

Graphene and Polarisable Nanoparticles: Looking Good Together?

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ABSTRACT

Several potentially interesting plasmonic effects can arise from combining graphene with polarisable nanoparticles (NPs), such as metallic or dielectric spheres, related to surface plasmon-polaritons (SPPs) supported by the latter in the terahertz (THz) spectral range. Owing to the electromagnetic coupling between the graphene SPPs and dipole moments of polarisable (nano-) particles deposited on top of it, the optical properties of such a composite system have some new features as compared to its constituents. First, the NP's polarizability is renormalized due to the electromagnetic back action of SPPs which are excited in graphene when an external propagating electromagnetic wave impinges on the particle. The coupling also results in a considerable enhancement of the THz radiation absorption in graphene, while the reflection drops to zero. This effect can be potentially interesting e.g. for cloaking in a certain THz frequency range.

Keywords: graphene, surface plasmon, nanoparticle, transmission, absorption.

1. INTRODUCTION

Surface plasmon-polaritons (SPPs) supported by a two-dimensional (2D) electron gas differ in some aspects from their classical counterparts that propagate along metal-dielectric interfaces [1]. In the case of graphene, the electron density oscillations are confined to the one-atom-thick layer, while the electromagnetic (EM) field decays on a much larger scale at both sides of the graphene sheet. Doped graphene sustains SPPs whose frequency (for the SPP wavevector, q , small compared to the Fermi wavevector) is proportional to $n^{1/4}\sqrt{q}$, where n is the 2D electron density [2]. The latter can be changed with an external gate electrode and therefore it is possible to control the SPP dispersion properties electrically. Soon after this theoretical prediction [3,4], it was demonstrated that such surface waves in the THz spectral range could be launched by patterning graphene [5] or just by using an atomic force microscope tip [6,7].

A polarisable particle located above the graphene sheet can also work as an antenna for the SPP excitation. The symmetry breaking by such a particle makes it possible the coupling of propagating EM waves to the SPPs. It has been shown theoretically that a polarisation and the corresponding electric current are induced on graphene by a non-dispersive polarisable particle and the radiation absorption is greatly enhanced by the excitation of either intraband (Drude-type) [8] or interband [9] graphene plasmons. With light emitting NPs (such as quantum dots, QDs) located in the vicinity of graphene, effects such as the superradiance [10], resonance energy transfer [11], and even the regime of strong coupling between the emitter and the plasmons [12] can be expected. In either case, there is a back action of the induced graphene polarisation onto the NP: for a QD it can lead to the establishing of a collective exciton-plasmon excitation [13] or to an enhanced absorption in the particle possessing an optical phonon mode [8] and an electrical instability owing to the energy transfer from drifting electrons to the particle [14]. Particularly attractive is the possibility of electrically controlling such processes by adjusting the gate voltage.

In this communication we analyse the optical properties, in the THz spectral range, of a monolayer of polarisable non-dispersive NPs deposited on a graphene sheet. We show that such a composite layer possesses a resonant mode whose frequency depends on the NP density and the graphene Fermi energy. Incident EM radiation can directly couple to this mode, leading to the enhanced absorption and also transmission. We also discuss the possibility of combining a number of such layers in order to obtain a spectrally broad resonance absorption and transmission.

2. EFFECTIVE SUSCEPTIBILITY OF A PARTICLE MONOLAYER ON GRAPHENE

2.1 Renormalized polarizability of a spherical particle over graphene

The light scattered by the particle can have large in-plane wavevector and couple to the SPPs. Because of the back action of the induced polarization charge on graphene, the particle's polarizability is renormalized. The renormalized polarizability is a tensor, $\hat{\alpha}$, with two distinct principal components, $\alpha_{xx} = \alpha_{yy}$ and α_{zz} (z axis is normal to the graphene sheet). While the bare polarizability of a noble metal spherical NP, α_0 , is dispersionless

in the THz range, the renormalized components do depend upon the frequency, as well as on NP's distance to the graphene sheet, h , and the Fermi energy, E_F , through graphene's optical conductivity [8]:

$$\alpha_{xx} = \frac{\alpha_0}{1 - 2\alpha_0 a(h, \omega)}; \quad \alpha_{zz} = \frac{\alpha_0}{1 - \alpha_0 a(h, \omega)}, \quad (1)$$

where

$$a(h, \omega) = \int_0^\infty r^p(q, \omega) q^2 e^{-2qh} dq$$

and $r^p(q, \omega)$ is the Fresnel coefficient of graphene covered interface between two semi-infinite dielectrics with dielectric constants ε_1 and ε_2 .

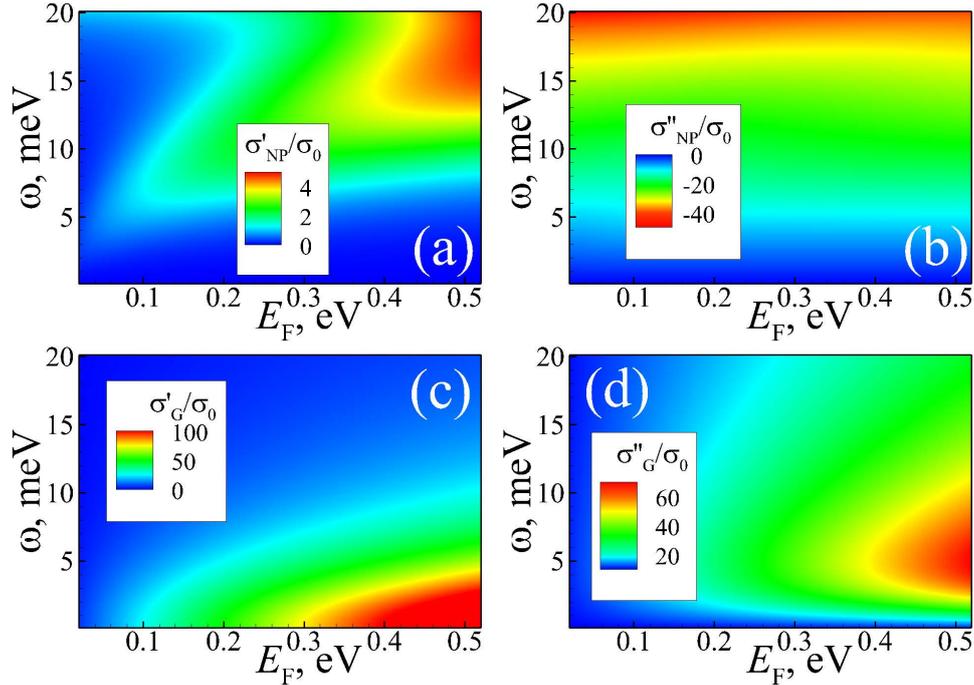


Figure 1. Real [panels (a, c)] and imaginary [panels (b,d)] parts of the effective optical conductivity of a NP monolayer, σ_{NP} (upper panels), and graphene optical conductivity, σ_G (lower panels), both normalised by the DC conductivity of graphene, $\sigma_0 = e^2/4\hbar$ versus frequency (ω) and graphene's Fermi energy for the following parameters: $h=R=10 \mu\text{m}$, $N_{NP} = 8 \times 10^4 \mu\text{m}^{-2}$, electron damping in graphene $\Gamma = 5 \text{meV}$.

2.2 Optical conductivity of a NP monolayer

In the simplest approximation of non-interacting particles, the 2D optical conductivity of a NP monolayer is [8]:

$$\sigma_{NP}(\omega) = -i\omega\alpha_{xx}N_{NP}, \quad (2)$$

where N_{NP} is the NP density. The dipole-dipole interactions can be included in the framework of so called coupled dipole equations (CDEs) [15], which read:

$$\mathbf{d}_i = \hat{\alpha}_i \left(\mathbf{E}_0 + \sum_{j \neq i} \hat{T}_{ij} \mathbf{d}_j \right), \quad (3)$$

where \mathbf{d}_i is the dipole moment of the i -th particle, \mathbf{E}_0 and the external field (of the incident EM wave), and \hat{T}_{ij} is the dipole-dipole interaction tensor, $\hat{T}_{ij} = \hat{T}_{ij}^0 + \Delta\hat{T}_{ij}$ with \hat{T}_{ij}^0 denoting the usual dipole-dipole interaction tensor in an infinite dielectric ($\propto r^{-3}$, r is the inter-particle distance), and $\Delta\hat{T}_{ij}$ representing the indirect particle-particle interaction through the polarisation induced on the graphene-covered interface, which depends on both r and h (see Ref. 16). The CDEs are solved numerically for a quadratic supercell (SC) where NPs are distributed over lattice sites using a Monte Carlo procedure [16] and the solution yields the dipole moments of all particles in the SC. They are used to calculate the SC susceptibility, which can also be expressed in terms of the optical conductivity, $\sigma_{NP}(\omega)$. As an example, Fig. 1 shows the real and imaginary parts of the latter as

functions of ω and the Fermi energy for a certain surface concentration of the particles. The optical conductivity of graphene, $\sigma_G(\omega)$, related to intraband transitions [2] is also shown for comparison.

3. REFLECTION, TRANSMISSION AND ABSORPTION BY GRAPHENE-NP COMPOSITE LAYERS

If the light wavelength is large compared to the NP's radius (which also determines the thickness of the composite layer), it can be easily shown that the optical conductivities of the NP monolayer and graphene simply sum up to determine the composite optical response. For instance, the reflection and transmission coefficients of the composite layer surrounded by two semi-infinite dielectrics with dielectric constants ε_1 and ε_2 are given by [8]:

$$\hat{r} = \frac{\sqrt{\varepsilon_1} - \sqrt{\varepsilon_2} - 4\pi[\sigma_{NP}(\omega) + \sigma_G(\omega)]/c}{\sqrt{\varepsilon_1} + \sqrt{\varepsilon_2} + 4\pi[\sigma_{NP}(\omega) + \sigma_G(\omega)]/c}, \quad (4)$$

$$\hat{t} = \frac{2\sqrt{\varepsilon_2}}{\sqrt{\varepsilon_1} + \sqrt{\varepsilon_2} + 4\pi[\sigma_{NP}(\omega) + \sigma_G(\omega)]/c}. \quad (5)$$

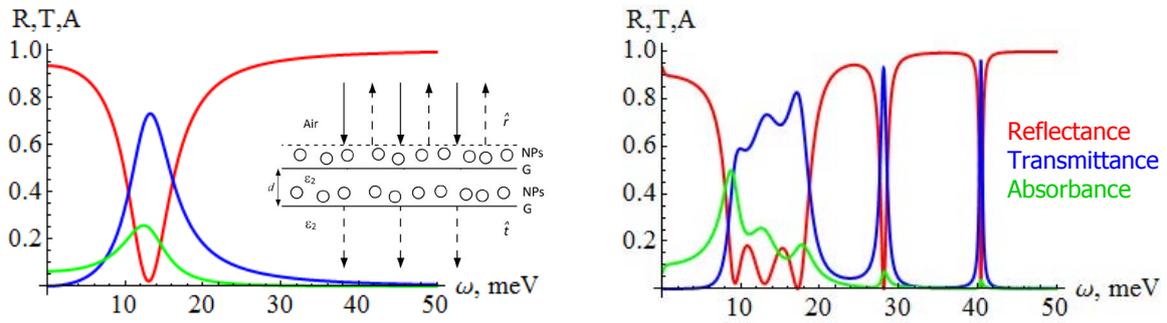


Figure 2. Normal incidence reflectance (red), transmittance (blue) and absorbance (green) spectra of a system composed of two graphene-NP composite layers separated by distance $d \approx 0$ (left) and $d = 47.5 \mu\text{m}$ (right). Particle's radius $R = 10 \mu\text{m}$, concentration $N_{NP} = 8 \times 10^{-4} \mu\text{m}^{-2}$, $h=R$, $E_F = 1 \text{ eV}$ for both layers, and $\varepsilon_1 = \varepsilon_2 = \varepsilon_3 = 1$.

From these relations we notice that when the (large) imaginary parts of $\sigma_{NP}(\omega)$ and $\sigma_G(\omega)$ compensate each other, which is possible according to Fig. 1, the denominator of Eqs. (4) and (5) decreases and it gives rise to enhanced transmission. Under this condition the reflection coefficient becomes very small and it corresponds to a spectral dip in the reflectance (similar to Fig. 2, left panel). Therefore the condition $\text{Im}[\sigma_{NP}(\omega) + \sigma_G(\omega)] = 0$ determines a resonant mode of the composite layer under normal incidence.

In order to enhance these effects, we can consider several composite layers in stack. Employing the transfer matrix method, one can obtain the following expressions for the reflection and transmission coefficients of the stack (the composite layers are separated by dielectric spacers and surrounded by semi- semi-infinite dielectrics ε_1 and ε_3):

$$\hat{r} = \frac{-\sqrt{\varepsilon_3}M_{11} + M_{12} - \sqrt{\varepsilon_1\varepsilon_3}M_{21} + \sqrt{\varepsilon_1}M_{22}}{\sqrt{\varepsilon_3}M_{11} - M_{12} - \sqrt{\varepsilon_1\varepsilon_3}M_{21} + \sqrt{\varepsilon_1}M_{22}}, \quad (4)$$

$$\hat{t} = \frac{2\sqrt{\varepsilon_3}}{\sqrt{\varepsilon_3}M_{11} - M_{12} - \sqrt{\varepsilon_1\varepsilon_3}M_{21} + \sqrt{\varepsilon_1}M_{22}}, \quad (5)$$

where M_{ij} are the elements of the transfer matrix. In the case of two composite layers separated by a spacer of thickness d and dielectric constant ε_2 (shown in the inset of Fig. 2) we have:

$$M_{11} = \cos(k_2d) - i \frac{4\pi\sigma_1}{c\sqrt{\varepsilon_2}} \sin(k_2d); \quad M_{12} = -\frac{4\pi(\sigma_1 + \sigma_2)}{c} \cos(k_2d) + i\sqrt{\varepsilon_2} \left(1 + \frac{16\pi^2\sigma_1\sigma_2}{c^2\varepsilon_2}\right) \sin(k_2d);$$

$$M_{21} = -\frac{i}{\sqrt{\varepsilon_2}} \sin(k_2d); \quad M_{22} = \cos(k_2d) - i \frac{4\pi\sigma_2}{c\sqrt{\varepsilon_2}} \sin(k_2d),$$

where $k_2 = \sqrt{\varepsilon_2}\omega/c$ and $\sigma_1(\sigma_2)$ denotes the effective conductivity, $[\sigma_{NP}(\omega) + \sigma_G(\omega)]$, of the first (second) composite layer. The transmission, reflection and absorption ($A = 1 - R - T$) spectra of the composite bilayer system are shown in Fig. 2 for two different values of the spacer thickness. We can see that for $d \approx 0$ the spectra

characterised by a single resonance node mode, very similar to those of a single composite layer [8] because the effective conductivities simply add in this case. However, for d comparable with the radiation wavelength, interference effects between the two layers lead to a considerable broadening of the nearly zero reflection band from ≈ 10 to ≈ 20 meV (Fig. 2, right panel). We can also notice higher Fabry-Perot-type modes at ≈ 30 and 40 meV, which are quite narrow because they are far away from the composite layers' resonant modes.

4. CONCLUSIONS

In conclusion, we briefly described some potentially interesting optical effects in the THz range that are related to surface plasmon-polaritons in graphene-based composite structures including polarisable (nano-) particles. The induced graphene polarisation, which propagates in the form of SPPs, produces a back-action on the particle and this coupling determines the EM properties of the composite system. It results in a considerable enhancement of the THz radiation absorption in graphene, while the reflection coefficient drops to zero. Stacks of multiple composite layers seem to be reasonably easy to make and their optical properties can be controlled by the interlayer distance and NPs' concentration. This effect can be potentially interesting e.g. for cloaking in a certain THz frequency range.

Some further effects can be expected, not considered here. For instance, one can use plasmonic particles with the surface plasmon resonance in the FIR range, e.g. made of a doped semiconductor. The interaction of localized surface plasmons (in the particle) and delocalized SPPs in graphene may lead to interesting physics (Fano resonances) as well as promising applications in sensors, switches, and filters, as suggested in Ref. [17] for another system. Also, shining light on a small particle located close to the graphene sheet produces a mechanical force on the particle, which can be explored for its optical manipulation.

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