Supplementary Information for:

Double-layer graphene for enhanced tunable infrared plasmonics

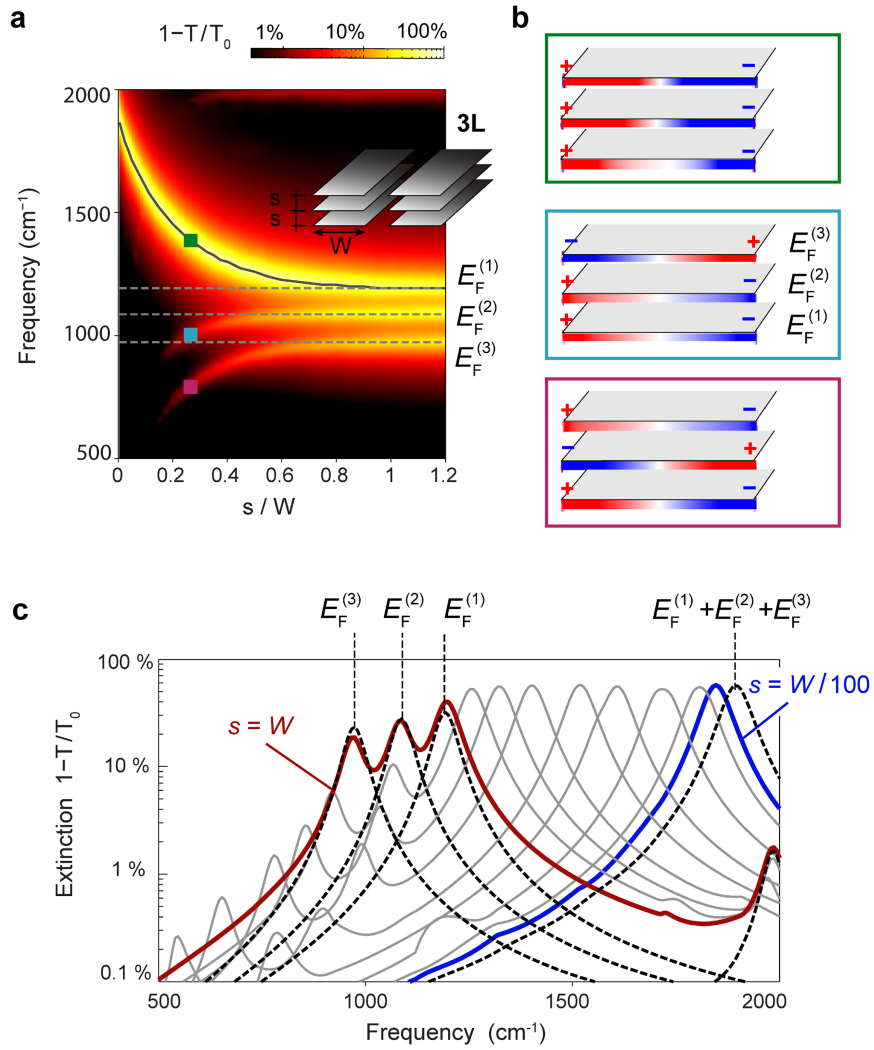
Daniel Rodrigo[1], Andreas Tittl[1], Odeta Limaj[1], F. Javier García de Abajo[2,3], Valerio Pruneri[2,3], and Hatice Altug[1]*

[1] Institute of BioEngineering, École Polytechnique Fédérale de Lausanne (EPFL), CH-1015 Lausanne, Switzerland.

[2] ICFO - Institut de Ciències Fotòniques, The Barcelona Institute of Science and Technology, 08860 Castelldefels (Barcelona), Spain

[3] ICREA - Institució Catalana de Recerca i Estudis Avançats, 08010 Barcelona, Spain

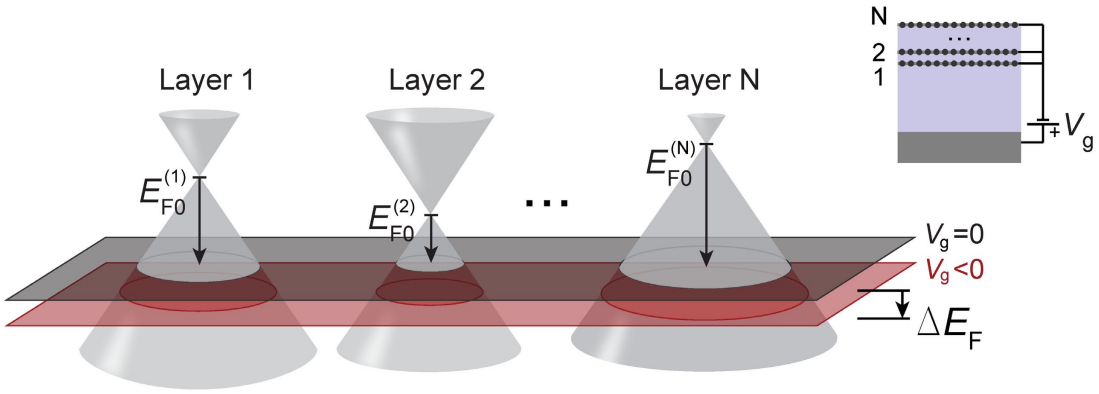
* Corresponding author: hatice.altug@epfl.ch



Supplementary Figure S1. (a) Simulated extinction coefficient of a 3L-GNRA. The three graphene layers have Fermi levels $|E_F^{(1)}|$=0.30 eV, $|E_F^{(2)}|$=0.25 eV, $|E_F^{(3)}|$=0.20 eV, relaxation time τ=100 fs and are embedded in a dielectric with refractive index n=1.41. The ribbon width is W=40 nm and the graphene layers are separated by a distance s. Dashed lines represent the resonances of the individual layers. (b) Electric charge distributions across the ribbons for the 3L-GNRA with s=W/4 at the three resonance frequencies indicated in the figure (780, 995, and 1400 cm^{-1}). (c) Simulated extinction spectra of the 3L-GNRA (solid lines) for an interlayer separation varying from s=W (red line) to s=W/100 (blue line). Dashed lines represent the extinction spectra of 1L-GNRAs with Fermi levels $|E_F^{(1)}|$, $|E_F^{(2)}|$, $|E_F^{(3)}|$ and $|E_F^{(1)}|+|E_F^{(2)}|+|E_F^{(3)}|$.

Supplementary Note 1. Electrostatic biasing in multi-layer graphene stacks (all layers having same doping type)

We consider a stack of *N* graphene layers in close proximity, that are electrically connected and are biased through a common backgate (Supplementary Figure S2). In this section we calculate the equivalent Fermi level of the biased multi-layer structure (E_F^{NL}). We assume that all graphene layers have the same doping type (p or n). The case when layers have different doping types p/n is addressed in Supplementary Note 2.



Supplementary Figure S2. Band diagram representation of a multi-layer graphene stack composed of two layers electrically interconnected. The black/red planes represent the Fermi level before/ after biasing.

The initial doping of each layer is $E_{F0}^{(i)}$ for i=1...N, defined as the Fermi level relative to its Dirac point. The Dirac cone of each layer shifts along the energy axis so the Fermi levels of all layers are aligned. After applying a biasing voltage V_g, each layer reaches a Fermi level $E_F^{(i)}$ and the increment of Fermi level is the same for all layers.

$$E_F^{(i)} - E_{F0}^{(i)} = E_F^{(1)} - E_{F0}^{(1)}, \qquad i = 1\ldots N \qquad \text{(S1)}$$

Additionally, the total amount of carriers injected by the bias is given by the charge in the capacitor formed by the thin SiO2 layer with surface capacitance C_{ox}:

$$\sum_{i=1}^{N} n_s^{(i)} - n_{s0}^{(i)} = C_{ox} V_g / e \qquad \text{(S2)}$$

Taking into account the relation between Fermi level and carrier density in graphene ($E_F = \hbar v_F \sqrt{\pi n_s}$) we have:

$$\sum_{i=1}^{N}\left(E_{\mathrm{F}}^{(i)}\right)^2-\left(E_{\mathrm{F0}}^{(i)}\right)^2=\pm A\,V_{\mathrm{g}} \qquad \text{(S3)}$$

where $A=C_{\mathrm{ox}}\hbar^2{v_F}^2\pi/e$ and the plus/minus sign accounts for the common doping type of the graphene layers.

Substituting (S1) in (S3),

$$0=\sum_{i=1}^{N}\left[\left(E_{\mathrm{F}}^{(1)}-E_{\mathrm{F0}}^{(1)}+E_{\mathrm{F0}}^{(i)}\right)^2-\left(E_{\mathrm{F0}}^{(i)}\right)^2\right]\mp A\,V_{\mathrm{g}}$$

$$0=\sum_{i=1}^{N}\left[\left(E_{\mathrm{F}}^{(1)}\right)^2+2\left(E_{\mathrm{F0}}^{(i)}-E_{\mathrm{F0}}^{(1)}\right)E_{\mathrm{F}}^{(1)}+\left(E_{\mathrm{F0}}^{(1)}\right)^2-2E_{\mathrm{F0}}^{(1)}E_{\mathrm{F0}}^{(i)}\right]\mp A\,V_{\mathrm{g}}$$

$$0=N\left(E_{\mathrm{F}}^{(1)}\right)^2+2\left(E_{\mathrm{F0}}^{\mathrm{NL}}-NE_{\mathrm{F0}}^{(1)}\right)E_{\mathrm{F}}^{(1)}+N\left(E_{\mathrm{F0}}^{(1)}\right)^2-2E_{\mathrm{F0}}^{(1)}E_{\mathrm{F0}}^{\mathrm{NL}}\mp A\,V_{\mathrm{g}}$$

where $E_{\mathrm{F0}}^{NL}=\sum_i E_{\mathrm{F0}}^{(i)}$.

Solving the second-order equation,

$$E_{\mathrm{F}}^{(1)}=\left[E_{\mathrm{F0}}^{(1)}-\frac{E_{\mathrm{F0}}^{\mathrm{NL}}}{N}\right]+\frac{1}{2N}\left[4\left(E_{\mathrm{F0}}^{\mathrm{NL}}-NE_{\mathrm{F0}}^{(1)}\right)^2-4N^2\left(E_{\mathrm{F0}}^{(1)}\right)^2+8NE_{\mathrm{F0}}^{(1)}E_{\mathrm{F0}}^{\mathrm{NL}}\pm 4NA\,V_{\mathrm{g}}\right]^{1/2}$$

$$E_{\mathrm{F}}^{(1)}=\left[E_{\mathrm{F0}}^{(1)}-\frac{E_{\mathrm{F0}}^{\mathrm{NL}}}{N}\right]+\left[\left(\frac{E_{\mathrm{F0}}^{\mathrm{NL}}}{N}\right)^2\pm\frac{1}{N}A\,V_{\mathrm{g}}\right]^{1/2}$$

The other solution of the second-order equation is non-physical because it does not satisfy $E_{\mathrm{F}}^{(1)}\Big|_{V_g=0}=E_{\mathrm{F0}}^{(1)}$.

Due to symmetry in the equations (variables can be exchanged without modifying the system of equations) the previous expression for layer *i*=1 is also valid for any layer.

$$E_{\mathrm{F}}^{(i)}=\left[E_{\mathrm{F0}}^{(i)}-\frac{E_{\mathrm{F0}}^{\mathrm{NL}}}{N}\right]+\left[\left(\frac{E_{\mathrm{F0}}^{\mathrm{NL}}}{N}\right)^2\pm\frac{1}{N}A\,V_{\mathrm{g}}\right]^{1/2}$$

Then the equivalent Fermi layer of the multi-layer stack is calculated by adding all the individual Fermi levels ($E_{\mathrm{F}}^{NL}=\sum_i E_{\mathrm{F}}^{(i)}$), which we initially assumed to have the same sign (same doping type).

$$E_{\mathrm{F}}^{\mathrm{NL}}=\sum_{i=1}^{N}\left[E_{\mathrm{F0}}^{(i)}-\frac{E_{\mathrm{F0}}^{\mathrm{NL}}}{N}\right]+N\left[\left(\frac{E_{\mathrm{F0}}^{\mathrm{NL}}}{N}\right)^2\pm\frac{1}{N}A\,V_{\mathrm{g}}\right]^{1/2}$$

$$E_{\mathrm{F}}^{\mathrm{NL}} = \left[(E_{\mathrm{F0}}^{\mathrm{NL}})^2 \pm NA\, V_{\mathrm{g}} \right]^{1/2}$$

Then,

$$(E_{\mathrm{F}}^{\mathrm{NL}})^2 = (E_{\mathrm{F0}}^{\mathrm{NL}})^2 \pm NA\, V_{\mathrm{g}}$$

and using the relation between Fermi level and carrier density ($E_{\mathrm{F}} = \hbar \mathrm{v}_F \sqrt{\pi n_s}$) we have

$$n_{\mathrm{s}}^{\mathrm{NL}} = n_{\mathrm{s0}}^{\mathrm{NL}} \pm NC_{\mathrm{ox}} V_{\mathrm{g}}/e$$

where $n_{\mathrm{s}}^{NL} = \left(\sum_i \sqrt{|n_{\mathrm{s}}^{(i)}|} \right)^2$ is the equivalent number of carriers in the multi-layer structure.

Therefore, when all the graphene layers have the same doping type (p or n), the equivalent carrier density in a multi-layer graphene stack varies linearly with the voltage V_{g} and biasing is enhanced by a factor *N* (the number of layers) respect to SLG.

Supplementary Note 2: Electrostatic biasing in multi-layer graphene stacks (general case)

We consider the general case of biasing a multi-layer graphene stack where the different layers may have different doping types (before or after applying a bias voltage). This can be generally solved by considering the same equations (S1) and (S2) from the previous section.

$$E_{\mathrm{F}}^{(i)} - E_{\mathrm{F0}}^{(i)} = E_{\mathrm{F}}^{(1)} - E_{\mathrm{F0}}^{(1)}, \qquad i = 1\ldots\mathrm{N} \qquad \text{(S1)}$$

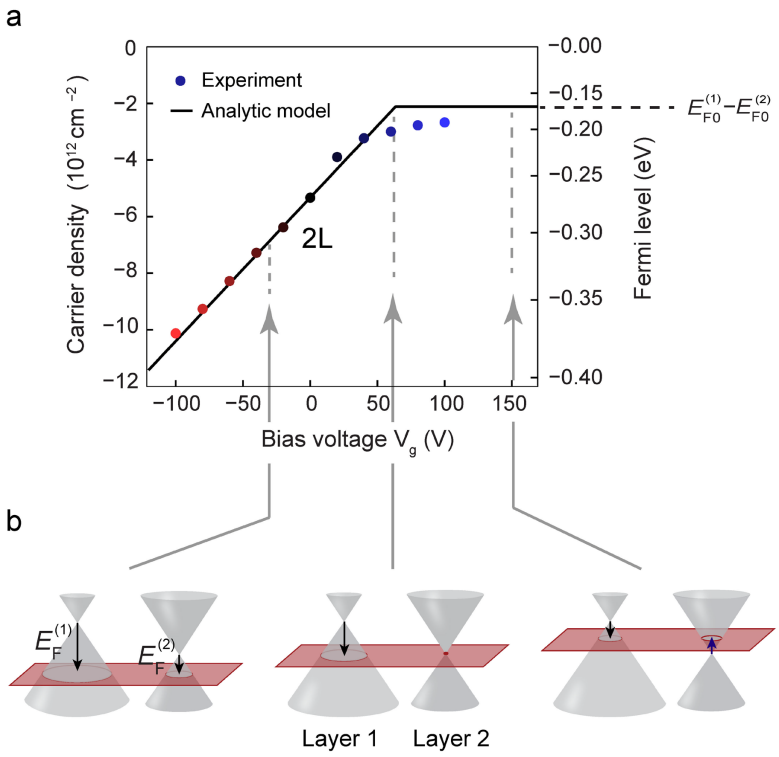
$$\sum_{i=1}^{N} n_{\mathrm{s}}^{(i)} - n_{\mathrm{s0}}^{(i)} = C_{\mathrm{ox}} V_{\mathrm{g}}/e \qquad \text{(S2)}$$

However, equation (S3) has to be modified to account for the sign of doping

$$\sum_{i=1}^{N} \left[\left(E_{\mathrm{F}}^{(i)}\right)^2 \mathrm{sgn}\left(E_{\mathrm{F}}^{(i)}\right) - \left(E_{\mathrm{F0}}^{(i)}\right)^2 \mathrm{sgn}\left(E_{\mathrm{F0}}^{(i)}\right) \right] = A\, V_{\mathrm{g}} \qquad \text{(S4)}$$

Solving equations (S1) and (S4) allows to calculate the equivalent Fermi level of the multi-layer stack, defined as $E_{\mathrm{F}}^{\mathrm{NL}} = \sum_i |E_{\mathrm{F}}^{(i)}|$. In contrast to the discussion in section (S3), the Fermi levels of each layer may have different signs and have to be added up in absolute value as a consequence of graphene ambipolarity.

In Supplementary Figure S3 we show the solution of the previous equations for a DLG (*N*=2) and compare them against the experimental data. We observe that this analytic model captures the saturation observed in the experimental data occurring for bias voltages above +40V. This saturation is caused by the Fermi level crossing the Dirac point of one of the layers, leaving the two layers with opposite doping types. In this case, a shift of the overall Fermi level will increase the absolute doping of one layer and decrease in the same amount the absolute doping of the second layer, which has different doping type. The net result when doping levels are added up $E_{\mathrm{F}}^{2L} = \left|E_{\mathrm{F}}^{(1)}\right| + \left|E_{\mathrm{F}}^{(2)}\right|$ is a zero variation, explaining the saturation effect. For voltages where the two layers have different doping types, the equivalent Fermi level of the structure remains constant at a level equal to the difference between the initial doping levels $E_{\mathrm{F}}^{2L}\big|_{\mathrm{saturation}} = \left|E_{\mathrm{F0}}^{(1)} - E_{\mathrm{F0}}^{(2)}\right|$. In our experimental data the Fermi level saturates between 0.15 eV and 0.20 eV, indicating that the two graphene layers in our device have significantly different doping levels. This is consistent with the device stack-up, where the graphene layers are exposed to different environments: the bottom layer is in contact with silica while the top layer is exposed to air. From the experimental data we estimate that the graphene layers have respective doping levels of approximately $E_{\mathrm{F}}^{(1)}$ =−0.22 eV and $E_{\mathrm{F}}^{(2)}$ =−0.05 eV.



Supplementary Figure S3. (a) Equivalent carrier density of 2L graphene extracted from experiments (circles) and calculations (solid line). The calculations are performed solving equations (S1) and (S2) for $E_F^{(1)}$ =−0.22 eV, $E_F^{(2)}$ =−0.05 eV. (b) Band diagram representation of the DLG for different biasing voltages. The red plane represents the Fermi level.