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Interaction between Graphene-coated Nanowires Revisited with Transformation Optics

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The interaction between graphene coated nanostructures provides interesting optical properties not found in isolated graphene plasmonic structures. However, full-analytical solutions, which can provide deep physical insights underlie the hybrid graphene plasmonic systems, are difficult to achieve. In this Letter, we deploy the theory of transformation optics to study the plasmonic interactions between two dielectric-core-graphene-shell nanowires. The scattering and absorption spectra as well as the field distributions are derived analytically. We find that the interaction between two graphene coated nanowires results in polarization-independent multi-frequency Fano dips, which show a broadband redshift of bonding modes and a blueshift of antibonding modes when the nanowires approach each other. The analytical tool presented here offers a rigorous study of graphene plasmonic compound and can be extended to treat more complicated cases. © 2017 Optical Society of America

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Graphene plasmonics have attracted considerable research interest in the past decades for its unique electro-optical properties. Compared to plasmons in noble metal, graphene plasmons feature their high spatial confinement and tunability based on chemical doping and electrical gating [1]. Moreover, shifting the operating wavelength from the optical to the terahertz and mid-infrared regime is essential for particular applications; for example, the detection of biomolecules [2]. Different structures have been proposed to exploit these properties, such as arrays of graphene nanoribbons [3, 4], nanodisks [5] and graphene conductivity gratings [6]. Among various designs, nanoparticles coated with a graphene monolayer have gained much attention. Superscattering peaks [7] and invisibility dips [8] based on the scattering cancellation of these core-shell structures can be observed in their scattering spectra. However, the majority of these studies have been focused on a single graphene coated particle [7, 9, 10]. Similar to its metallic counterpart [11], for a graphene-coated nanowire dimer, the localized surface plasmon modes of single graphene-coated nanowire can interact with each other to form pairs of bonding and antibonding modes, which will further split apart when increasing

the coupling strength. This kind of plasmonic hybridization can provide additional opportunities to engineer the light harvesting abilities and multi-frequency Fano resonances through varying the geometry of the structure. In [12], coupled mode equations for a pair of graphene-coated nanowires are derived to get the conversion frequency for second harmonic generation. However, the calculation of the eigen mode profiles and mode indices still relies on numerical Finite element method (FEM) calculations.

Usually, an analytical solution of the optical response of a two-dimensional (2D) core-shell dimer can be obtained by using classical Mie scattering theory with the aid of multiple scattering. However, this strategy involves higher-order cylindrical harmonics whose treatment can be computationally consuming, especially when separations go down to the sub-nanometre scale. To overcome these drawbacks, transformation optics [13-15] has been proven as a powerful analytical tool to describe the optical properties of singular plasmonic structures, such as crescents [16], kissing cylinders [17] and spheres [18]. By mapping the original geometry into a more symmetric geometry, the efficiency is improved and more physical insights are gained through revealing the hidden symmetry [19]. However, traditional transformation theory cannot be applied to graphene-coated structure because unlike the in-plane permittivity, the conductivity of graphene is no longer preserved under the transformation.

In this letter, we propose a modified TO approach to study the interaction between graphene coated nanowires by introducing a spatially dependent conductivity of graphene in the transformed frame. We show that although this spatial dependence breaks the rotational symmetry of the transformed geometry, it only adds a minor complication to the problem, resulting in a tri-diagonal scattering matrix. Such a matrix can be easily inverted and diagonalized. Based on this modified TO method, the scattering and absorption cross sections are analytically derived and verified by comparing with numerical simulated results. We show that the core-shell dimer under study can support broadband spectrum response with multi-frequency Fano dips under both x and y polarized illuminations, contrary to conventional plasmonic dimers which can achieve broadband response only under one specified polarization [20]. Also, the field distributions are investigated to gain physical insight into the plasmonic modes.

As shown in Fig. 1, as the first step of the TO approach, we transform the original dimer system in Fig 1 (a) into an annular

Fig. 1. Schematics of the transformation. (a) The starting structure consist of two core-shell nanowires. (b) Performing an inversion about the inversion point give rise to an annulus system.

system (Fig 1 (b)) with much more symmetry. The conformal mapping relating the two systems is defined by

$$\xi' = \frac{g^2}{\xi - R_0} \quad (1)$$

This transformation maps points at infinity to the inversion point, R_0 . Note that a similar conformal transformation was applied to study a pair of metal nanospheres separated by a small gap [18].

We shall assume that the cores of the dimer in Fig. 1 (a) are comprised of homogeneous dielectrics defined by a permittivity ϵ_s (here we choose glass as the core material with $\epsilon_s = 2.1025$), immersed in a homogeneous environment defined by a relative permittivity ϵ_b . As for the shell, in the THz range the graphene monolayer is characterized by its surface conductivity σ' in the original frame which can be calculated from the Kubo formula $\sigma'(\omega, \mu_c, \Gamma, T) = \sigma_{intra} + \sigma_{inter}$, where the intraband and interband conductivity can be approximated as [17]

$$\sigma_{intra} = i \frac{e^2 k_B T}{\pi \hbar^2 (\omega + i2\Gamma)} \left(\frac{\mu_c}{k_B T} + 2 \ln(e^{-\mu_c/k_B T} + 1) \right) \quad (2)$$

$$\sigma_{inter} = \frac{ie^2}{4\pi\hbar} \ln \left(\frac{2|\mu_c| - (\omega + i2\Gamma)\hbar}{2|\mu_c| + (\omega + i2\Gamma)\hbar} \right) \quad (3)$$

where e is the element charge, k_B is the Boltzmann constant, Γ is the phenomenological scattering rate, and μ_c is the chemical potential which can be tuned through the gate voltage and/or chemical doping. Here we assume room temperature $T=300$ K, the scattering rate $\Gamma = 0.11$ meV, and the chemical potential is 0.5 eV.

For the case where the shell is made of semiconductor of a certain thickness, its permittivity can be related to the conductivity by

$$\epsilon = 1 + i \frac{\sigma'}{\omega \epsilon_0 \Delta d'} \quad (4)$$

We choose Indium antimonide (InSb) in our calculation as a counterpart to graphene for its small energy band gap and large electronic mobility [21]. The values of permittivity of InSb at THz frequencies are similar to those of metals in the visible and near infrared [22, 23]. The complex permittivity of InSb can be modeled through a Drude-like formula $\epsilon(\omega) = \epsilon_\infty [1 - \omega_p^2 / (\omega^2 + i\omega\gamma)]$ with parameters obtained by fitting experimental data ($\epsilon_\infty = 15.68$, $\omega_p = 1.52 \times 10^{13}$ rad/s, $\gamma = 1.79 \times 10^{12}$ rad/s) [20].

Under the conformal map, the in-plane components of the electric permittivity are conserved [24, 25]. This implies that based on the relation $\sigma = \epsilon \cdot \Delta d$ (where Δd is an infinitesimal distance), the conductivity transforms according to Δd . Assuming in the original frame $\Delta d' = \xi'_0 - \xi'$, in the transformed frame we have

$$\sigma = \frac{|\xi - R_0|^2}{g^2} \sigma' = \frac{\rho^2 + R_0^2 - 2\rho R_0 \cos \varphi}{g^2} \sigma' \quad (5)$$

Different from the metallic nanowire dimers in previous studies where the permittivity of the materials remains unchanged [26, 27], Eq. (5) indicates that graphene is altered under 2D conformal mappings and the transformed graphene acquires a highly non-uniform conductivity, which complicates the boundary equations.

Next, we solve for the electrostatic potential $\varphi(\rho)$ in the simplified annulus frame and hence find $\varphi(\rho')$ in the original frame. $\varphi(\rho)$ in the annulus frame can be expressed as

$$\begin{aligned} & \sum_n a_n^{in} (\rho / R_0)^n, \quad \rho < R_1 \\ & \sum_n \left[(a_n^+ + a_n^{s+}) (\rho / R_0)^n + a_n^- (\rho / R_0)^{-n} \right], \quad R_0 > \rho > R_1 \\ & \sum_n \left[a_n^+ (\rho / R_0)^n + (a_n^- + a_n^{s-}) (\rho / R_0)^{-n} \right], \quad R_2 > \rho > R_0 \\ & \sum_n a_n^{out} (\rho / R_0)^{-n}, \quad \rho > R_2 \end{aligned} \quad (6)$$

where (ρ, φ) is the polar coordinate system (angular factor $\cos n\varphi$ and $\sin n\varphi$ is omitted here) centered at the origin of the Cartesian coordinate system (x, y) , and $a_n^{s\pm}$ is the source term associated with the incident wave. It has been shown in a previous study [28] that the incident plane wave in the origin frame can be represented as a point dipole in the transformed frame. Then by applying boundary conditions across the inner and outer radius of the annulus (i.e. $\Delta E_\varphi = 0$ and $\Delta D_\rho = -i \frac{\sigma}{\omega} \frac{\partial E_\varphi}{\partial \varphi}$ deduced from the continuity of the tangential component of the electric field E_φ and the normal component of the electric displacement field D_ρ take into account the surface charge), we obtain the following matrix equation,

$$\mathbf{R}|a\rangle = \mathbf{S}(|a^s\rangle + |a^r\rangle) \quad (7)$$

where $|a\rangle$ and $|a^s\rangle$ are the scattering coefficients and the source coefficients vectors, respectively. To take account of radiation damping, a radiative correction term $|a^r\rangle$ is included, which are the expansion coefficients associated with a fictive absorbing particle [29]. The matrices $\mathbf{R}$ and $\mathbf{S}$ are calculated as

$$\mathbf{R} = \begin{bmatrix} \mathbf{P} - \mathbf{I} & (\mathbf{P} + \mathbf{I}) \cdot \mathbf{U} \\ (\mathbf{Q} + \mathbf{I}) \cdot \mathbf{V} & \mathbf{Q} - \mathbf{I} \end{bmatrix}, \quad \mathbf{S} = - \begin{bmatrix} \mathbf{P} - \mathbf{I} & 0 \\ 0 & \mathbf{Q} - \mathbf{I} \end{bmatrix} \quad (8)$$

Where $\mathbf{U}$ and $\mathbf{V}$ are diagonal matrices, whose elements are given by,

$$U_{nn'} = \left(\frac{R_0}{R_1} \right)^{2n} \delta_{n,n'}, \quad V_{nn'} = \left(\frac{R_2}{R_0} \right)^{2n} \delta_{n,n'}, \quad (9)$$

Fig. 2. Bonding (blue stars) and anti-bonding (red dots) Modes for (a) glass-core-graphene-shell dimer and (b) InSb dimer for various separations (radius $r_1 = r_2 = 1 \mu\text{m}$).

$\mathbf{I}$ denotes a unity matrix, and $\mathbf{P}$ and $\mathbf{Q}$ are tri-diagonal matrices:

$$\begin{aligned}
P_{nn'} &= \frac{\varepsilon_s}{\varepsilon_0} \delta_{n,n'} + \frac{i\sigma' R_1}{\omega \varepsilon_0 \varepsilon_b g^2} \\
&\left[n(1 + \frac{R_0^2}{R_1^2}) \delta_{n,n'} - \frac{(n-1)^2}{n} \delta_{n,n'+1} - \frac{(n+1)^2}{n} \frac{R_0^2}{R_1^2} \delta_{n,n'-1} \right], \\
Q_{nn'} &= \frac{\varepsilon_s}{\varepsilon_0} \delta_{n,n'} + \frac{i\sigma' R_2}{\omega \varepsilon_0 \varepsilon_b g^2} \\
&\left[n(1 + \frac{R_0^2}{R_2^2}) \delta_{n,n'} - \frac{(n-1)^2}{n} \delta_{n,n'+1} - \frac{(n+1)^2}{n} \frac{R_0^2}{R_2^2} \delta_{n,n'-1} \right],
\end{aligned} \tag{10}$$

Note that the conductivity of the graphene in the transformed frame is spatially variant, with dependence on the azimuthal angle φ . To eliminate the φ -dependence in the final set of boundary equations, we make use of trigonometric identities, resulting in a tridiagonal system of equation where the n -th term couples to $n \pm 1$ terms.

Substituting the solution of the above equation system in the expression of electrostatic polarizability of the structure, we arrive at the electrostatic polarizability γ_x and γ_y for $\hat{x}$ polarized (along dimer's axis, see the schematic in Fig.1) and $\hat{y}$ polarized illumination. And finally, extinction, scattering, and absorption cross-sections can then be deduced as

$$\sigma_{x,y}^{ext} = k_0 \text{Im} \left\{ \frac{\gamma_{x,y}}{1 - ik_0^2 \gamma_{x,y} / 8} \right\}, \sigma_{x,y}^{sca} = \frac{k_0^3}{8} \left| \frac{\gamma_{x,y}}{1 - ik_0^2 \gamma_{x,y} / 8} \right|^2, \tag{11}$$

$$\sigma_{x,y}^{abs} = \sigma_{x,y}^{ext} - \sigma_{x,y}^{sca}$$

Figure 2 shows how the resonant frequencies of eigenmodes shift when varying the gap size between two nanowires with radius $r_1 = r_2 = 1 \mu\text{m}$ in the configurations of (a) a glass-core-graphene shell dimer and (b) an InSb dimer without any shell. We plot the resonant frequencies for both $\hat{x}$ and $\hat{y}$ polarized illuminations. It can be clearly observed that the modes fall into two branches corresponding to two different polarized incident waves; the odd bonding modes ($\hat{x}$ polarized) denoted by blue stars and the even anti-bonding modes ($\hat{y}$ polarized) denoted by red dots. As shown in Figs. 2 (a) and (b), for both structures the odd modes redshift as the nanowires approach each other, whereas the even modes blueshift as the gap reduces. This can be understood by the odd and even nature of modes with respect to the symmetry plane along the y direction, which is well illustrated in their field profiles (Fig. 3). For the odd modes, as the nanowires approach each other, the attraction of opposite charges drags down the modes' frequencies. In contrast, for the even modes, the repulsion of like charges drives up the resonant frequencies as the gap is reduced.

Although the resonant frequency of dimers with and without a shell moves in a similar fashion when altering the gap size, they exhibit entirely different patterns. In Fig. 2(b), for InSb dimer without shell, when the gap is large and comparable to dimer's radius, all the modes are resonant at the vicinity of the surface plasma frequency ω_{sp} , which is similar to an individual nanowire. As the nanowires approach, more odd resonance modes of smaller orders (n) are excited and their frequencies are redshifted toward zero frequency when the gap decreases. While the odd modes spread out to form a continuous and broadband spectrum, the even modes, on the other hand, tend to the bulk plasma frequency

Fig. 3. The potential distribution shown in real space and in the transformed space for glass-core-graphene-shell dimer ($r=1 \mu\text{m}$, separation= $0.1 \mu\text{m}$). The mode order increase from left panel to right panel from dipole mode ($m=1$) to quadrupole mode ($m=2$). For each order, the corresponding dark eigenmode at the same frequency is also shown next to the bright mode. The upper panel corresponding to odd bonding modes, the lower panel is for even anti-bonding modes.

ω_p and are affected by the gap size only to a small extent compared to the odd modes. However, for the glass dimer coated with a graphene monolayer (Fig. 2(a)), branches of the even modes spread out and are paired with the odd modes instead of being constrained to frequencies between ω_{sp} and ω_p , leading to a broadband response for both $\hat{x}$ and $\hat{y}$ polarized illumination. This is a major difference from conventional plasmonic dimers (without graphene shell) which obtain a broadband response only at certain polarization [20]. Furthermore, unlike the InSb dimer case where the odd bonding modes extend to zero frequency as the gap size decreases, for the glass-core-graphene shell dimer, odd and even modes saturate to a finite frequency when the gap size is decreased.

In order to gain more physical insights into the resonant features of the glass-core-graphene-shell dimer, we study the field distributions of eigenmodes. The structure considered here consists of two $1 \mu\text{m}$ -radius glass-core-graphene-shell nanowires separated by $0.1 \mu\text{m}$. Figure 3 displays the potential distribution of dipole ($m=1$) and quadrupole modes ($m=2$) under $\hat{x}$ (upper panel) and $\hat{y}$ (lower panel) polarized incident waves in the physical and transformed spaces, respectively. The field profiles of the corresponding dark modes, which refers to non-radiative modes that cannot couple directly with the incident plane wave, are also shown next to that of the radiative bright mode. From Fig. 3 it can be seen that if the potential distribution in the real space is relatively smooth, in the transformed space it will become highly localized, and vice versa. Note that in the transformed annulus region, the left-hand side of the annulus is transformed to the gap while the right-hand side is mapped to infinity. Previous studies [18] showed that for an annulus comprised of homogenous materials, the mode is almost uniformly distributed. On the contrary, for the graphene case, the mode tends to stay on the right-hand side of the annulus. Correspondingly, in the physical space, the energy is well spread out, leading to the absence of strong hot spots.

Fig. 4. (a) Scattering and (b) absorption cross section for glass-core-graphene-shell nanowires with different radius (5 μm (black), 2 μm (blue), 1 μm (red)) separated by 0.1 μm under x-polarized illumination (odd bonding modes). Theory predictions (solid lines) are compared with simulations (circles).

To further demonstrate the validity of our theory, in Fig. 4 the calculated scattering and absorption spectra of the glass-core-graphene-shell dimer are compared with the results of numerical simulations (COMSOL Multiphysics). Good agreement is observed when the radius of nanowire is relatively small (1–2 μm). For larger structure dimensions, the theoretical predictions are slightly blueshifted compared to the numerical results. This is due to the retardation effects which are not considered in our approach [30]. However, the magnitude and line shape of the spectra are still quantitatively well predicted for nanowire radii up to 5 μm . Two Fano dips can be observed in the scattering spectra which are shifted towards longer wavelength as the radius increases. The dip at the longer wavelength results from the destructive interference between dipole ($n=1$) and quadrupole ($n=2$) modes that are resonant at each side of the dip supported by the dimer [31], which can be regarded as a bright mode and a dark mode which interfere to form the Fano resonance. On the other hand, the sharp invisibility dip located at the shorter wavelength can be attributed to the scattering cancellation of the dielectric core and monolayer graphene shell, similar to the plasmonic cloak and the mantle cloak operating at visible and RF frequencies respectively [8, 32, 33]. The emergences of these Fano dips can lead to huge field enhancement in because energy can be harvested without radiating into the far field [26]. This suggests potential applications in sensitive micro spectroscopy and nonlinear devices.

In conclusion, we have studied the plasmonic interactions between dielectric dimers coated with graphene monolayer using a modified transformation-optics approach. Comparison with simulation has shown that our theoretical model can predict the magnitude and line shape of the spectra for nanowire radii up to 5 μm . Our results show polarization independent broadband properties and multi-frequency Fano dips, contrary to conventional plasmonic dimer structures.

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