Electrically Tunable Terahertz Plasmonic Metasurfaces Employing Multilayer Graphene

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Abstract: We demonstrate electrically tunable terahertz metasurfaces employing multilayer graphene realized by repeated transfer and stacking of monolayer graphene. Such multilayer graphene plasmonic structures exhibit significant increase of plasmonic resonance frequency compared to monolayer graphene structures.

1. Introduction

Graphene plasmonics features two key advantages, i.e. exceedingly high spatial confinement and large frequency tunability, compared to conventional noble metal based plasmonics in the mid-infrared and terahertz (THz) spectral regions, and has recently found a variety of applications in THz photonic devices [1]. Nevertheless, the frequency tuning range of plasmonic devices based on monolayer graphene is ultimately limited by its carrier density tuning range. Here, we demonstrate that the achievable resonance frequency range of graphene-based plasmonic devices can be significantly increased by employing multilayer graphene structures [2]. Our experimental characterizations of plasmonic metasurfaces consisting of double-layer and three-layer graphene ribbon arrays show that their gate-tunable THz plasmonic resonance frequencies exhibit approximately 20% and 30% increase, respectively, compared to that of the corresponding monolayer graphene ribbon array. Furthermore, contrary to the previous prediction [3], employing even more graphene layers for such plasmonic structures yields little additional benefit, as the interlayer charge screening effect leads to negligible gate-induced carrier density in the additional graphene layers. Our findings provide new insights for designing and optimizing graphene-based plasmonic structures for various photonic device applications, such as modulators, sensors and detectors.

2. Device Structure and Fabrication

The schematic of the demonstrated multilayer graphene THz plasmonic metasurfaces is illustrated in Fig. 1(a). The multilayer graphene is realized by repeatedly transferring large-area monolayer graphene (synthesized on copper foil by chemical vapor deposition) onto the substrate using the standard PMMA-based graphene transfer process. The substrate is a lightly doped silicon substrate with a ~300 nm silicon dioxide layer on the surface, which also functions as the back-gate for the graphene structures. After the transfer process, we pattern periodic arrays of graphene ribbons with ~1 μm ribbon width and 2 μm periodicity using photolithography followed by oxygen plasma etching. Metal contacts (Ti/Au) are then deposited on the patterned graphene structures using an electron beam evaporator. Several graphene ribbon array plasmonic metasurfaces with the number of graphene layers ranging from 1 to 4 are fabricated. Figure 1(b) shows a scanning electron microscopy (SEM) image of a fabricated three-layer graphene ribbon array.

3. Experimental Results

We characterize the carrier density dependent plasmonic resonances of the fabricated multilayer graphene ribbon arrays using a Fourier transform infrared spectrometer (FTIR). All the measurements are conducted with the devices placed inside the vacuum chamber of the FTIR, and transmission extinction spectra of each device at various back-gate voltages (graphene carrier densities) are obtained. Figure 1(c) shows the normalized transmission extinction spectra (solid curves) of a three-layer graphene ribbon array at various gate voltages relative to the charge neutrality point ($\Delta V_{\text{gate}} = |V_{\text{gate}} - V_{\text{CNP}}|$), in comparison to those of a monolayer graphene ribbon array (dashed curves). It can be clearly seen that as ΔV_{gate} changes from 45 V to 90 V, the plasmonic resonance frequency of the three-layer graphene ribbons varies in the range of $\sim 200\ \text{cm}^{-1}$ to $225\ \text{cm}^{-1}$ (~6.0 to 6.8 THz), which is significantly higher than that of monolayer graphene ribbons in the same ΔV_{gate} range. Figure 1(d) shows a comparison of the plasmonic resonance frequencies of three graphene ribbon arrays with different numbers of layers (i.e. 1 to 3 layers) at various ΔV_{gate}. It is evident that the plasmonic resonance frequency increases significantly when the number of layers varies from one to three. However, we also observe that employing even more graphene layers yields little additional increase of the plasmonic resonance frequency. As discussed in details in the next section, the increase of the plasmonic resonance frequency with the number of graphene layers is a direct consequence of the THz optical conductivity of a multilayer graphene being larger than that of a monolayer graphene with the same total carrier density; however, due to a strong interlayer charge screening effect, this benefit is only significant for double-layer and three-layer graphene.

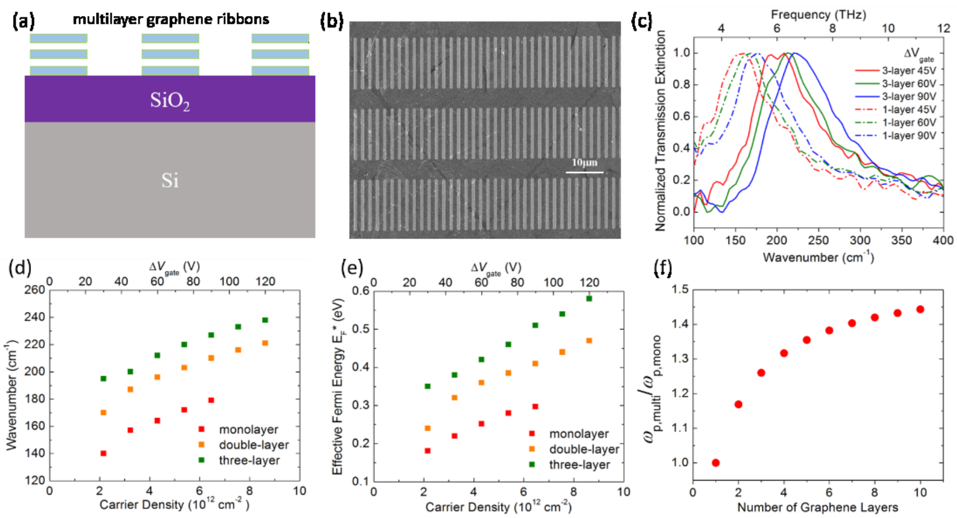

Figure 1 (a) Schematic of investigated multilayer graphene plasmonic metasurfaces. (b) SEM image of a three-layer graphene ribbon array. (c) Normalized transmission extinction spectra of the three-layer graphene ribbon array at various ΔV_{gate}, in comparison to those of the monolayer graphene ribbon array. (d) Plasmonic resonance frequency versus total gate-induced carrier density for three devices with different number of graphene layers. (e) Extracted effective Fermi energy E_F^* versus total gate-induced carrier density for the same three devices in (d). (f) Theoretically calculated graphene plasmonic resonance frequency enhancement factor ($\omega_{\text{p,multi}}/\omega_{\text{p,mono}}$) versus number of stacked graphene layers.

4. Theoretical Model and Simulation

In the THz frequency range, the optical conductivity of a stacked multilayer graphene is the sum of the optical conductivity of each monolayer graphene layer, as long as the thickness of the multilayer graphene is much smaller than the wavelength of interest. The optical conductivity of each monolayer graphene layer is proportional to its Fermi energy, and the graphene Fermi energy E_F is proportional to $\sqrt{n}$, where n is the carrier density of the monolayer graphene. Therefore, when a total carrier density is distributed to several graphene layers, the total optical conductivity is larger than the optical conductivity of a monolayer graphene with the same carrier density [2,3]. We can define an effective Fermi energy $E_F^* = \sum_{i=1}^{N} E_{F,i}$ for an N-layer graphene where $E_{F,i}$ is the Fermi energy for the i-th graphene layer, and hence in this aspect a multilayer graphene is equivalent to a monolayer graphene with the effective Fermi energy E_F^*. We extract the E_F^* by fitting simulation spectra to the experimental ones with E_F^* as the fitting parameter. Figure 1(e) shows the extracted E_F^* as a function of ΔV_{gate} for the same three devices in Fig. 1(d), and it is evident that E_F^* increases significantly as the number of graphene layers increases from one to three. However, as revealed by our theoretical calculation of the carrier density distribution in multilayer graphene which takes into account the strong interlayer charge screening effect, when employing even more graphene layers, the layers above the third one obtain a negligible carrier density and thus have relatively small contribution to the total optical conductivity [2]. Therefore, employing multilayer graphene structures with more than 3 layers does not further improve the plasmonic resonance frequency range significantly. Figure 1(f) shows our calculated plasmonic resonance frequency enhancement factor versus graphene layer number, which is in good agreement with our experimental observation.

5. Conclusion

In summary, we have systematically studied the gate-tunable THz plasmonic resonances of graphene ribbon array structures with different numbers of graphene layers. We find that for double-layer and three-layer graphene structures, the non-equal distributions of gate-induced carriers in different graphene layers lead to a significantly larger total optical conductivity, which in turn produces a considerably higher plasmonic resonance frequency. However, employing even more graphene layers yields little additional benefit in this aspect. Our study provides new insights for designing and optimizing plasmonic structures based on multilayer graphene, which may exhibit crucial advantages over monolayer graphene plasmonic structures for various photonic and optoelectronic device applications.

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References

[1] T. Low, P. Avouris, "Graphene Plasmonics for Terahertz to Mid-Infrared Applications," ACS Nano **8**, 1086-1101 (2014).

[2] G. Li *et al.*, "Multilayer Graphene Terahertz Plasmonic Structures for Enhanced Frequency Tuning Range," ACS Photonics, in press.

[3] D. Rodrigo *et al.*, "Double-Layer Graphene for Enhanced Tunable Infrared Plasmonics," Light: Sci. Appl. **6**, e16277 (2017).