**Supplementary Information for**

**Ultrafast terahertz transparency boosting in graphene meta-cavities**

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According to the study of ultrafast transient response revealed by previous work, monolayer and multilayer graphene shows a transient decrease or increase in terahertz conductivity by ultrafast optical excitation \([1],[2]\). Although a variety of studies have been performed on graphene, very few fundamental studies have been done to explore the possibilities of patterned graphene-based devices for ultrafast photonics \([3-7]\). Here, we exploit the photoexcitation properties of the graphene to actively modulate the amplitude in a patterned graphene-embedded hybrid device on an ultrafast time scale.

**S1 Theoretical analysis and Simulation**

**Fig. S1a** shows a simulation results of the metal structure. Terahertz wave excites the dipole resonant modes of rings, and a resonance peak is induced through the mutual coupling effect, with a strong local field bounded in the gap. In this case, a small change in the graphene conductivity is not sufficient to cause large frequency shifts and mainly induces significant amplitude modulation under optical pumping.

Excluding the effects of graphene, **Fig. S1b** and c also shows that the field strength in the gap depends on the width of the rings. The nonlinear response of the system could be greatly enhanced by properly choosing geometric parameters of metasurface cavity.
Figure S1. Simulation of SRRs-SiC and terahertz oscillation field enhancement for GMCs. a. The simulated spectra of SRRs-SiC. b. The integration of the nonlinear field along \( l \) versus rings width \( (w) \). c. Field confinement in the gap between the rings plotted along the gap (vertical axis) for different rings width at 0.5 THz.

We calculate the relationship of the Drude weight \( D \), the frequency and the decrease in the the real part of graphene conductivity, as shown in Fig.S2a. Fig. S2b demonstrate the simulated transmission of GMCs. We decrease the graphene conductivity from 3.22\( G_0 \) to 2.38\( G_0 \), corresponding transmission of terahertz waves at 0.5 THz boosts from 0.54 a.u. to 0.65 a.u. level.

To explore the underlying physics of the active transparency boosting behavior, the widely used two-oscillator model is adapted to analyze the near-field interaction between the two rings in the unit cell [8],[9]:

\[
\begin{align*}
  x_1 + \gamma_1 x_1 + \omega_0^2 x_1 + \kappa x_2 &= g_1 E, \\
  x_2 + \gamma_2 x_2 + \omega_0^2 x_2 + \kappa x_1 &= g_2 E
\end{align*}
\]  

(S1)

Here \( x \) and \( \gamma \) are the amplitude and the damping rate, subscripts 1 and 2 represent the inner and outer resonators, respectively. \( g \) is a geometric parameter indicating how strong the resonator couples with the incident electromagnetic wave \( E \). In this equation, the \( \kappa \) denotes the coupling coefficient. Compared with the terahertz wavelength, the thickness of the meta-graphene layer is negligible and the transmission coefficient can be expressed as:
where \( n_{\text{sap}} \) is the refractive index of the substrate and \( c \) is the light velocity in vacuum. The magnetic susceptibility \( \chi_e \) is expressed by \( \gamma, \kappa \) and \( g \), which characterizes the polarization intensity of the metasurface unit cell to the incident electromagnetic field. Fig. S2c shows the fitting results to transmission spectra via various graphene conductivity, exhibiting good agreement with the experiment. The fitting parameters \( \gamma_1, \gamma_2 \) and \( \kappa \) are shown in Fig. S2b. It was observed that the damping coefficients \( \gamma_1 \) and \( \gamma_2 \) do not significantly change with the decrease of graphene conductivity, while the coupling coefficient \( \kappa \) increases significantly and then remains almost constant. Therefore, utilization of the metasurface cavity structure helps to increase the terahertz intensity via coupling effect, offering higher enhancement efficiency.

**Figure S2.** a, The relationship of the Drude weight \( D \), the frequency and the decrease in the real part of graphene conductivity, wherein the oscillation intensity \( F=4.3 \times 10^8 \Omega^{-1}\text{s}^{-1} \); line width \( \gamma=11.5 \text{ THz} \); resonant frequency \( \omega_0/2\pi=2.7 \text{ THz} \). b, Simulated transmission of GMCs-SiC. c and d are fitting transmission coefficients and corresponding parameters under various graphene conductivity, respectively.
**S2. Nano-fabrication and characterizations of the graphene metasurface cavities**

We fabricate an array of hybrid structure consisting of multilayer graphene and metal resonators by standard photolithography, as shown in Fig. S3. First, a large-scale graphene multilayer (1×1 cm²) is grown on a 4H-SiC substrate by CVD method. Then, a 20 nm-thick Au layer is deposited on the surface of the graphene to prevent it from being contaminated and damaged by PMMA. The PMMA pattern of the graphene region is produced by standard lithography process, and then the Au layer and the graphene layer outside the region are etched using KI+ I₂ solution and oxygen plasma equipment, respectively. Through photoetching, electron-beam evaporation, and lift-off processes, a complex metal layer of Au/Ti (200/10 nm) is orderly deposited onto the sample to form metal structure. The improved fabrication approach is easy operation. Most importantly, it keeps the graphene layer away from the undesired photo-resist contamination, surface air oxidation and damage during the fabrication process.

**Figure S3. Fabrication.** Fabrication flow of the graphene metasurface cavities. Here we show 8 steps to realize the GMCs, by depositing graphene, nano-patterning and lithography etching.

We use X-ray photoelectron spectroscopy to estimate the number of multilayer graphene layers. Fig.S4a shows the XPS spectrum of C 1s, as shown by the black line. The spectral line presents two peaks, the first one corresponding to the C in the substrate SiC (282.5eV), and the second peak derived from the sp² hybrid C atom in graphene (284.5eV) [10],[11]. Using attenuation models, graphene thickness can be estimated [12]:

\[
t_{\text{graphene}} = \lambda_C \ln \left( \frac{I^C_{\text{graphene}}}{I^C_{\text{SiC}} + I^C_{\text{buffer}}} + 1 \right)
\]

(S3)
where $\lambda_c=1.8$ is the electron escape depth, $I_{\text{graphene}}^c$, $I_{\text{SiC}}^c$ and $I_{\text{buffer}}^c$ is the integral area of graphene, SiC and C atom in the buffer layer obtained by Gaussian peak division method. The final estimate is about seven layers of graphene. Besides, Raman spectroscopy is measured using Renishaw inVia Raman Microscopes with 532 nm laser excitation at room temperature 300K (Fig. S4b). As the Raman spectrum suggests, the dashed orange curve plots the measured result of the SiC substrate, background noise in the band 1500 cm$^{-1}$ to 1900 cm$^{-1}$ is induced by the phonons in SiC. The solid blue curve plots the case of the GMCs, here two dominant Raman peaks of graphene, the G peak (~1582 cm$^{-1}$) and the 2D peak (2696 cm$^{-1}$) are clear, verifying the existence of graphene layers. Furthermore, we measure the Raman spectra of different samples with diverse doping states, as shown in Fig. S4c and S4d. With varied $E_F$, spectral shift and intensity alteration of the G peak and the 2D peak in samples are characterized. Typically, for the amplitude of graphene Fermi level $|E_F|$ increases from 0 eV to higher, one can see the intensity increment and spectral right shift on G peak, meanwhile intensity decrement and spectral left shift on 2D peak [13].

**Figure S4. Characterization of the graphene in GMCs.** a, The X-ray photoelectron spectroscopy of multilayer graphene. Here the peak at 282.5 eV denotes the C-Si key on boundary, while the peak at 284.5 eV presents the C-C key in graphene. b, Raman spectrum of the graphene on the metasurface cavities (solid blue curve) and SiC substrate (dashed orange curve). c and d, Measured Raman spectra at points with different doping state (or $E_F$) on different GMCs samples. In c and d, the arrows mark Fermi level increases from 0 to 0.3 eV.
S3. Extended information of experiment

Fig. S5 illustrates the experimental setup. The laser source passes through a beam splitter with the average power of 1kW. 70% of the output of the laser is used for the generation of THz waves. The other part of the laser is used as the pumping light in our GMCs sample. The on-sample spot diameter is 1 cm (larger than the terahertz spot diameter of 2~3 mm), which ensures the uniform optical excitation on the planar surface of the sample. So, the maximum peak power of the pump laser reaches 10 GW/cm² on sample. A silicon wafer (1 mm) is placed behind the first parabolic mirror to block the laser, and only allow the terahertz wave to pass through. The temporal delay between THz pulses and laser pulses is tuned with the optical delay line (Newport M-436). When the two pulses overlap in time, the terahertz resonance gradually increased, and the resonance intensity reached the maximum at 0 ps. And we can obtain the obvious THz amplitude enhancement in the spectral domain after using the standard Fourier transform of the terahertz time-domain waveform.

Figure S5. Experimental setup. a. Schematic diagram of the setup, with both terahertz wave excitation and detection. b. Pictures of the setup. Left: the whole setup on optical table; right: the GMCs sample on holder.
Figure S6. Characterization of the laser source and THz transmission. a. Measured pulses of laser source with a repetition rate of 1kHz. b. Measured self-beat-note of the laser source, The SNR is > 60 dB, demonstrating the high stability of the laser source. c. Measured temporal traces of the terahertz transmission and the corresponding Fourier transform spectrum.

Here we introduce the light source in detail. A Ti: sapphire laser (Newport Corporation, SOL-ACE35F1) with central wavelength 800 nm and 35 fs pulse duration is used as the laser pump. The time-domain of the laser source is shown in Fig. S6a, it illustrates stable pulses of the laser source with a fixed repetition rate of 1kHz. Fig. S6b plots the electrical self-beating notes of the laser, the SNR of the beating-line is higher than 60 dB. The laser focuses into a 1 mm thick <110> ZnTe crystal, thus THz wave is generated though optical rectification in ZnTe. In the spectral domain, the generated THz pulses is broadband, from about 200 GHz to typically 3 THz (Fig. S6c) [14].

In Fig. S7a, we test the long-term power stability of the laser source. The laser power uncertainty is < 0.1 dB with a continuous measurement of 60 minutes at room temperature. Besides, we also measure the stability of the THz intensity generated by the laser through ZnTe (Fig. S7b). Due to the fluctuations of environmental temperature and humidity, the uncertainty of the THz signal is a bit higher than the laser source (~0.25dB).
Figure S7. Long-term stability and reliability of the laser pump and the terahertz source. a. The laser power is measured continuously in 1 hour, and the result shows the power uncertainty <0.15dB. b. The THz intensity is measured continuously in 1 hour, and the result shows the power uncertainty <0.25dB

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