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Selective excitation of multipolar surface plasmon in a graphene-coated dielectric particle by Laguerre Gaussian beam

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Localized surface plasmonic resonance has attracted extensive attention since it allows for great enhancement of local field intensity on the nanoparticle surface. In this paper, we make a systematic study on the excitation of localized surface plasmons of a graphene coated dielectric particle. Theoretical results show that both the intensity and frequency of the plasmonic resonant peak can be tuned effectively through modifying the graphene layer. Furthermore, high order localized surface plasmons could be excited and tuned selectively by the Laguerre Gaussian beam, which is induced by the optical angular orbital momentum transfer through the mutual interaction between the particle and the helical wavefront. Moreover, the profiles of the multipolar localized surface plasmons are illustrated in detail. The study provides rich potential applications in the plasmonic devices and the wavefront engineering nano-optics.

Keywords: localized surface plasmon, Mie theory, Laguerre Gaussian beam

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1. Introduction

Localized surface plasmon resonance in metal nanoparticles that results in the resonant absorption, scattering, and near field enhancement around the particle can be tuned across a broadband range from visible to far-infrared wavelength band. The resonant enhancement allows for a wide range of scientific and technological applications, such as optical tweezers,[1–3] surface-enhanced Raman scattering (SERS),[4–6] nano-antennas,[7–10] plasmonic devices,[11] and biosensing.[12,13]

Recently, graphene with two-dimensional Dirac fermions has attracted intensive investigation[14,15] due to its excellent physical properties.[16,17] Graphene decorated nanomaterials and nanostructures have widely been used in the photonic and optoelectronic fields such as photovoltage device,[18] photodetectors,[19] optical modulators,[20] and medical science applications. Since the experimental fabrication of curved and spherical graphene sheets is demonstrated,[21] graphene modulated localized surface plasmon resonance and SERS on nanoparticles are realizable. In this paper, utilizing Lorenz–Mie theory, we make a systematical study on the multipolar localized surface plasmons of the graphene coated dielectric particle excited by Gauss beams with complex wavefront. The detailed results are shown in the following.

2. Theoretical model

In the Lorenz–Mie theory, the general electromagnetic wave can be expanded into vector wave functions in the spherical coordinate system with the origin coincide with particle center[22]

$$E_r^{(\mathrm{i})}(r,\theta,\varphi)=\frac{1}{\tilde{r}^2}\sum_{l=1}^{\infty}\sum_{m=-l}^{l} l(l+1)A_{lm}\psi_l(\alpha\tilde{r})Y_{lm}(\theta,\varphi),$$
$$H_r^{(\mathrm{i})}(r,\theta,\varphi)=\frac{1}{\tilde{r}^2}\sum_{l=1}^{\infty}\sum_{m=-l}^{l} l(l+1)B_{lm}\psi_l(\alpha\tilde{r})Y_{lm}(\theta,\varphi), \quad (1)$$

where the expansion coefficients A_{lm} and B_{lm} that characterize the incident electric and magnetic fields are[23]

$$A_{lm} = \frac{1}{l(l+1)\psi_l(\alpha_2)} \times \int_{0^{2\pi}}\int_0^{\pi} E_r^{(\mathrm{i})}(R,\theta,\varphi)Y_{lm}^*(\theta,\varphi)\sin\theta\mathrm{d}\theta\mathrm{d}\varphi, \quad (2)$$
$$B_{lm} = \frac{1}{l(l+1)\psi_l(\alpha_2)} \times \int_{0^{2\pi}}\int_0^{\pi} H_r^{(\mathrm{i})}(R,\theta,\varphi)Y_{lm}^*(\theta,\varphi)\sin\theta\mathrm{d}\theta\mathrm{d}\varphi. \quad (3)$$

Here $\alpha = n_2 k_0 R$, $\tilde{r} = r/R$, λ is the wavelength, $k_0 = 2\pi/\lambda$ is the wave vector in vacuum, n_2 is the refractive index of the surrounding medium, R is the particle radius, θ and φ are the polar and azimuthal angles, respectively, and $Y_{lm}(\theta,\varphi)$ is the spherical harmonics.

The following procedure is used to obtain the electromagnetic wave solution for the scattering and internal fields of the graphene coated dielectric particle by applying proper boundary conditions at the surface of the particle. In the study, the graphene layer coating is considered as a surface current surround the spherical particle. The boundary condition at the

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particle surface is that the tangential component of the electric field E is continuous and the discontinuity of the tangential component of the magnetic field H is proportional to the surface current density $\sigma(\omega)\boldsymbol{E}_{\rm p}$,[24]

$$\boldsymbol{e}_r \times (\boldsymbol{E}_{\rm i} + \boldsymbol{E}_{\rm s} - \boldsymbol{E}_{\rm w}) = 0,$$

$$\boldsymbol{e}_r \times (\boldsymbol{H}_{\rm i} + \boldsymbol{H}_{\rm s} - \boldsymbol{H}_{\rm w}) = \sigma(\omega)\boldsymbol{E}_{\rm p}, \tag{4}$$

where $\boldsymbol{E}_{\rm p}$ is the tangential component of the total electric field, and $\boldsymbol{e}_r$ is the normal vector. By applying the boundary conditions, the expansion coefficients for the scattered electromagnetic field by the graphene coated particle are derived as[25]

$$a_l = -\frac{\tilde{n}\psi_l(\alpha_1)\psi_l'(\alpha_2) - \psi_l(\alpha_2)\psi_l'(\alpha_1) + (1/n_2)S(\omega)\cdot\psi_l'(\alpha_1)\psi_l'(\alpha_2)}{\tilde{n}\psi_l(\alpha_1)\xi_l'(\alpha_2) - \xi_l^{(1)}(\alpha_2)\psi_l'(\alpha_1) + (1/n_2)S(\omega)\cdot\psi_l'(\alpha_1)\xi_l'(\alpha_2)}, \tag{5}$$

$$b_l = -\frac{\tilde{n}\psi_l'(\alpha_1)\psi_l(\alpha_2) - \psi_l(\alpha_1)\psi_l'(\alpha_2) + (1/n_2)S(\omega)\cdot\psi_l(\alpha_1)\psi_l(\alpha_2)}{\tilde{n}\psi_l'(\alpha_1)\xi_l(\alpha_2) - \psi_l(\alpha_1)\xi_l^{(1)\prime}(\alpha_2) + (1/n_2)S(\omega)\cdot\psi_l(\alpha_1)\xi_l(\alpha_2)}, \tag{6}$$

where $\tilde{n} = n_1/n_2$, and n_1 is the refractive index of the particle. Also, the multipole expansion coefficients for the internal electromagnetic field inside the particle are obtained through the boundary conditions as

$$c_l = \tilde{n}\frac{\psi_l(\alpha_2)\xi_l'(\alpha_2) - \psi_l'(\alpha_2)\xi_l^{(1)}(\alpha_2)}{\tilde{n}\psi_l(\alpha_1)\xi_l'(\alpha_2) - \xi_l^{(1)}(\alpha_2)\psi_l'(\alpha_1) + (1/n_2)S(\omega)\cdot\psi_l'(\alpha_1)\xi_l'(\alpha_2)}, \tag{7}$$

$$d_l = \frac{\psi_l'(\alpha_2)\xi_l(\alpha_2) - \psi_l(\alpha_2)\xi_l^{(1)\prime}(\alpha_2)}{\tilde{n}\psi_l'(\alpha_1)\xi_l(\alpha_2) - \psi_l(\alpha_1)\xi_l^{(1)\prime}(\alpha_2) + (1/n_2)S(\omega)\cdot\psi_l(\alpha_1)\xi_l(\alpha_2)}, \tag{8}$$

where $\xi_l^{(1)} = \psi_l - {\rm i}\chi_l$, ψ_l and χ_l are the Riccati–Bessel functions.

The optical conductivity of the graphene layer $\sigma(\omega)$ can be derived from the Kubo formula.[26–28] For simplicity, we only consider the low-temperature conditions where the absolute value of Fermi level $E_{\rm F}$ of graphene is much larger than $k_{\rm B}T$, where $k_{\rm B}$ is the Boltzmann constant and T is the absolute temperature. The optical conductivity $\sigma(\omega)$ is the summation over the intraband $\sigma_{\rm intra}(\omega)$ and interband $\sigma_{\rm inter}(\omega)$ contributions: $\sigma(\omega) = \sigma_{\rm intra}(\omega) + \sigma_{\rm inter}(\omega)$. The Drude conductivity describes the intraband conductivity part

$$\sigma_{\rm intra}(\omega) = \sigma_0\frac{4E_{\rm F}}{\pi(\Gamma_{\rm c} - {\rm i}\hbar\omega)}, \tag{9}$$

and the interband optical conductivity is

$$\begin{aligned}\sigma_{\rm inter}(\omega) = \sigma_0\Bigg[&1 + \frac{1}{\pi}\arctan\left(\frac{\hbar\omega - 2E_{\rm F}}{\Gamma_{\rm c}}\right) \\ &- \frac{1}{\pi}\arctan\left(\frac{\hbar\omega + 2E_{\rm F}}{\Gamma_{\rm c}}\right) \\ &- \frac{\rm i}{2\pi}\ln\frac{(\hbar\omega + 2E_{\rm F})^2 + \Gamma_{\rm c}^2}{(\hbar\omega - 2E_{\rm F})^2 + \Gamma_{\rm c}^2}\Bigg],\end{aligned} \tag{10}$$

where $\sigma_0 = e^2/4\hbar$ is the universal conductivity of graphene, e is the electronic charge, $\hbar$ is the reduced Planck constant, and $\Gamma_{\rm c}$ is the damping constant. The optical conductivity is a function of the incident wave frequency, the Fermi level $E_{\rm F}$ and damping constant $\Gamma_{\rm c}$ of graphene. The dimensionless surface conductivity $S(\omega)$ of the particle can be modified by changing the optical conductivity of graphene $\sigma(\omega)$, which has the relationship $S(\omega) = {\rm i}\sigma(\omega)(\mu_0/\varepsilon_0)^{1/2}$, where μ_0 and ε_0 are the permeability and permittivity in vacuum, respectively.

The optical extinction, scattering, and absorption spectra of a nano-particle irradiated by an electromagnetic wave can be obtained by the Mie expansion coefficients

$$\begin{aligned}W_{\rm ext} &= {\rm Re}(\boldsymbol{E}_{\rm i}\times\boldsymbol{H}_{\rm s}^* + \boldsymbol{E}_{\rm s}\times\boldsymbol{H}_{\rm i}^*) \\ &= -\frac{c}{8\pi}k_{\rm ext}^2{\rm Re}\sum_{lm} l(l+1)\left[{\rm Re}(A_{lm}a_{lm}^* + B_{lm}b_{lm}^*)\right], \\ W_{\rm sca} &= {\rm Re}(\boldsymbol{E}_{\rm s}\times\boldsymbol{H}_{\rm s}^*) \\ &= \frac{c}{8\pi}k_{\rm ext}^2{\rm Re}\sum_{lm} l(l+1)\left[|a_{lm}|^2 + |b_{lm}|^2\right], \\ W_{\rm abs} &= W_{\rm ext} - W_{\rm sca} = -\frac{c}{8\pi}k_{\rm ext}^2{\rm Re}\sum_{lm} l(l+1) \\ &\quad\times\left[|a_{lm}|^2 + |b_{lm}|^2 + {\rm Re}(A_{lm}a_{lm}^* + B_{lm}b_{lm}^*)\right].\end{aligned} \tag{11}$$

The corresponding normalized optical cross section of a spherical particle can be derived as

$$\begin{aligned}&Q_{\rm ext} = W_{\rm ext}/(I_{\rm i}\cdot\sigma), \quad Q_{\rm sca} = W_{\rm sca}/(I_{\rm i}\cdot\sigma), \\ &Q_{\rm abs} = W_{\rm abs}/(I_{\rm i}\cdot\sigma),\end{aligned} \tag{12}$$

where $I_{\rm i}$ is the average intensity of light, $\sigma = \pi R^2$ is the geometric cross-section of the spherical particle. For the Gaussian beam, $I_{\rm i}$ is the field intensity at the center of the beam waist. As indicated, the optical spectra of nano particles contribute from different plasmonic modes categorized by the angular index l, such that the excitation of the multipolar localized surface plasmon should not only depend on the properties of the particle, but also on the beam profile and the wavefront.

3. Results and discussion

The schematic of scattering diagram is shown in Fig. 1. A graphene-coated polystyrene spherical particle is irradiated by

a focused Gaussian beam with waist radius $\omega_0 = 5$ μm. The ambient medium is air. The refractive index of the polystyrene particle is $n_1 = 1.59$. First we investigate the local surface plasmonic spectra for the graphene-coated dielectric particle excited by the Gaussian wave. As shown in Fig. 2(a), a resonant absorption peak is excited in the optical spectra due to the plasmonic response of the graphene layer, with hardly any scattering by the particle. The plasmonic resonant peak is mainly generated by the dipole mode resonance of the localized surface plasmon of the particle. We further analyze the impact of the particle size and the characteristics of the graphene coating on the plasmonic response of the particle.

As observed in Fig. 2(b), with the increase of the particle size, the frequency of the resonant peaks red shifts, while the magnitude of the peak remains almost unchanged. In Fig. 2(c), with the increase of the Fermi energy E_F of the graphene coating, which can be implemented through chemical doping, the corresponding wavelength of the resonant peak decreases gradually, simultaneously the magnitude of the peak is enhanced significantly. The mechanism of blueshifts of the resonant peaks with the particle size and Fermi energy of graphene can be understood from the surface plasmon excited in graphene by the Gaussian wave. With the particle size decreasing, the resonant wavelength becomes short, the corresponding resonant peak blueshifts. Furthermore, when the Fermi energy of the graphene coating increases, the localized surface plasmon is enhanced, the corresponding plasmonic resonant peak of the particle is improved and blueshifts. It demonstrates that both the intensity and resonant frequency of the plasmonic peak can be modified effectively by tuning the properties of the graphene layer, which provides functional localized surface plasmons manipulation for plasmonic devices.

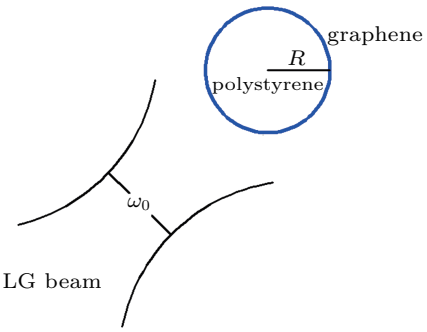


Fig. 1. Excitation of the localized surface plasmon on a graphene-coated polystyrene spherical particle with radius R by Gaussian beam.

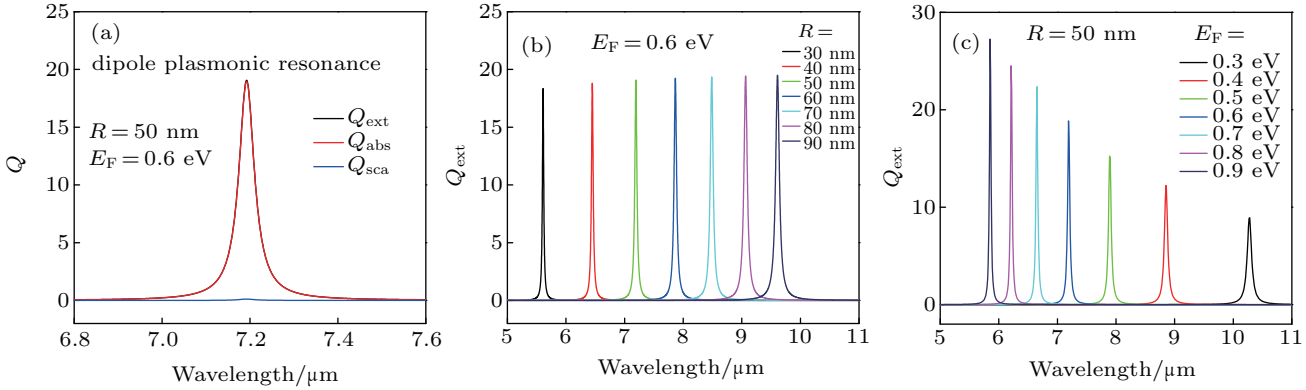


Fig. 2. (a) Local surface plasmonic spectra for a graphene-coated dielectric particle excited by a Gaussian wave (particle radius $R = 50$ nm, Fermi level $E_F = 0.6$ eV); (b) and (c) the optical extinction spectra (Q_{ext}) of the graphene-coated polystyrene particle with the variation of the particle size and the Fermi level of graphene, respectively.

However, as demonstrated above, the high order plasmonic mode cannot be excited through modifying the properties of the graphene coating. In view of this limitation, we proceed to explore the optical resonance of the graphene coated dielectric particle interact with beams with complex wavefront, the Laguerre Gaussian (LG) beam, to further investigate the high order plasmonic resonant mode response. The LG beam possesses the cylindrical symmetry and helical wavefront with a special vortex phase $\exp(\mathrm{i}s\varphi)$, where s is the azimuthal mode index.[29] Suppose the LG beam is polarized along x direction and propagating along z direction, the vector potential $\boldsymbol{A}$ can be expressed as

$$\boldsymbol{A} = \hat{e}_x u(r,\theta,z)\exp(\mathrm{i}kz),$$

$$u(r,\theta,z) = A_{ps}\left(\frac{w_0}{w_z}\right) f_{ps}(\rho)\exp\left(-\mathrm{i}\frac{kr^2}{2R(z)}\right) \times \exp(\mathrm{i}s\varphi)\exp\left[\mathrm{i}(2p+s+1)\zeta(z)\right], \quad (13)$$

where the parameters are related with the Rayleigh range $z_R = kw_0^2/2$,

$$w_z = w_0\left[1+\left(\frac{z}{z_R}\right)^2\right]^{1/2},$$
$$\zeta(z) = \tan^{-1}\left(\frac{z}{z_R}\right), \quad R(z) = \frac{z^2+z_R^2}{z},$$
$$f_{ps}(\rho) = \rho^s L_p^s(\rho^2)\exp(-\rho^2/2),$$
$$\rho = \frac{\sqrt{2}r}{w_z}, \quad A_{ps} = \frac{2}{w_0\sqrt{1+\delta_{0s}}}\sqrt{\frac{p!}{\pi(p+s)!}}, \quad (14)$$

and $L_p^s\left(\rho^2\right)$ is the generalized Laguerre polynomial. Through the Maxwell's equations

$$\begin{aligned}\boldsymbol{H} &= (1/\mu)\nabla\times\boldsymbol{A},\\ \boldsymbol{E} &= -\mathrm{i}\omega\left[\boldsymbol{A}+\left(1/k^2\right)\nabla(\nabla\cdot\boldsymbol{A})\right],\end{aligned}\tag{15}$$

the electromagnetic fields of the Laguerre Gaussian beam can be derived as[30]

$$\begin{aligned}\boldsymbol{H} &= \mathrm{i}\omega\sqrt{\frac{\varepsilon}{\mu}}A_{ps}\left(\frac{w_0}{w_z}\right)\exp\left(\mathrm{i}\frac{kr^2}{2R(z)}\right)\\ &\times\exp\left[\mathrm{i}\left(2p+l+1\right)\zeta(z)\right]\exp(\mathrm{i}kz)\\ &\times\left\{\boldsymbol{e}_y+\left\{\left[-\frac{r\cos\theta}{R(z)}f_{ps}(\rho)+\mathrm{i}\frac{\sqrt{2}}{kw_z}\cos\theta\frac{f_{ps}(\rho)}{\partial r}\right]\right.\right.\\ &\times\cos(s\theta)-\mathrm{i}s\frac{\sin\theta}{kr}\sin(s\theta)f_{ps}(\rho)\left.\left.\right\}\boldsymbol{e}_z\right\},\end{aligned}\tag{16}$$

$$\begin{aligned}\boldsymbol{E} &= \mathrm{i}\omega A_{ps}\left(\frac{w_0}{w_z}\right)\exp\left(\mathrm{i}\frac{kr^2}{2R(z)}\right)\\ &\times\exp\left[\mathrm{i}\left(2p+l+1\right)\zeta(z)\right]\exp(\mathrm{i}kz)\\ &\times\left\{\boldsymbol{e}_x+\left\{\left[-\frac{r\cos\theta}{R(z)}f_{ps}(\rho)+\mathrm{i}\frac{\sqrt{2}}{kw_z}\cos\theta\frac{f_{ps}(\rho)}{\partial r}\right]\right.\right.\\ &\times\cos(s\theta)+\mathrm{i}s\frac{\sin\theta}{kr}\sin(s\theta)f_{ps}(\rho)\left.\left.\right\}\boldsymbol{e}_z\right\}.\end{aligned}\tag{17}$$

Figures 3(a) and 3(b) show the modal profile of cross section $f_{ps}(\rho)$ of an LG beam of order $p=5$, $s=5$. As the figure shows, the cross section of the high order LG beam exhibits the cylindrical symmetry and a complex helical wavefront.

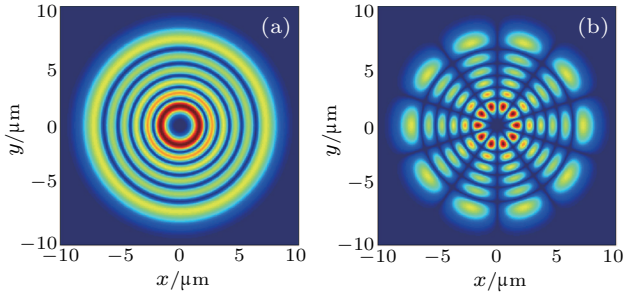


Fig. 3. Modal profile of cross section $f_{ps}(\rho)$ of LG beam of order $p=5$, $s=5$: (a) $\exp(is\theta)$, (b) $\cos(s\theta)$. Beam waist $\omega_0=5$ μm.

The optical extinction spectra of a graphene coated polystyrene particle excited by the LG electromagnetic wave described by Eqs. (16) and (17) with order $p=5$ and vortex phase $s=0$–5 are shown in Fig. 4. The color lines in the figure represent the extinction spectra contributed by all modes and discrete plasmon modes of dipole, quadrupole, octupole, $l=4$, $l=5$, respectively. As the figure indicates, with the vortex phase of the LG beam increasing, the lower order localized surface plasmonic modes diminish gradually, while the higher order localized surface plasmons are excited in the high frequency band. Specifically, the dipole mode accounts for the major part of plasmonic resonance for the lower order LG beam (LG_{50}, LG_{51}). With the helical phase s growing, the dipole mode diminishes gradually and the octupole mode emerges (LG_{52}) and dominates (LG_{53}). With the helical phase further increasing, the high order mode of $l=5$ is excited (LG_{54}) and dominates (LG_{55}), the resonant frequency blue shifts gradually.

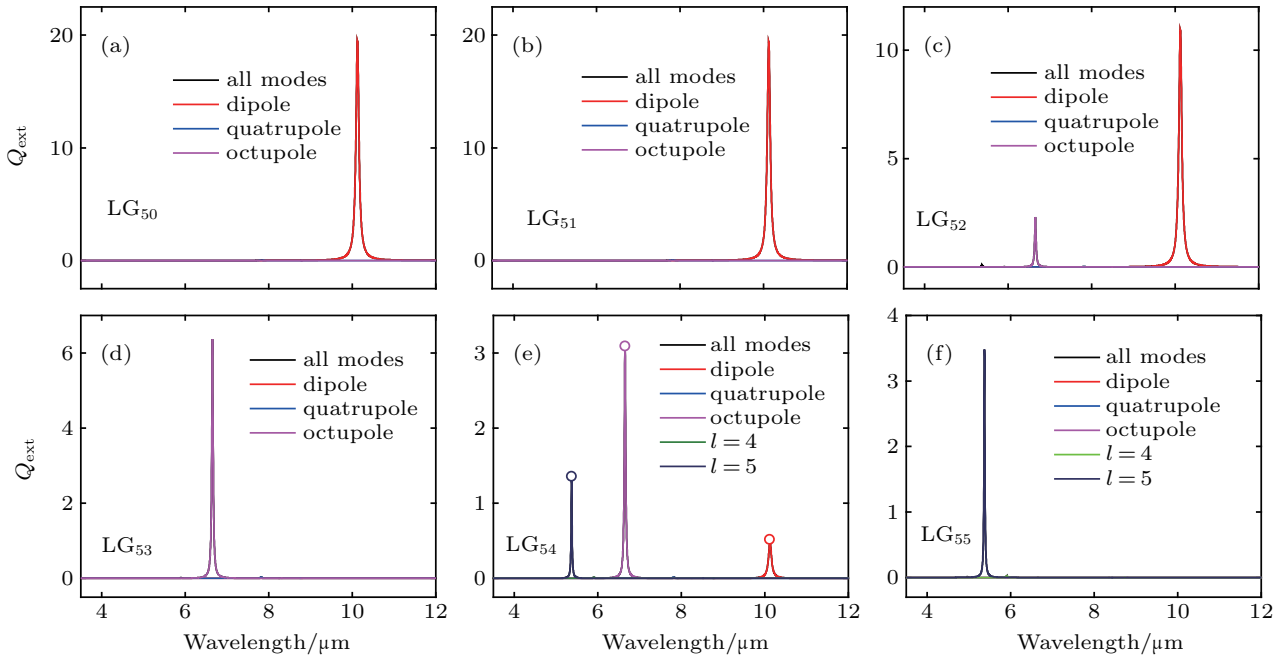


Fig. 4. Optical extinction spectra Q_{ext} of a graphene coated dielectric particle excited by LG beam of order $p=5$ with vortex phase factor $s=0$–5. The color lines represent the extinction spectra contributed by all modes and the discrete plasmonic modes of dipole, quadrupole, octupole, $l=4$, $l=5$, respectively. $R=100$ nm, $E_{\mathrm{F}}=0.6$ eV.

The investigation above demonstrates that the multi-polar localized surface plasmons of the graphene coated dielectric particle could be excited and tuned selectively by modifying the helical wavefront of the LG beam. The underlying physics can be understood from the mutual interaction of the particle and the helical wavefront of the high order LG beam which carries angular orbital momentum (OAM) of $s\hbar$ per photon.[29] The OAMs of the high order LG beam are transferred to the graphene coated dielectric particle under the resonant absorbing condition, exciting the corresponding mode of localized surface plasmons.[31,32] The selective multi-polar surface plasmons excitation through the wavefront engineering of optical beams could open a new path for the study on enhancing light–matter interaction.[33]

In further study, we illustrate the profile of multipolar mode of the electric field of the localized surface plasmon of the graphene coated dielectric particle excited by the LG beam in Fig. 5. As illustrated in the figure, by the different resonant frequency excitation, the field distribution around the particle exhibits the corresponding multipolar plasmonic profile, which further indicates the selective excitation of the high order localized surface plasmons by the LG beam.

Besides the local field distribution, we also examine the electric charge distribution of the graphene coated dielectric particle excited by the LG beam, since the charge pattern can offer a clear picture of the nature of a particular plasmonic wave mode at resonance within the particle.[34,35] In particular, the graphene coating is considered as a surface current surround the particle. The polarized electric charge distribution of the graphene coated dielectric particle excited by the LG beam for different plasmonic mode is shown in Fig. 6, corresponding to the multipolar mode of the localized surface plasmon in Fig. 5, respectively. It is shown that the charge density is mainly distributed on the surface of the particle, which is due to the surface plasmonic resonance of the graphene coating, and the charge density distributions inside the dielectric particle exhibit the exact localized surface plasmonic resonant mode patterns that correspond to the local field in Fig. 5. The analysis further demonstrates the effective excitation of the high order localized surface plasmons of the graphene coated dielectric particle by the LG beam.

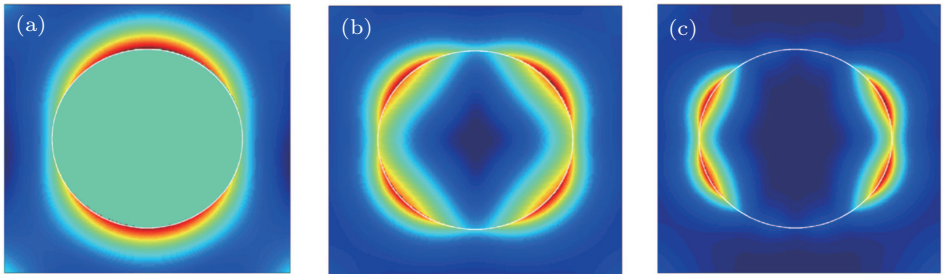


Fig. 5. Multipolar mode profile of electric field of the localized surface plasmon of the graphene coated dielectric particle excited by LG beam, panels (a)–(c) correspond to the red, magenta, and navy blue circles in Fig. 4(e), representing different plasmonic modes, respectively.

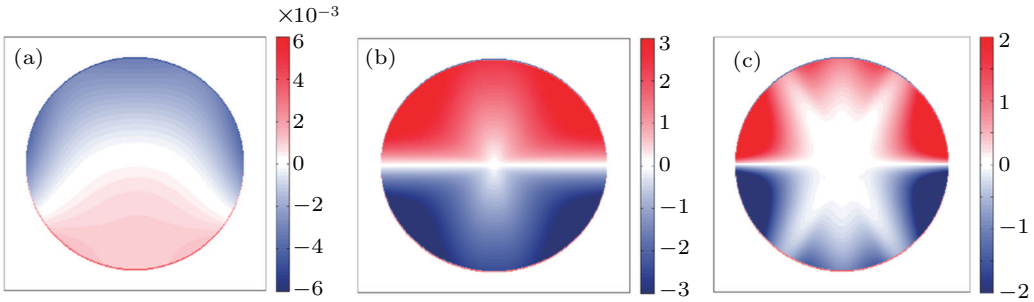


Fig. 6. Electric charge distribution of the graphene coated dielectric particle excited by LG beam for different localized plasmonic mode, corresponding to the multipolar mode of the localized surface plasmon in Fig. 5.

4. Conclusion and perspectives

We have made a systematic investigation on the excitation of localized surface plasmon of the graphene coated dielectric particle. It is found that both the intensity and resonant wavelength of the plasmonic peak can be tuned effectively through modifying the graphene layer. Furthermore, high order localized plasmons could be excited and tuned selectively by an LG beam, which is induced by the mutual interaction of the particle with the helical wavefront. Simultaneously, the multipolar mode profile of the localized surface plasmon is illustrated in detail. These investigations can find rich potential applications in the plasmonic devices and the wavefront engineering of optical beams for enhancing the light–matter interaction.

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