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Citation: Applied Physics Letters 109, 031909 (2016); doi: 10.1063/1.4959553
View online: http://dx.doi.org/10.1063/1.4959553
View Table of Contents: http://scitation.aip.org/content/aip/journal/apl/109/3?ver=pdfcov
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Unconventional Fano effect based spectrally selective absorption enhancement in graphene using plasmonic core-shell nanostructures

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(Received 10 April 2016; accepted 12 July 2016; published online 21 July 2016)

We present a design of multilayer core-shell nanostructures formed by introducing a dielectric gap shell layer between a silver core and a monolayer graphene shell for spectrally selective absorption enhancement in graphene based on an unconventional Fano effect. We demonstrate that this mechanism enables great flexibility in the choice of parameters of the proposed structures for the achievement of a relatively large and narrow-band absorption enhancement in graphene. Furthermore, we also demonstrate that such a spectrally selective absorption enhancement in graphene is highly tunable and can be optimized by controlling either the core or the shell parameters. These unique absorption properties may have applications in color-selective photodetectors and image sensors. Published by AIP Publishing. [http://dx.doi.org/10.1063/1.4959553]

Graphene, a two-dimensional honeycomb sheet composed of carbon atoms, exhibits unique electronic and optical properties, which enable its use in a wide range of nanophotonic and optoelectronic devices.1 The relatively weak light absorption (∼2.3%) of a single layer of undoped graphene,2,3 however, severely hampers its applications in devices such as photodetectors4 and tunable modulators.5 Light absorption of graphene could be greatly enhanced by exploiting either propagating6,7 or localized surface plasmon resonances (SPRs)8,9 supported by graphene with appropriate doping. However, these methods are not applicable for the absorption enhancement of graphene in the visible and near-infrared (vis-NIR) spectral range due to the lack of graphene plasmon in this wavelength range.10 A possible way to enhance the vis-NIR light-graphene interactions is to integrate graphene with resonant Fabry-Perot11,12 or photonic crystal microcavities,13–15 where absorption enhancement is gained from the increased effective interaction length. In addition, based on the ability of metallic nanostructures to concentrate light into subwavelength volumes and produce highly localized fields,16 the combination of graphene with conventional plasmonic nanostructures has also been proposed and demonstrated to improve interaction between graphene and vis-NIR light.17–23 In those studies involving metal-graphene hybrid nanostructures, main research focus has been placed on achieving the highest possible absorption enhancement17–23 or broadening its bandwidth,17,20 while narrowing the bandwidth of the absorption enhancement to overcome the limitation induced by the wavelength-independent absorption characteristics of graphene,3 which may have important applications in color-selective photodetectors and image sensors.23,24 has received relatively little attention. Recently, by exploiting the interaction between the graphene and the Fano-like guided mode resonances, a large enhancement of graphene absorption over a narrow bandwidth of few nanometers has been attained in graphene-dielectric grating systems.25,26

In this letter, instead of using array-induced Fano resonances in a periodic structure,25,26 we investigate metal-dielectric-graphene core-shell resonators (MDGCSRs) that exploit unconventional Fano resonances arising from the interference between two electromagnetic Mie modes with the same multipole moment inside the graphene shell27 to realize a large and narrow-band absorption enhancement in graphene as compared to the single-layer metal-graphene core-shell nanostructures. We demonstrate that by simultaneously varying two of the three parameters of the MDGCSRs: the core radius, the thickness, and the refractive index of the dielectric gap shell layer, the spectrally selective absorption enhancement in graphene is able to be easily tuned to different specified visible light wavelengths and, more importantly, can be optimized at each selective wavelength. These unique absorption properties make the proposed MDGCSRs attractive candidates for applications in color-selective photodetectors and image sensors.23,24

We first consider a simple metal-graphene core-shell resonator (MGCSR) consisting of a silver nanosphere wrapped directly by a monolayer graphene, as schematically depicted in the inset of Fig. 1(a). Throughout this paper, the problem of absorption of light by a spherical concentric core-shell particle is solved analytically using Mie theory,28 in which contributions from all the dominant multipolar terms have been taken into account. In the calculations, the monolayer graphene is assumed to be 0.335-nm-thick,10 and its permittivity is described by a Drude-Lorenz model.29 The dielectric constant of silver is described by a Drude model $\varepsilon_{Ag} = 3.7 - \frac{f_p^2}{f(f + i\gamma)}$, where $f_p = 2196$ THz is the plasma frequency of bulk silver and $\gamma$ is the frequency of electron collisions. The efficiency of absorption in the graphene shell ($Q_{gra}$) is given by $Q_{gra} = \sigma_{gra}/\sigma_{geom}$, where $\sigma_{gra}$ is the absorption cross section for the graphene shell and $\sigma_{geom} = \pi r^2$ is the geometrical cross-section with $r$ being the
outer radius of the core-shell nanoparticle. In order to focus on the essence of the underlying physics, we will begin our discussion by neglecting the dissipative loss of silver ($\gamma_r = 0$ THz). The effect of realistic metal loss on the graphene absorption will be discussed later in this work. Since there is only one degree of the freedom in the MGCSRs, the graphene absorption efficiency can be optimized at any given wavelength by varying the radius of the silver core. The calculated $Q_{gra}$ is plotted in Fig. 1(a) as a function of silver core radius ($R_{in}$) and wavelength, which indeed demonstrates that at each wavelength there exists a particular core radius such that the absorption efficiency can reach its maximum.

By further summarizing the achievable maximum absorption efficiency and the corresponding core radius in Fig. 1(b), it is clearly seen that due to the retardation effect, the spectral position of the maximum graphene absorption redshifts with increasing the silver core radius [blue line in Fig. 1(b)], thus providing the spectral tunability of graphene absorption upon the change of the core size. However, the achievable maximum absorption efficiency is found to decay exponentially with red-shifting the spectral position of graphene absorption and is very weak ($Q_{gra} < 0.6$) in the whole visible region [red line in Fig. 1(b)].

In the following, we demonstrate that it is possible to greatly enhance the visible light absorption of graphene by introducing a dielectric gap shell layer between the silver core and the monolayer graphene to form a MDGCSR present in the inset of Fig. 2(a). For such a multilayered core-shell structure, there are three degrees of freedom including the core size ($R_{in}$), the dielectric shell thickness ($t$), and the refractive index of dielectric shell ($n$). Figure 2(a) shows the graphene absorption efficiency calculated at a given wavelength of $\lambda = 400$ nm for the MDGCSRs with the same dielectric shell refractive index of $n = 1.56$ but different core sizes and dielectric shell thicknesses (a) and for the MDGCSRs with the same dielectric shell thickness of $t = 12$ nm but different core sizes and dielectric shell refractive indexes (c). The inset in (a) shows a schematic of a MDGCSR. Marked points SI in (a) and S2 in (c) indicate the achievable maximum efficiency and the corresponding parameters. (b) and (d) Analytically calculated (red lines) and numerically simulated (blue lines) graphene absorption efficiency spectra of a MDGCSR with the optimized parameters corresponding to the points SI (b) and S2 (d), and the Fano fitting results (open symbols).

Fano resonances arising from the interference between two Mie modes with the same multipole moments excited in different layers of the particle. Figure 2(b) shows the graphene absorption efficiency spectrum of the optimized MDGCSR with a set of parameters ($R_{in} = 6$ nm, $n = 1.56$, $t = 15.2$ nm) [marked point SI in Fig. 2(a)], in which a Fano-type line shape is expectedly observed around the wavelength of $\lambda = 400$ nm.

To prove that we are dealing with the Fano resonance in this case, we will fit the absorption efficiency spectrum with the Fano formula $F(\varepsilon) = \sigma_{bg} + \sigma_0(\varepsilon + q^2)/(1 + q^2)$, where $\sigma_{bg}$ and $\sigma_0$ are the background and normalized efficiency, $q$ is the asymmetry parameter, and $\varepsilon = 2(\lambda - \lambda_{res})/\Gamma$ with $\lambda_{res}$ and $\Gamma$ being the resonance position and linewidth, respectively. At the wavelengths of $\lambda_{max} = \lambda_{res} + \Gamma/(2q)$ and $\lambda_{min} = \lambda_{res} - \Gamma/q/2$, the Fano formula can achieve its maximal value $F_{max} = \sigma_{bg} + \sigma_0(1 + q^2)$ and minimal value $F_{min} = \sigma_{bg}$, respectively. Note here that $\lambda_{max}$, $\lambda_{min}$, $F_{max}$, and $F_{min}$ ($\sigma_{bg}$) can be directly acquired from the calculated efficiency spectrum. Therefore, in the fitting procedure, only $\lambda_{res}$ and $q$ are fitting parameters, while $\sigma_0$ and $\Gamma$ are derived from $\sigma_0 = (F_{max} - \sigma_{bg})/(1 + q^2)$ and $\Gamma = 2q(\lambda_{max} - \lambda_{min})/(1 + q^2)$, respectively. The fitting result (open triangles in Fig. 2(b)) is found to be in excellent agreement with the calculated spectrum [red line in Fig. 2(b)] in the vicinity of the resonance, indicating that the graphene absorption efficiency enhancement is due to the excitation of the unconventional Fano resonance in the MDGCSRs. The derived linewidth is as narrow as $\Gamma = 0.82 \pm 8.4 \times 10^{-5}$ nm, demonstrating a highly spectral selective absorption enhancement in graphene.
could also be optimized to maximize the graphene absorption efficiency in MDGCSRs. As a demonstration, the MDGCSRs are assumed to have a fixed shell thickness of \( t = 12 \text{ nm} \). The graphene absorption efficiency at \( \lambda = 400 \text{ nm} \) is plotted in Fig. 2(c) as a function of the core radius and the dielectric shell refractive index. The absorption efficiency of the MDGCSRs with the core radius of \( R_{\text{in}} = 8.4 \text{ nm} \) and the shell refractive index of \( n = 1.58 \) [Fig. 2(c)] reaches a value of \( Q_{\text{gra}} = 10.3 \). As a result of the excitation of the sharp unconventional Fano resonance, the absorption efficiency spectrum calculated for the MDGCSR with the optimized parameters expectedly shows a Fano-like line shape [red line in Fig. 2(d)], which is well described by the Fano fit [open triangles in Figs. 2(b) and 2(d)] with a derived narrow linewidth of \( \Gamma = 2.6 \pm 8.4 \times 10^{-3} \text{ nm} \) around the target wavelength of \( \lambda = 400 \text{ nm} \). Figures 2(b) and 2(d) also show the efficiency spectra calculated using the three-dimensional finite-element software COMSOL Multiphysics (blue lines), in which the transition boundary condition is used on the dielectric-air boundary to model a monolayer graphene shell layer. The simulated results are found to be in good agreement with the results calculated using Mie theory except some small deviation (less than 0.007) around the minimum efficiency. Furthermore, the simulated absorption efficiency spectra can also be well described by the above mentioned Fano formula [open circles in Figs. 2(b) and 2(d)], in which the derived linewidths of \( \Gamma = 0.8 \pm 1.6 \times 10^{-3} \text{ nm} \) and \( 2.4 \pm 3.4 \times 10^{-3} \text{ nm} \) are reasonably consistent with those obtained from the Fano fitting to the analytically calculated spectra (\( \Gamma = 0.82 \pm 8.4 \times 10^{-5} \text{ nm} \) and \( 2.6 \pm 8.4 \times 10^{-3} \text{ nm} \)), verifying the correctness and accuracy of the Mie theory.

Although the graphene absorption efficiency at a given wavelength has been demonstrated to reach its maximum value only for the MDGCSRs with a particular set of optimized parameters, the MDGCSRs with other parameters could still exhibit relatively large absorption efficiency. To demonstrate this, the graphene absorption efficiency at \( \lambda = 400 \text{ nm} \) is calculated for the MDGCSRs with a fixed silver core radius of \( R_{\text{in}} = 10 \text{ nm} \) and plotted in Fig. 3(a) as a function of the dielectric shell thickness and refractive index. As marked by the point \( S4 \) in Fig. 3(a), when the shell thickness and shell refractive index are chosen to be \( t = 10.9 \text{ nm} \) and \( n = 1.6 \), the absorption efficiency could reach its maximum value of \( Q_{\text{gra}} = 10.3 \), demonstrating that the graphene absorption efficiency could be optimized by simultaneously tuning the thickness and the refractive index of the dielectric shell layer in MDGCSRs. For clarity, a dark-red colored area is used in Fig. 3(a) to represent the relatively large absorption efficiency above the value of 9, where the absorption efficiency is about 16 times larger than the achievable maximum graphene absorption efficiency \( (Q_{\text{gra}} = 0.55) \) at \( \lambda = 400 \text{ nm} \) in a MGCSR shown in Fig. 1(b). Figure 3(b) shows the analytically calculated and simulated graphene absorption efficiency spectra of the MDGCSRs with three different sets of parameters \( (n = 1.68, t = 7.2 \text{ nm}), (n = 1.6, t = 10.9 \text{ nm}), \) and \( (n = 1.57, t = 14 \text{ nm}) \), which correspond to the points \( S3, S4, \) and \( S5 \) located inside the dark-red colored region of Fig. 3(a). In each case, the calculated and simulated spectra are in good agreement, and the linewidths derived from the Fano fitting to two spectra are also reasonably consistent with each other and below 7 nm [Fig. 3(b)]. In addition, Fig. 3(b) also shows that the linewidths of the unconventional Fano resonances for three cases are different, indicating a dependency of the thickness and the refractive index of the shell. To more clearly demonstrate this, Fig. 3(c) shows graphene absorption efficiency spectra calculated for the MDGCSRs with the same core radius of \( R_{\text{in}} = 10 \text{ nm} \) and the same shell refractive index of \( n = 1.6 \) but different shell thicknesses of \( t = 9.5 \text{ nm}, 10.5 \text{ nm}, \) and \( 15 \text{ nm} \), which correspond to marked points \( S3, S4, S5, S6, S7, S8, \) and \( S9 \) in (a), respectively. It is clearly seen from Fig. 3(c) that the linewidth is significantly narrowed from \( \Gamma = 4.6 \pm 0.002 \text{ nm} \) to \( 2.5 \pm 0.001 \text{ nm} \) as the shell thickness increases from \( t = 9.5 \text{ nm} \) to \( 15 \text{ nm} \). Meanwhile, the efficiency spectra of the MDGCSRs with the same core radius of \( R_{\text{in}} = 10 \text{ nm} \) and the same shell thickness of \( t = 10.5 \text{ nm} \) but different shell refractive indices of \( n = 1.55, 1.6, \) and \( 1.65 \) [marked points \( S8, S4 \) and \( S9 \) in Fig. 3(a)] are calculated and presented in Fig. 3(d), in which the linewidth is found to be only slightly narrowed from \( \Gamma = 3.9 \pm 0.001 \text{ nm} \) to \( 3.7 \pm 0.001 \text{ nm} \) when the shell refractive index decreases from \( n = 1.65 \) to \( n = 1.55 \).

So far, we have only demonstrated that the graphene absorption efficiency could be greatly enhanced at the wavelength of \( \lambda = 400 \text{ nm} \) due to the excitation of the unconventional Fano resonance in the MDGCSRs. In the frame of Mie theory, the scattering coefficients are dependent on the particle size and refractive index.\(^{28}\) This makes it possible to tune the resonance wavelength of the unconventional Fano resonance and thus the spectral position of the narrow-band absorption enhancement in graphene by varying the silver core radius, the dielectric shell thickness, or the dielectric

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**FIG. 3.** (a) Graphene absorption efficiency at a given wavelength of \( \lambda = 400 \text{ nm} \) calculated for the MDGCSRs with the same silver core radius of \( R_{\text{in}} = 10 \text{ nm} \) but different dielectric shell thicknesses and refractive indexes. The dark–red colored area represents the high absorption efficiency larger than 9. (b), (c), and (d) Absorption efficiency spectra of the MDGCSRs with parameters of \( (n = 1.68, t = 7.2 \text{ nm}), (n = 1.6, t = 10.9 \text{ nm}), (n = 1.57, t = 14 \text{ nm}), (n = 1.6, t = 9.5 \text{ nm}), (n = 1.6, t = 15 \text{ nm}), (n = 1.55, t = 10.9 \text{ nm}), \) and \( (n = 1.65, t = 10.9 \text{ nm}) \), which correspond to the marked points \( S3, S4, S5, S6, S7, S8, \) and \( S9 \) in (a), respectively.
shell refractive index of the MDGCSRs. As an example, we demonstrate here that by fixing the silver core radius to $R_{in}=15$ nm and simultaneously varying the other two parameters, the enhanced optical absorption of graphene could be tuned to longer wavelengths and could be optimized at each selective wavelength. Figures 4(a) and 4(b) show the graphene absorption efficiency calculated at $\lambda = 550$ nm and 700 nm, respectively, for the MDGCSRs with the same core radius of $R_{in}=15$ nm but different shell thicknesses and shell refractive indexes. In each case, there exists a particular combination of the shell thickness and shell refractive index, for example, $(n = 2.83, t = 11.7$ nm) at $\lambda = 550$ nm [marked point $S10$ in Fig. 4(a)] and $(n = 3.47, t = 19.5$ nm) at $\lambda = 700$ nm [marked point $S11$ in Fig. 4(b)], to make the graphene absorption efficiency reach the maximum value. Furthermore, the absorption efficiency of the MDGCSRs with other choice of the shell thickness and shell refractive index located inside the dark-red colored region, as shown in Figs. 4(a) and 4(b), could exceed the efficiency of $Q_{gra} = 10$. In the above cases, perfect silver (without loss) is used. However, in the practice the metal loss has to be taken into account. For this purpose, the frequency of electron collisions in the Drude model is assumed to be $\gamma_e = 4.34$ THz. In addition, a more realistic case where the permittivity of silver is taken from the experimental data by Johnson and Christy $^{34}$ is also investigated here. Figure 4(c) shows the graphene absorption efficiency spectra calculated in the presence of metal loss for the MDGCSRs with a fixed core radius of $R_{in}=15$ nm, in which the shell thickness and the refractive index of the shell are optimized so that the graphene absorption efficiency can reach its maximum value at the wavelength of $\lambda = 550$ nm. For comparison, Fig. 4(c) also shows the graphene absorption efficiency spectrum of the MDGCSR with a lossless silver core and the optimized parameters [marked point $S10$ in Fig. 4(a)]. It is clearly seen from Fig. 4(c) that the achievable maximum graphene absorption efficiency drops from $\sim 10$ to $\sim 5.1$ when $\gamma_e = 4.34$ THz is taken into account in the Drude model and will further drop to $Q_{gra} \approx 2.0$ if the realistic silver is considered, which is still about 37 times larger than the achievable maximum graphene absorption efficiency at $\lambda = 550$ nm in a MGCSR [$Q_{gra} \approx 0.054$, Fig. 1(b)]. In addition to the reduction in the achievable maximum graphene absorption efficiency, the finite dissipation of the silver core will broaden the bandwidth of the graphene absorption. For example, the derived linewidth is found to increase from $\sim 6.8$ nm for lossless silver to $\sim 12.7$ nm for lossy silver described by the Drude model with $\gamma_e = 4.34$ THz and $\sim 23.7$ nm for the realistic silver [Fig. 4(c)].

In summary, we demonstrate that metal-dielectric-graphene multilayered core-shell nanostructures could exhibit large and narrow-band absorption enhancement in graphene as a result of the excitation of the sharp unconventional Fano resonances. Such a mechanism enables great flexibility in the choice of parameters of the proposed structures for the achievement of a relatively large and narrow-band absorption enhancement in graphene. We also demonstrate that the spectrally selective absorption enhancement in graphene could be easily tuned within a wide visible wavelength range and could be optimized at the corresponding wavelength by controlling either the core or the shell parameters. We suggest that the proposed MDGCSRs can be prepared by using the wet-chemistry method to coat silver nanospheres with a dielectric layer, $^{35}$ followed by electrostatic wrapping of graphene sheets on the surfaces of the as-prepared silver-dielectric core-shell nanoparticles. $^{36}$ Our results imply that the proposed MDGCSRs with unique absorption properties would be attractive candidates for applications in color-selective photodetectors and image sensors. $^{23,24}$ We also hope that our strategy could be straightforwardly applied to enhance absorption in other quasi-2D materials.

The authors thank the support by the State Key Program for Basic Research of China (SKPBR) under Grant Nos. 2012CB921501 and 2013CB632703, and the National Nature Science Foundation of China (NSFC) under Grant Nos. 11474215, 91221206, 11321063 and 51271092.

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