

Graphene superstrates for metamaterials

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Abstract

The electromagnetic properties of a photonic metamaterial are dramatically modified by a graphene monolayer superstrate. The strong polarizability of the graphene layer combined with the Fano-type, resonant plasmonic modes supported by the metamaterial leads to substantial red-shift of the narrow metamaterial resonances. These frequency shifts translate to multi-fold increase in the measured transmission at a specific wavelength. In our experiments we used chemical vapor deposited (CVD) graphene on structurally related metamaterial samples with plasmonic modes at different wavelength positions covering a spectral range from 1600 nm to 1800 nm.

1. Introduction

Since its discovery in 2004 [1], graphene, a flat monolayer of carbon atoms arranged in a honey-comb lattice, attracts growing interest for its unique electronic properties. Graphene demonstrates a particularly strong field-effect related to its ballistic electronic transport [1], that combined with its planar nature, can lead to promising applications in nanoscale electronics [2]. However, the extraordinary electronic properties of graphene do not lead to equally exciting optical properties. Although graphene is known to support plasmons at THz frequencies [3], at optical wavelengths it is barely visible and special techniques are considered in order to verify its presence [4-6]. Here we study the electromagnetic response of a composite medium consisting of single-layer graphene deposited on a metamaterial that supports "trapped mode" plasmonic resonances. It consists of a two-dimensional array of asymmetrically-split ring (ASR) slits on a gold film. Metamaterials of this type are known to show Fano anti-symmetric resonant lines in transmission with enhanced local fields that appear "trapped" in the vicinity of the nanostructure [7]. Such a configuration is of substantial interest both in terms of enabling high-contrast detection of graphene monolayers as well as in terms of possible metamaterial-based sensing applications, where graphene serves as a continuous adsorption layer.

2. Samples and experimental setup

The metamaterial structures were fabricated by focused ion beam milling through a 65 nm thick gold film evaporated on a 102 nm thick Si₃N₄ membrane. Gold film roughness of less than 5 nm was obtained with low pressure (10⁻⁸ mbar) thermal evaporation. We manufactured three almost identical sets of samples with five metamaterial arrays each. The overall size of the arrays was 22x22 μm², while the unit cell D varied among the five arrays from 711 nm to 839 nm. For wavelengths longer than the unit cell side, the metamaterial arrays do not diffract.

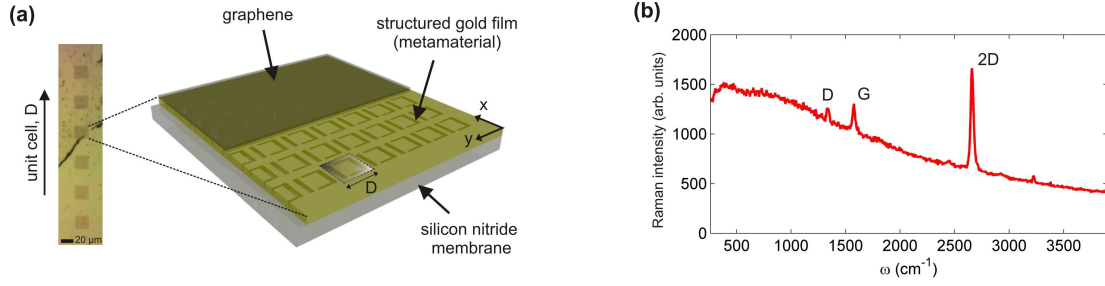


Fig. 1. (a) Schematic of the fabricated complementary split-ring metamaterial. (b) Raman spectrum of graphene on the split-ring metamaterial.

The graphene film was grown on polycrystalline Cu foils by low pressure CVD process [8]. After the growth, the graphene-on-Cu samples were coated with PMMA and were then transferred on to the substrates with the metamaterial structures, where the PMMA was removed by acetone. Raman spectroscopy was carried out to assess the quality and uniformity of the CVD graphene with a WITTEC CRM200 Raman system using a 100X objective lens with a numerical aperture (NA) of 0.95. Figure 1b shows a Raman spectrum with peaks typical for single layer graphene, including a 2D band with a full width at half-maximum (FWHM) of 30 cm^{-1} located at 2670 cm^{-1} and a G band with a much lower Raman intensity than that of the 2D.

In our experiment, part of the sample containing three metamaterial arrays was not covered by graphene, while the rest of the sample containing twelve different metamaterial arrays of five different sizes was covered by epitaxial graphene. The spectral response of the metamaterial before and after graphene deposition was measured experimentally in a micro-photospectrometer (CRAIG). The electric field of the incident field was polarized along the y-axis of Fig. 1a.

3. Results and discussion

In Figure 2a, we present characteristic transmission spectra of an ASR array with a unit cell size $D=711 \text{ nm}$, before and after graphene deposition. The transmission spectrum of the bare metamaterial shows a distinct peak, followed by a transmission dip (marked α) that we attribute to the trapped-mode resonance, while a second transmission peak (marked as β) can be seen at longer wavelengths. The effects of graphene as a metamaterial superstrate are consistent with that of a thin lossy dielectric superstrate: The trapped-mode resonance shifts towards lower frequencies due to the polarizability of graphene, while due to Joule losses the resonance becomes broader with lower (higher) transmission at the transmission maxima (minimum). As a result, although transmission at the peak decreases, when transmission is monitored at a fixed wavelength, a strong increase will be observed due to the resonance shift. This transmission increase can be as high as 250% and is related to the geometry of the array. Arrays of different unit cell size support trapped-mode resonances at different frequencies and, consequently, with increasing unit cell size, the transmission increase appears at longer wavelengths. Importantly the transmission spectra of the uncovered samples before and after deposition of graphene remained unchanged indicating negligible contamination during the graphene transfer process.

In Fig. 2b, we present the relative resonance shift as a function of the unit cell size. Here the relative shift is defined as $(\lambda_f - \lambda_i)/\lambda_i$, where λ_i and λ_f are the positions of the resonances (transmission dip (α) and transmission peak (β)) before and after graphene deposition, respectively. A consistent wavelength shift varying between 5% and 10% is observed for all cases. Intriguingly, the long-wavelength transmission peak (β) appears to experience a much smaller wavelength shift than the trapped-mode resonance. Indeed, for all samples covered with the graphene layer, the wavelength shift is smaller than 2%, although transmission at the maximum drops significantly. We attribute this difference in behavior to the peculiarities of the field structure associated with the trapped-mode and the long-wavelength resonance. It is known that the trapped-mode excitation includes a strong electric dipole

component perpendicular to the ASR plane, while the excitation at the long-wavelength resonance can be represented by a magnetic dipole parallel to the array's plane [9]. We argue that this distinct difference in the nature of the two resonances coupled to the highly anisotropic electric and magnetic optical frequency polarizabilities of graphene leads to the different reactions of the metamaterial to the presence of the graphene layer: a strong response at the trapped-mode resonance frequency and a much weaker response at the long-wavelength resonance.

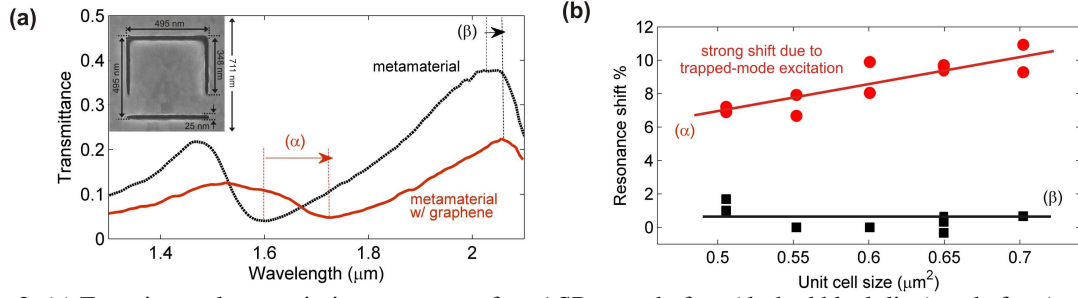


Fig. 2. (a) Experimental transmission spectrum of an ASR array before (dashed black line) and after (red solid line) deposition of graphene for unit cell size $D=711$ nm as shown in the inset. The vertical dashed lines mark the position of the trapped-mode (α) and the long-wavelength dipole (β) resonance, before and after graphene deposition. (b) Wavelength shift of the trapped-mode (α) and of the dipole (β) resonance resulting from the deposition of the graphene layer as a function of unit cell size.

4. Conclusion

In summary, we have demonstrated that trapped-mode metamaterial arrays constitute ideal substrates to enhance the transmission visibility of graphene in the optical part of the spectrum. The observed experimental ratios can exceed 250% with an absolute transmission level of about 10% at a specific wavelength that can be tuned in a broad spectral range by appropriate scaling of the metamaterial structure. This provides a very robust and simple method of detecting graphene and suggests optical sensor applications based on graphene-metamaterial systems.

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