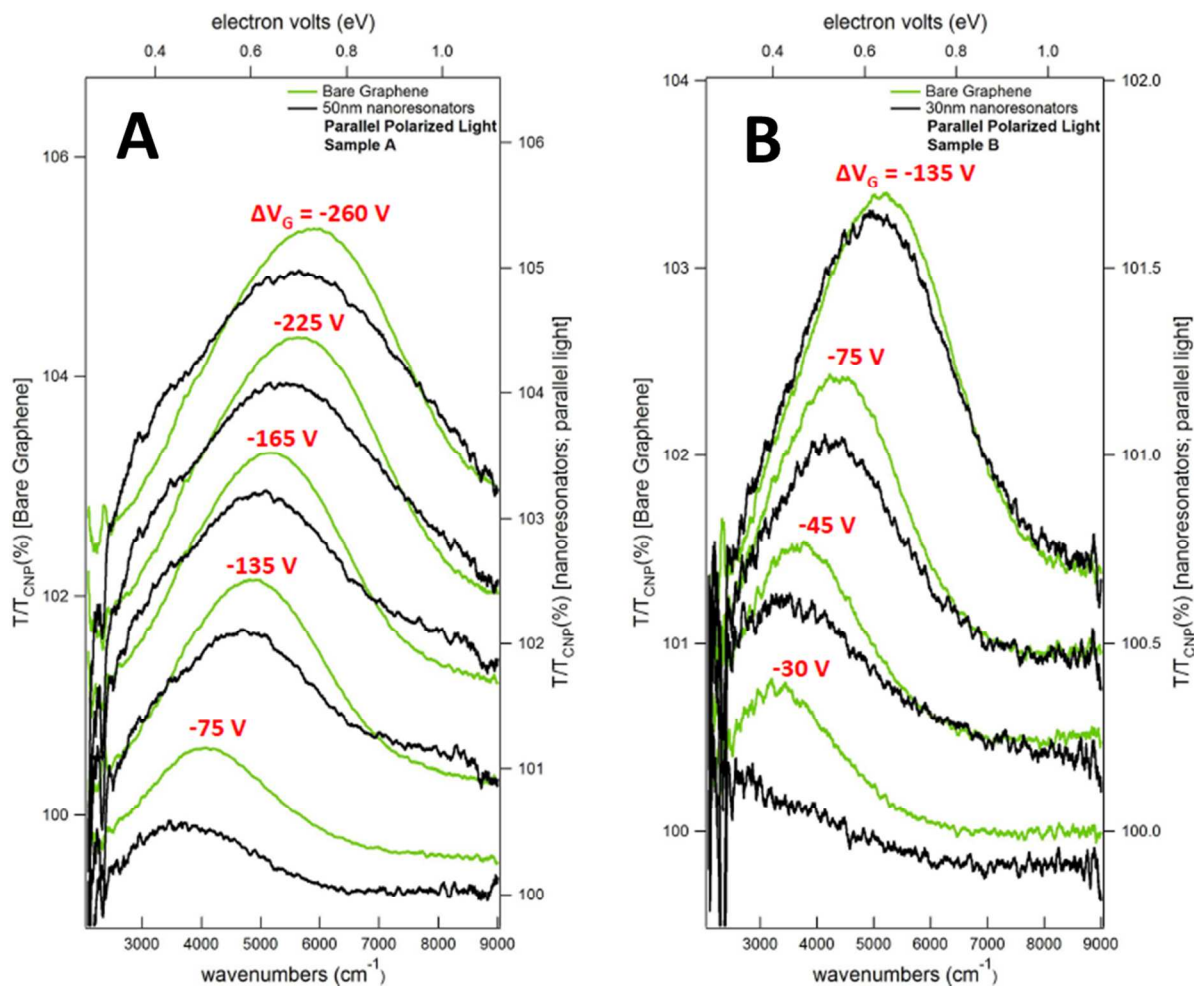


Figure S2. Normalized transmission spectra obtained from graphene nanoresonators at different gate voltages (black lines) compared to spectra obtained at identical gate voltages from the bare graphene (green lines) adjacent to the nanoresonator patterns. Data was collected with light aligned parallel to the graphene nanoresonators. Sample A was prepared using an Iron(III) Chloride etch, and sample B was prepared using an Ammonium Persulfate etch.

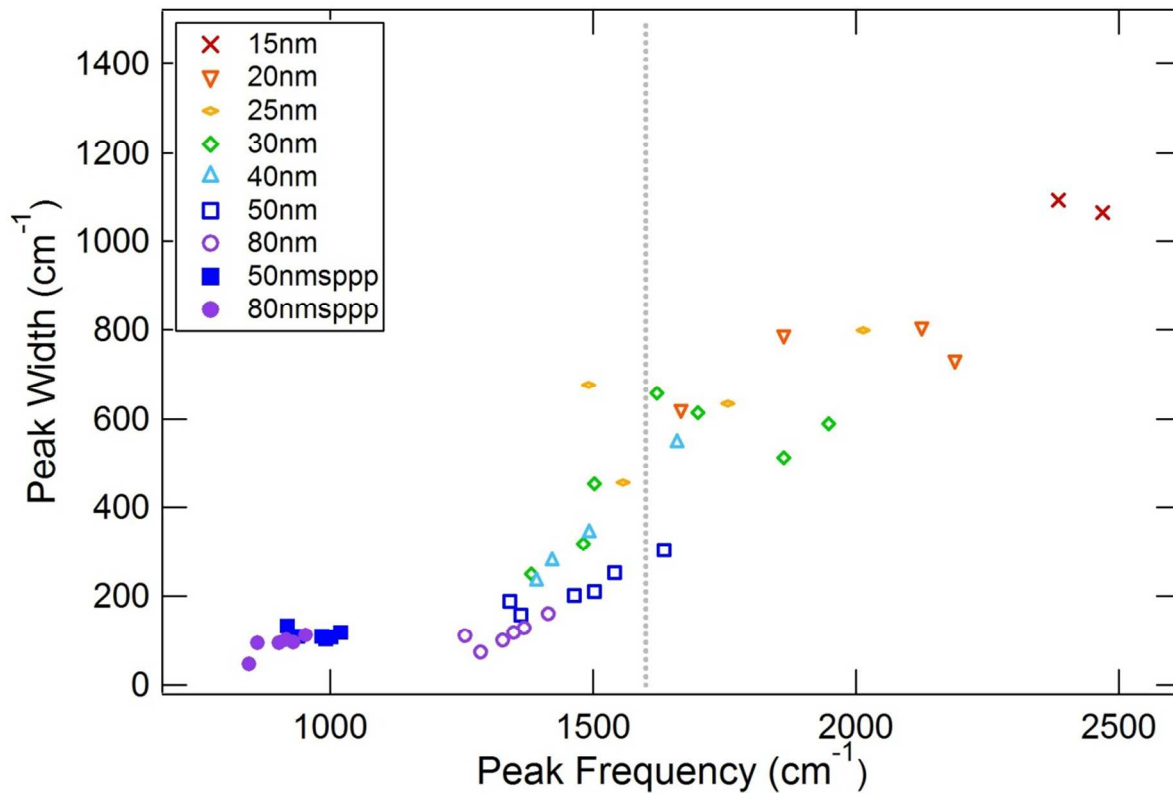


Figure S3. Dependence of peak width $\Delta\omega$ on peak frequency of GP (open colored symbols) and SPPP (filled colored symbols) resonances. The characteristic width of nanoresonator varies from 15nm to 80nm. The dotted vertical line indicates the energy of in-plane optical phonons of graphene.

1. Thongrattanasiri, S.; Silveiro, I.; de Abajo, F. J. G. *Applied Physics Letters* **2012**, 100, (20), 201105-4.
2. Li, Z. Q.; Henriksen, E. A.; Jiang, Z.; Hao, Z.; Martin, M. C.; Kim, P.; Stormer, H. L.; Basov, D. N. *Nat Phys* **2008**, 4, (7), 532.
3. Jablan, M.; Buljan, H.; Soljacic, M. *Phys Rev B* **2009**, 80, (24), 245435.