Optimizing Broadband Terahertz Modulation with Hybrid Graphene/Metasurface Structures

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Supplementary Information

S I: Theoretical derivation of THz power transmission T as a function of the average local field enhancement in the slit (η):

In the coordinate system shown in Figure S1, $z = 0$ denotes the interface between the slit array and medium on the transmission side. To relate electrical fields in both regions, we apply the tangential continuity boundary condition from Faraday's law at the interface:

$$E_x^{III}(x, z = 0) = E_x^{II}(x, z = 0) \quad (1)$$

$E_x^{II}(x, z)$, $E_x^{III}(x, z)$ are the x component of electric field in region II and region III respectively.

We perform Fourier Transform (FT) with respect to variable x on both sides of Eq. (1). Note that both sides are periodic functions with a period of P. The FT of left hand side (LHS) results in a sum of planar waves for arbitrary z:

$$E_x^{III}(x, z) = \frac{2\pi}{P} \sum_{n=-\infty}^{\infty} \tilde{E}_x^{III}(k_n) e^{i\zeta_n z} e^{ik_n x} \quad (2)$$

while $k_n = 2\pi \frac{n}{P}$ and $\tilde{E}_x^{III}(k_n)$ denotes the corresponding Fourier amplitudes. The wave vector in z for each term is determined by Helmholtz equation as $\zeta_n = \sqrt{(\frac{2\pi}{\lambda})^2 - {k_n}^2}$, where λ is the effective wavelength in region III. Eq. (2) is also known as the Rayleigh-Bloch Expansion.

In the subwavelength regime (in which the slit-array device is designed to work) where $\lambda > P$, it is easy to see that ζ_n is always imaginary except for the term $n = 0$. Since imaginary ζ_n leads to evanescent waves that decay in z direction and cannot propagate into far field in region III ($z \gg \lambda$), the far field transmission (P_{trans}) is solely determined by the $n = 0$ term of Eq. (2). In other words, only the zeroth order of transmission contributes to our measured transmission in experiment.

Mathematically we have:

$$E_x^{III}(x, z \to \infty) = \frac{2\pi}{P} \tilde{E}_x^{III}(0) e^{i\zeta_0 z} \quad (3)$$

Therefore, the transmission intensity can be written as $P_{trans} = \frac{1}{2} E_x H_y^* = \frac{|\frac{2\pi}{P}\tilde{E}_x^{III}(0)|^2}{2Z_{III}}$, where Z_{III} is the electromagnetic impedance in region III.

The FT of the right hand side (RHS) of Eq. (1) can be written in a similar form as well:

$$E_x^{II}(x, z = 0) = \frac{2\pi}{P} \sum_{n=-\infty}^{\infty} \tilde{E}_x^{II}(k_n) e^{ik_n x} \qquad (4)$$

Substitute Eq. (4) and Eq. (2) to Eq. (1), we have:

$$\sum_{n=-\infty}^{\infty} \tilde{E}_x^{III}(k_n) e^{ik_n x} = \sum_{n=-\infty}^{\infty} \tilde{E}_x^{II}(k_n) e^{ik_n x} \qquad (5)$$

By Eq. (3), (5) and orthogonality of each Fourier Transform component, we can rewrite electrical field at far field in Region III as:

$$E_x^{III}(x, z \to \infty) = \frac{2\pi}{P} \tilde{E}_x^{III}(0) = \frac{2\pi}{P} \tilde{E}_x^{II}(0) \quad (6)$$

On the other hand, from the inverse FT we have:

$$\frac{2\pi}{P} \tilde{E}_x^{II}(0) = \frac{1}{P} \int_{-D/2}^{D/2} E_x^{II}(x, z = 0) dx \quad (7)$$

Since the tangential field $E_x^{II}(x, z = 0)$ is zero everywhere on top of gold (gold is assumed to be Perfect Electric Conductors in THz regime), we have:

$$E_x^{III}(x, z \to \infty) = \frac{1}{P} \int_{-w/2}^{w/2} E_x^{II}(x, z = 0) dx = \frac{\eta E_0 w}{P} \quad (8)$$

Where $\eta \equiv \frac{1}{w} \frac{\int_{-\frac{w}{2}}^{\frac{w}{2}} E_x^{II}(x,z=0)dx}{E_0}$ is explicitly the average field enhancement factor, defined as $\langle \frac{E_{gap}}{E_0} \rangle$, inside the slit. E_0 is the field amplitude of incident THz wave.

Putting all these pieces together, we have the final expression for THz power transmittance:

$$T = \frac{P_{trans}}{P_{incident}} = \frac{\frac{1}{2Z_I}\left(\frac{\eta E_0 w}{P}\right)^2}{\frac{1}{2Z_{III}}E_0^{\ 2}} = \frac{Z_{III}}{Z_I}\left(\frac{\eta w}{P}\right)^2 \qquad (9)$$

Z_{III} is the impedance in the region of incident light (region III). When $Z_I = Z_{III}$, we have:

$$T = \left(\frac{\eta w}{P}\right)^2 \qquad (10)$$

This is the Eq. (1) in the main text.

S II: Determine the conductivity of graphene

For the devices we show THz modulation in the main text (Fig. 3 and Fig. 4) we simultaneously measure the resistance of the device in a two terminal geometry by applying a small bias. The resistance we measure is the sum of contact resistance and that of graphene sheet carrying current. Considering the geometry factor and assuming a gating capacitance for ion gel, we have the following expression:

$$R = R_{contact} + \frac{1}{ge\mu\sqrt{n_{imp}^2 + \left(\frac{C|V-V_{cnp}|}{e}\right)^2}} = R_{contact} + \frac{1}{ge\sqrt{(\mu n_{imp})^2 + \left(\frac{\mu C|V-V_{cnp}|}{e}\right)^2}} \qquad (11)$$

Where V_{cnp} is the charge neutral point (CNP) of graphene, C is the gating capacitance, V is the gating voltage for electrostatic gating, μ is graphene mobility, n_{imp} is impurity doping at $V = V_{cnp}$, g is geometry factor of the graphene in study, and e is the electron charge.

For our control device of bare graphene, we measure the geometry ratio to be $g = \frac{w}{L} = \frac{2}{3}$. For the slit hybrid device where the major conducting channel is the graphene outside of the slit array (graphene on top of gold slit array is shorted), we measure the geometry factor to be $g = \frac{w}{L_{out}} = 4$.

For both devices, we fit the experimental data according to Eq. (11). We fix the values for g, V_{cnp} . $R_{contact}, \mu n_{imp}$ and μC are fitting parameters. V is the fitting variable.

For the control device of bare graphene, $R_{contact} = 190\ \Omega$, $\mu C = 5000\ \mu F/(Vs)$, $\mu n_{imp} = 1500\ \times 10^{12}\ (Vs)^{-1}$. The value of μC agrees with previous studies for ion-gel gated graphene[1,2], where $\mu \sim 1000\ cm^2/(Vs)$ and $C \sim 6\ \mu F/cm^2$. For the graphene-slit hybrid device, we have the best fitting (red trace in Fig. S2b) with the fitting parameters $R_{contact} = 140\ \Omega, \mu C = 1100\ \mu F/(Vs)$, $\mu n_{imp} = 1200 \times 10^{12}\ (Vs)^{-1}$.

With the value of $R_{contact}$, we can extract graphene conductivity for different gate voltages using

$$\sigma = \frac{1}{(R - R_{contact})g}$$

We use this derived graphene conductivity to plot Fig 4 (c) in the main text.

S III: Fabrication of the periodic gold lit array device

The periodic gold slit structure is fabricated on top of 300 $nm\ SiO_2/Si$ substrate with standard photolithography followed by 5 nm/80 nm Ti/Au evaporation and lift-off. The low resistivity ($10 - 20\ Ohm \cdot cm$) silicon substrate is chosen to avoid substantial THz absorption by the substrate.

A typical optical image of the $2 - 20\ um$ (width $2\ um$ and period $20\ um$) periodic gold slits array is shown in Figure S4. The pattern is uniform and the lift off is clear with no noticeable metal shortage between the slits across the entire device region ($4\ mm$ by $5\ mm$).

S IV: Simulation for THz transmission

A series of Finite Element Method (FEM) simulation is performed to obtain the simulation results used in relevant figures in the main text, using the 2D Radio Frequency (RF) frequency domain analysis module in Comsol 4.4.

To simulate the THz transmission of bare graphene device, we set up a periodic boundary condition for a unit cell with period of 20 um in the x (horizontal) axis. The unit cell is divided into three layers in z (vertical) axis to construct the $air - SiO_2 - Si$ interface. Thickness of SiO_2 layer is set to be 300 nm, and the thickness for air and silicon layer is $100\ um$. Two ports are set up on the top (Port 1, for wave excitation) and bottom (Port 2, for transmission measurement) of the unit cell, as shown in Fig. S4a. The thickness for Si layer is chosen so that transmission is measured sufficiently far away from the grating structure to avoid interference of evanescent waves near the slit array. THz wave is excited from port 1 and propagates to port 2 with a normal incident angle and TM polarization (only E_x , H_y is non-zero). A line current $I_x = \sigma E_x$ is set up at the $air - SiO_2$ interface to simulate the interaction of transferred graphene sheet on top of SiO_2 with local electrical field, where σ is the conductivity of graphene. The simulation zone is depicted in Fig. S4a. Mesh size is refined until the result converges.

Modulation on THz transmission is calculated by sweeping σ from $4\ G_0$ to $80\ G_0$ and recording the power transmission $|S_{21}|^2$ at frequency of 1THz

For the graphene-slit hybrid device, a similar unit cell is used. The geometry is divided into four layers to construct $air - slit\ array - SiO_2 - Si$ interface. Thickness of the slit array layer is $80\ nm$ and SiO_2 layer is 300 nm thick, while the thickness for Air and Si layer remains $100\ um$. Perfect Electrical Conductor (PEC) condition is applied to the boundaries of gold in the slit array. Material inside the slit is *air* and slit width is 2 μm. A line current $I_x = \sigma E_x$ is set at the $air - slit\ array$ interface to simulate graphene transferred on top of slit device. Excitation and transmission ports are set similarly as the bare graphene device case. The simulation zone is depicted in Fig. S4b.

Modulation of THz transmission is obtained by sweeping σ and recording $|S_{21}|^2$ at frequency of 1THz, in a similar fashion as in the bare graphene case.

Removing the line current I_x and sweeping frequency instead, we obtain THz transmission data for the slit device alone. The field profile and local field enhancement can be calculated using numerical tools in Comsol. In particular, local field enhancement η is calculated by integrating $|E_x|$ along the $air - slit\ array$ interface divided by the width w of the slit.

Reference:

(1) Horng, J.; Chen, C.; Geng, B.; Girit, C.; Zhang, Y. *Phys. Rev. B* **2011**, *83*, 165113.

(2) Kim, B. J.; Jang, H.; Lee, S.-K.; Hong, B. H.; Ahn, J.-H.; Cho, J. H. *Nano Lett.* **2010**, *10*, 3464–3466.

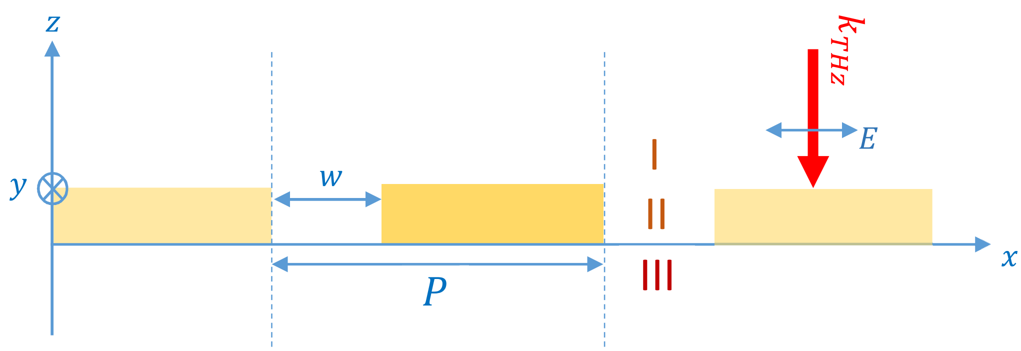

Figure S1. Schematic representation of the periodic slit arrays in $x - z$ plane. Yellow blocks represent the part of gold. P is the period of the gold slit arrays and w is the width of the slit. TM polarized THz light is normally incident from Region I on the slit array (Region II), and the far field transmission is measured in Region III.

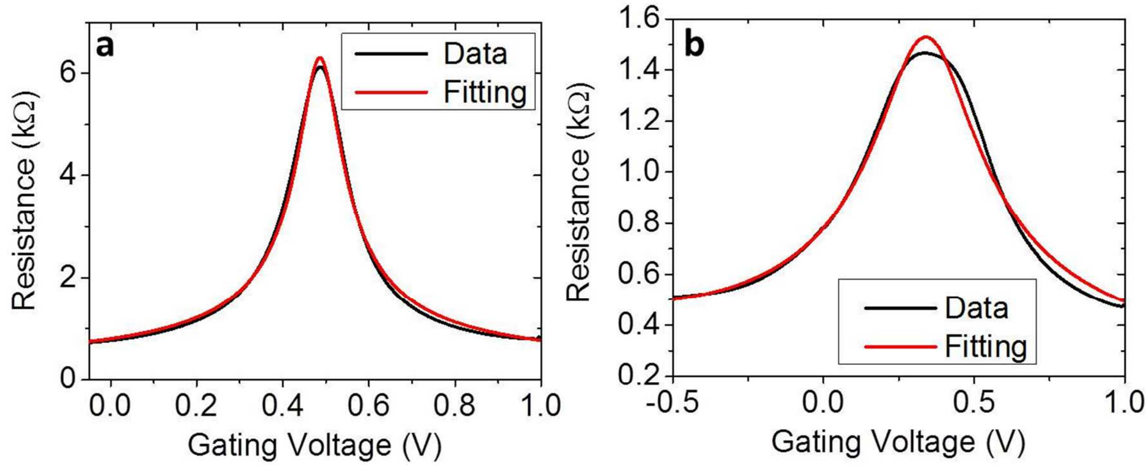

Figure S2. Electrical transport characterization simultaneously taken during the THz transmission measurement (Figure 4 in the main text). (a) Resistance measured for the bare graphene device on SiO2/Si substrate. (b) Resistance measured for the graphene-slit array hybrid device on SiO2/Si substrate.

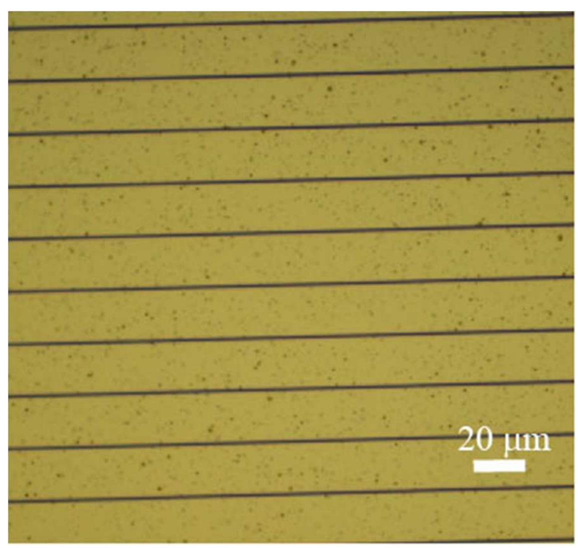

Figure S3. Optical image of a 2-20 μm gold slit-array device.

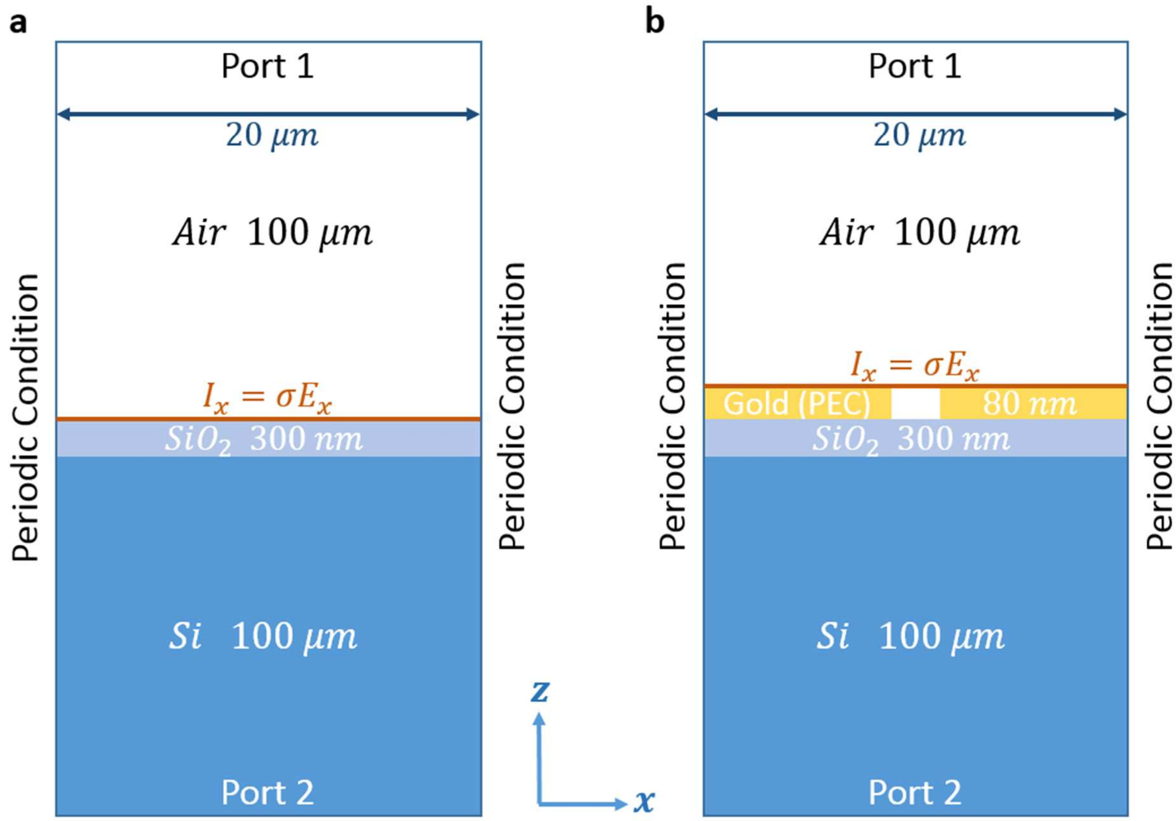

Figure S4: FEM simulation set-up. (a) The set-up for bare graphene device. (b) The-set up for graphene-slit array hybrid device. In both cases a line current with $I_x = \sigma E_x$ is used to simulate the graphene sheet, with σ being the conductivity of graphene.