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Geometrical Parameter Effect on Plasmonic Scattering of Bimetallic Three-Layered Nanoshells

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Abstract: Enhanced scattering from local surface plasmon resonance by light has attracted much attention due to its special applications in sensor, cell, and biological imaging. Here, we investigate the ratio of scattering to absorption in bimetallic three-layered nanoshells with different geometrical parameters using quasi-static theory. We show that the ratio of scattering to absorption strongly depends on the inner radius, shell thickness, middle dielectric function, and surrounding medium function. To gain insight into the effect of such geometrical parameters on the plasmonic scattering, we also provide a comparison between silver–dielectric–gold nanoshells and gold–dielectric–silver nanoshells. This work provides an alternative approach to analyze the optical properties of bimetallic three-layered nanoshells with potential applications in sensors and photo-detectors.

Keywords: bimetallic three-layered nanoshells; scattering cross section; scattering efficiency; quasi-static theory

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1. Introduction

Due to their unique optical and optoelectrical properties, the local surface plasmon resonance (LSPR) of noble metal nanostructure induced by the collective motion of conduction electrons has attracted widespread attention from surface-enhanced Raman scattering (SERS) [1,2], sensor [3,4], cell, and biological imaging [5,6]. Different metallic nanostructures are designed theoretically and experimentally to achieve wavelength matching for specific applications [7–11]. LSPR can be well tuned from the ultraviolet to the infrared region by adjusting its geometrical parameter and surrounding medium function [12,13]. Unlike solid nanoparticles, noble nanoshells exhibit tunable plasmon resonance with minimal absorption in the near-infrared region by adjusting its geometrical parameters. Plasmon hybridization theory can explain the physical mechanism of nanoshells well [14,15], and the interaction between the plasmon responses of a solid sphere and a cavity will produce two new plasmon modes (the higher energy and the lower energy). Xia et al. have synthesized concentric nanoshells with a diameter of 50 nm, containing a gold core, a tunable silicon dielectric layer, and an outer gold nanoshell [16]. Theoretically, Khosravi et al. have studied the light scattering from two alternating concentric double silica-gold nanoshells by using quasi-static theory. The result has demonstrated that the intensity and position of the scattering peaks depend on shell thicknesses [17]. Shirzaditabar et al. have studied geometrical parameters' effect on the local electric field of silver–dielectric–silver multi-layer nanoshell; it is found that the resonance wavelength exhibits a red-shift by increasing the silver core radius [18,19]. Campbell and Ziolkowski have designed concentric double-negative metamaterial spheres. The results show that absorption can be overcome, and new optical properties can be observed in coated shell particles. In addition,

the coated nanoparticle can be applied in the design of highly subwavelength optical amplifiers [20–23]. Zouros and Tsakmakidis have presented a high permittivity semiconductor structure capable of breaking the scattering efficiency's fundamental single-channel limit and demonstrated the feasibility of obtaining magnetically-tunable directionality inversion in spherical microresonators (THz antennas) coated by magneto-optical materials [24,25].

Although both the scattering and absorption result from LSPR, the intensity of light scattering is usually less than that of light absorption. Thus, improving the ratio of scattering to absorption is an essential research topic. To extract the contribution of scattering from LSPR, Daneshfar et al. have studied the effect of various plasmonic materials on the scattering-to-absorption ratio of metal-dielectric two-layered nanoshell with quasi-static theory. The result shows that the ratio of scattering to absorption strongly depends on the shape and different materials [26]. Using Mie's theory, Bansal et al. have examined the absorption and scattering of semiconductor-coated nanoshells. They found that absorption and scattering were blue-shifting with increasing core radius, and red-shattering with increasing surrounding medium function [27]. A recent study on the scattering of $Ag_{1-x} - Cu_x$, $Ag_{1-x} - Au_x$ and $Au_{1-x} - Cu_x$ alloy nanoshell has demonstrated that the scattering enhancement can be controlled and tuned by adjusting the composition and surrounding medium [28]. Using quasi-static theory, Zhu et al. have found that the maximum effective scattering occurs at a thick thickness of nanoshell due to the plasmon mode transformation [29].

In this work, we theoretically examine the geometrical parameters' effect on plasmonic scattering of bimetallic three-layered nanoshells. We show that the ratio of absorption to scattering depends on its geometrical parameters and surrounding medium function. Furthermore, our simulations demonstrate that the ratio of scattering to absorption can be improved by increasing the inner core radius to a suitable wavelength and will be reduced by increasing the thickness of the middle dielectric layer and middle dielectric function. This work has the potential applications of bimetallic three-layered nanoshells in SERS, biological tissues, cells, and sensors.

2. Calculation Model and Theoretical Method

The geometry of bimetallic three-layered nanoshells is shown in Figure 1, in which the inner nanosphere has a radius r_1 and a complex dielectric function ε_1 , the outer nanoshell has a thickness of $r_3 - r_2$ and a complex dielectric function ε_3 , the inner nanosphere and outer nanoshell are separated by a dielectric layer with a thickness of $r_2 - r_1$ and a dielectric function ε_2 , and the total geometry is suspended in surrounding medium function ε_4 . The total diameter of the nanoshell is much smaller than the incident wavelength. Thus, the quasi-static theory can be used to calculate the scattering and absorption efficiency [29,30], and the electric potential that satisfies Laplace's equation reads

$$\nabla^2 \varphi = 0. \quad (1)$$

In the spherical coordinate system, the solution of Equation (1) with the electric potential $\varphi_i(r, \theta)$ in each region can be given by [31]

$$\varphi_i(r, \theta) = (A_i r + B_i r^{-2}) \cos \theta, (i = 1, \dots, 4) \quad (2)$$

where r and θ correspond to the radial distance and the polar angle, respectively, and the coefficients A_i and B_i are determined by satisfying the following electromagnetic field boundary conditions. That is, the electric potential should be continued at the adjacent surface

$$\varphi_i(r, \theta)|_{r=r_i} = \varphi_{i+1}(r, \theta)|_{r=r_i}, \quad (3)$$

and the normal derivative of electric potential at the surface should satisfy

$$\varepsilon_i \frac{\partial \varphi_i(r, \theta)}{\partial r} \Big|_{r=r_i} = \varepsilon_{i+1} \frac{\partial \varphi_{i+1}(r, \theta)}{\partial r} \Big|_{r=r_i} \quad (4)$$

because of the continuity of the normal component of the displacement field at the boundary.

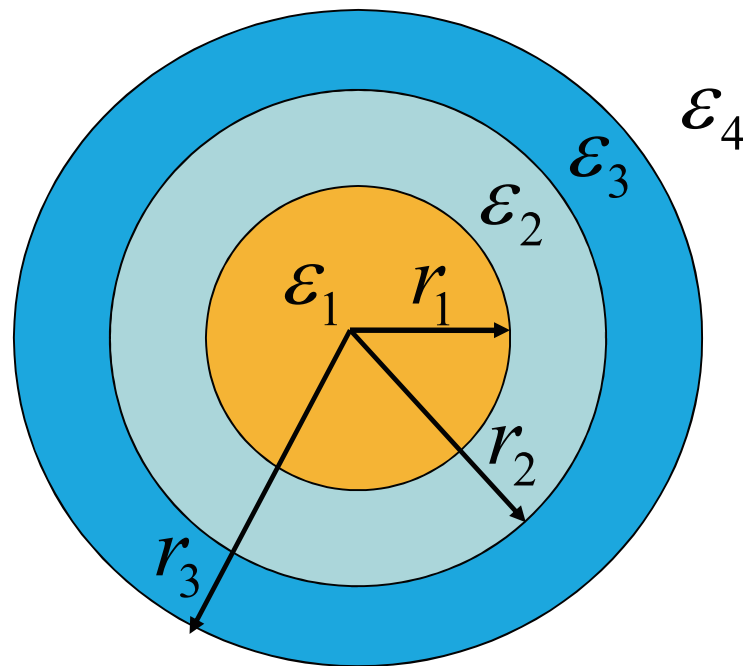


Figure 1. The geometry of bimetallic three-layered nanoshell. r_1 , r_2 , and r_3 are the radii of inner core, middle dielectric layer, and outer nanoshell. ϵ_1 , ϵ_2 , ϵ_3 , and ϵ_4 are the dielectric functions for the inner core, dielectric layer, outer nanoshell, and surrounding medium, respectively.

Considering the natural boundary conditions, in the inner core region $i = 1$, $B_1 = 0$. In the surrounding medium region $i = 4$, far from the nanoshell, the electric potential can be expressed as $\varphi_4 = -E_0 r \cos \theta$, where the magnitude of the incident electric field $E_0 = -A_4$. The other coefficients A_1, A_2, A_3 and B_2, B_3, B_4 can be derived from Equations (3) and (4). By using the gradient of the electric potential, the electric field intensity E in each region can be obtained

$$E_i(r, \theta) = -\nabla \varphi_i(r, \theta) = -(A_i - 2B_i r^{-3}) \cos \theta \vec{e}_r + (A_i + B_i r^{-3}) \sin \theta \vec{e}_\theta, (i = 1, \dots, 4). \quad (5)$$

Since the particle is much smaller than wavelength, the induced field in the region outside the shell is the same as a dipole with an effective dipole moment P ($P = \epsilon_4 \alpha E_4$), where α is the polarizability. As a result, the scattering and absorption cross section can be derived by using optical scattering theory [32]

$$C_{sca} = \frac{k^4 |\alpha|^2}{6\pi \epsilon_0^2} \quad (6)$$

$$C_{abs} = k \text{Im}(\alpha) \quad (7)$$

where k is the wave number, ϵ_0 is the permittivity of free space, and Im is imaginary part. In this paper, the ratio of scattering to absorption (scattering efficiency) is defined as $\frac{C_{sca}}{C_{abs}}$.

The complex dielectric functions of gold and silver have real and imaginary frequency-dependent components. In the Drude model [33], the complex dielectric function reads

$$\epsilon(\omega) = \epsilon_\infty - \frac{\omega_p^2}{\omega^2 + i\omega\gamma} \quad (8)$$

where ϵ_∞ is the value of the dielectric constant at high frequency, ω_p is the bulk plasmon frequency, ω is the angular frequency of the incident field, and γ is the damping constant

from the scattering of noble metal electrons. In this calculation, the values of parameters for silver are $\gamma_{Ag} = 3.2258 \times 10^{13} \text{ s}^{-1}$, $\epsilon_{\infty}^{Ag} = 4.039$, and $\omega_p^{Ag} = 9.1721 \text{ eV}$, and for gold $\gamma_{Au} = 1.07 \times 10^{14} \text{ s}^{-1}$, $\epsilon_{\infty}^{Au} = 8.7499$, and $\omega_p^{Au} = 9.0146 \text{ eV}$.

3. Numerical Results and Discussion

Figure 2 shows the spectra of light absorption, scattering, and scattering efficiency as a function of wavelength, where the geometric parameters are $[r_1, r_2, r_3] = [10 \text{ nm}, 25 \text{ nm}, 30 \text{ nm}]$ and $[\epsilon_2, \epsilon_4] = [2.5, 1.77]$. By examining the spectra of silver–dielectric–silver three-layered nanoshell as a function of wavelength in Figure 2a, we can see that three peaks $|\omega_{-}^{-}\rangle$, $|\omega_{-}^{+}\rangle$ and $|\omega_{+}^{-}\rangle$ appear in the spectra, caused by the plasmon hybridization [14,15,17,18]. Experimentally, the two peaks at longer wavelengths $|\omega_{-}^{-}\rangle$ and $|\omega_{-}^{+}\rangle$ can be easily detected. We can also see two longer absorption wavelength peaks at $\lambda = 648 \text{ nm}$ and $\lambda = 412 \text{ nm}$. The longest wavelength $\lambda = 648 \text{ nm}$, denoted as $|\omega_{-}^{-}\rangle$, corresponds to an anti-symmetric coupling between the solid inner sphere and outer bonding nanoshell. The peak at the wavelength $\lambda = 412 \text{ nm}$, denoted as $|\omega_{-}^{+}\rangle$, is caused by a symmetric coupling between the solid inner sphere and outer bonding nanoshell. The corresponding scattering peaks are located at $\lambda = 650 \text{ nm}$ ($|\omega_{-}^{-}\rangle$) and $\lambda = 412 \text{ nm}$ ($|\omega_{-}^{+}\rangle$). Unlike the intensity of absorption, the intensity of scattering intensity is much weaker than that of absorption. Therefore, the scattering efficiency as a function of wavelength in Figure 2a corresponds to the relative scattering intensity between scattering and absorption. Three peaks appear in the optical spectrum, for which two main peaks with a larger ratio of scattering efficiency are located at $\lambda = 402 \text{ nm}$ and $\lambda = 660 \text{ nm}$. This implies that the underlying mechanism results in the peak of scattering efficiency differing from the plasmon resonance wavelength of absorption and scattering peaks. In addition, the scattering-efficiency peaks are wider than the absorption and scattering peaks. Experimentally, it is crucial to improve the scattering efficiency for cell and biomedical imaging applications with minimized absorption by adjusting its geometric parameters. Figure 2b shows the spectra of the silver–dielectric–gold three-layered nanoshell. Two main absorption peaks appear at about $\lambda = 414 \text{ nm}$ and $\lambda = 730 \text{ nm}$, which can be attributed to the plasmon hybridization between a solid silver sphere and a gold nanoshell, indicating that the LSPR could be modulated by adjusting the material composition. Compared with the silver–dielectric–silver nanoshell, the scattering efficiency peaks of the silver–dielectric–gold nanoshell appeared at $\lambda = 454 \text{ nm}$, and $\lambda = 814 \text{ nm}$ demonstrated that the scattering efficiency strongly depends on its material component. To help understand the effect of geometric parameters on the optical properties, Figure 2c,d show the spectra of gold–dielectric–gold and gold–dielectric–silver nanoshells. The spectra of the gold–dielectric–gold and gold–dielectric–silver nanoshells are similar to that of Figure 2a,b, where the scattering efficiency of the gold–dielectric–silver nanoshell is higher than that of the gold–dielectric–gold nanoshell. As a result, the scattering efficiency can be improved by adjusting the geometric parameters.

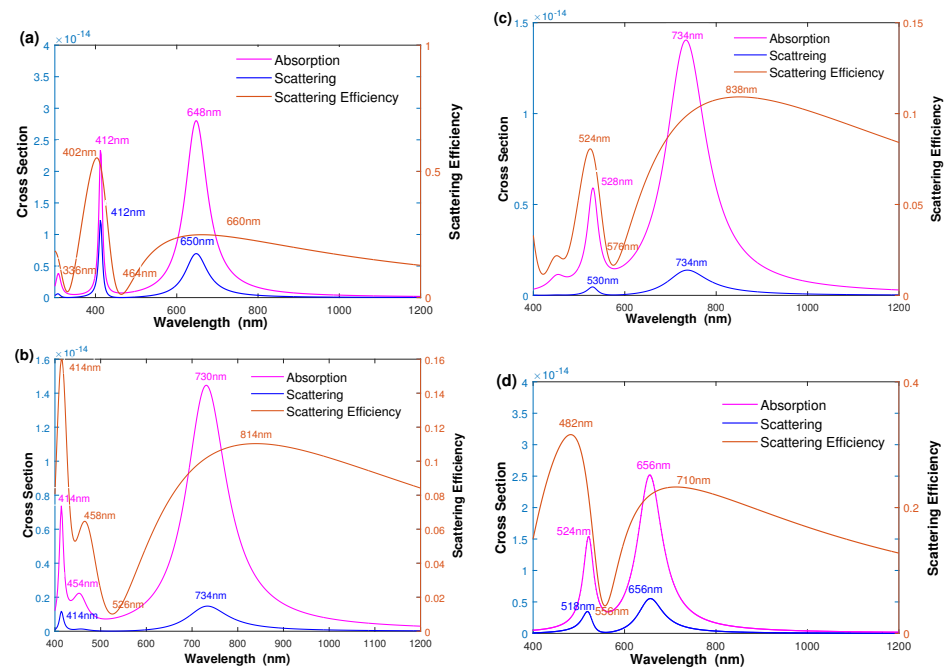


Figure 2. The spectra of absorption, scattering, and scattering efficiency are plotted as a function of wavelength. (a) Silver–dielectric–silver nanoshell, (b) silver–dielectric–gold nanoshell, (c) gold–dielectric–gold nanoshell, and (d) gold–dielectric–silver nanoshell.

3.1. The Effect of Inner Core Radius on Scattering Efficiency

Figure 3 shows the effect of inner radius r_1 on the scattering efficiency of silver–dielectric–gold, where the inner silver radius r_1 is increased from $r_1 = 4$ nm to $r_1 = 16$ nm with $[r_2, r_3] = [20$ nm, 25 nm], and $[\varepsilon_2, \varepsilon_4] = [2.0, 4.0]$ for Figure 3a, $[\varepsilon_2, \varepsilon_4] = [2.0, 2.0]$ for Figure 3b, and $[\varepsilon_2, \varepsilon_4] = [4.0, 2.0]$ for Figure 3c respectively. The results show that the scattering efficiency of silver–dielectric–gold strongly depends on the inner core radius r_1 , ε_2 and ε_4 . As seen from Figure 3a, the intensity of each scattering-efficiency peak becomes intense and then reduces quickly with increasing the inner radius r_1 . The intensity of the scattering-efficiency peak reduces first and reaches the minimum when the inner core radius $r_1 = 10$ nm; it then becomes intense quickly as the inner radius r_1 increases. The spectral width of the scattering efficiency is wider as the inner core radius increases, where the peak position takes place at a red-shift [11,14,18]. The inset of Figure 3a shows the effect of inner core radius r_1 on the scattering cross section, plotted as the function of wavelength; the scattering section of $|\omega_{-}^{+}\rangle$ mode takes place a red-shift, and its intensity reduces with increasing inner core radius r_1 . Figure 3b,c show the influence of the middle dielectric function ε_2 and surrounding dielectric function ε_4 on scattering efficiency, where $[\varepsilon_2, \varepsilon_4] = [2.0, 2.0]$ in Figure 3b, and $[\varepsilon_2, \varepsilon_4] = [4.0, 2.0]$ in Figure 3c. Figure 3b,c show that the scattering efficiency strongly depends on the middle dielectric function ε_2 and surrounding dielectric function ε_4 . The intensity becomes intense, and the spectral width becomes wider as the ratio of $\varepsilon_2/\varepsilon_4$ increases. In addition, the spectra of gold–dielectric–silver nanoshell are also shown in Figure 3d–f. Compared with Figure 3a–c, the scattering efficiencies of gold–dielectric–silver nanoshells are similar to those of silver–dielectric–gold nanoshells. The scattering efficiency strongly depends on the inner core radius r_1 and the ratio of $\varepsilon_2/\varepsilon_4$, which becomes intense, becomes wider, and has a red-shift with increasing r_1 and $\varepsilon_2/\varepsilon_4$. The scattering efficiency of gold–dielectric–silver nanoshell is higher than that of silver–dielectric–gold nanoshell under the same geometric parameter.

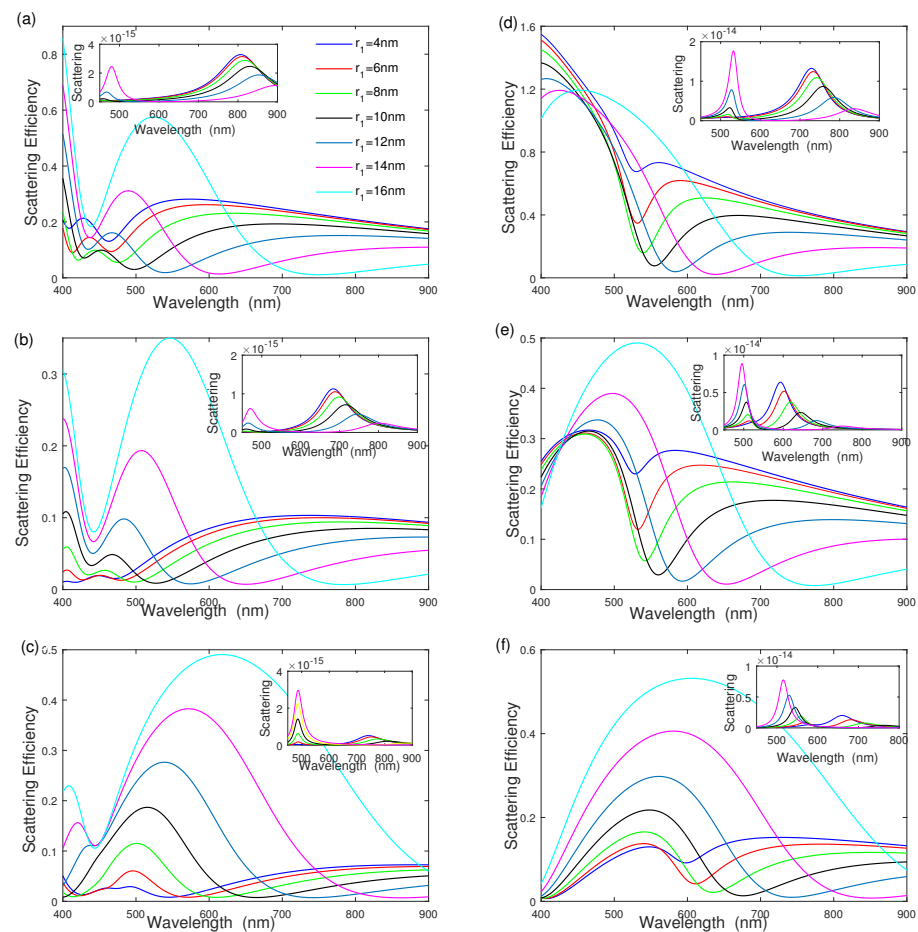


Figure 3. The effect of inner core radius r_1 on scattering efficiency and scattering. (a–c) Silver–dielectric–gold nanoshell with $[\varepsilon_2, \varepsilon_4] = [4.0, 2.0]$, $[\varepsilon_2, \varepsilon_4] = [2.0, 2.0]$, and $[\varepsilon_2, \varepsilon_4] = [2.0, 4.0]$; (d–f) gold–dielectric–silver nanoshell with $[\varepsilon_2, \varepsilon_4] = [4.0, 2.0]$, $[\varepsilon_2, \varepsilon_4] = [2.0, 2.0]$, and $[\varepsilon_2, \varepsilon_4] = [2.0, 4.0]$, respectively.

3.2. The Effect of Separated Dielectric Layer Thickness on Scattering Efficiency

To fully understand the influence of geometrical parameters on the scattering efficiency, we examine the effect of separated dielectric layer thickness $r_2 - r_1$ on the scattering efficiency of silver–dielectric–gold nanoshells and gold–dielectric–silver nanoshells with different middle dielectric radius r_2 in Figure 4. The middle dielectric layer radius r_2 is increased from $r_2 = 7$ nm to $r_2 = 16$ nm, whereas the radii of the inner core and outer shell are fixed at $r_1 = 5$ nm and $r_3 = 25$ nm. Figure 4a shows that the intensity of scattering efficiency decreases quickly as the middle dielectric wall thickness increases from $r_2 - r_1 = 2$ nm to $r_2 - r_1 = 11$ nm with $[\varepsilon_2, \varepsilon_4] = [4.0, 2.0]$. The scattering-efficiency intensity remains at a constant when the middle dielectric thickness is large enough ($r_2 - r_1 = 10$ nm). The scattering efficiency exhibits a blue shift with the increasing thickness of the middle dielectric layer. The inset of Figure 4a shows the scattering cross section has a blue-shift with increasing the thickness of $r_2 - r_1$ due to plasmon hybridization [14,29]. The corresponding physical phenomenon implies that the thickness of the outer nanoshell reduces by increasing the middle dielectric layer radius. The influences of the middle dielectric function and surrounding medium function on scattering are also exhibited in Figure 4b,c with $[\varepsilon_2, \varepsilon_4] = [2.0, 2.0]$ and $[\varepsilon_2, \varepsilon_4] = [4.0, 2.0]$ respectively, the same as that in Figure 3a–c. In addition, the effects of middle dielectric thickness on the gold–dielectric–silver nanoshell are also given in Figure 4d–f, showing similar features to that in Figure 4a–c.

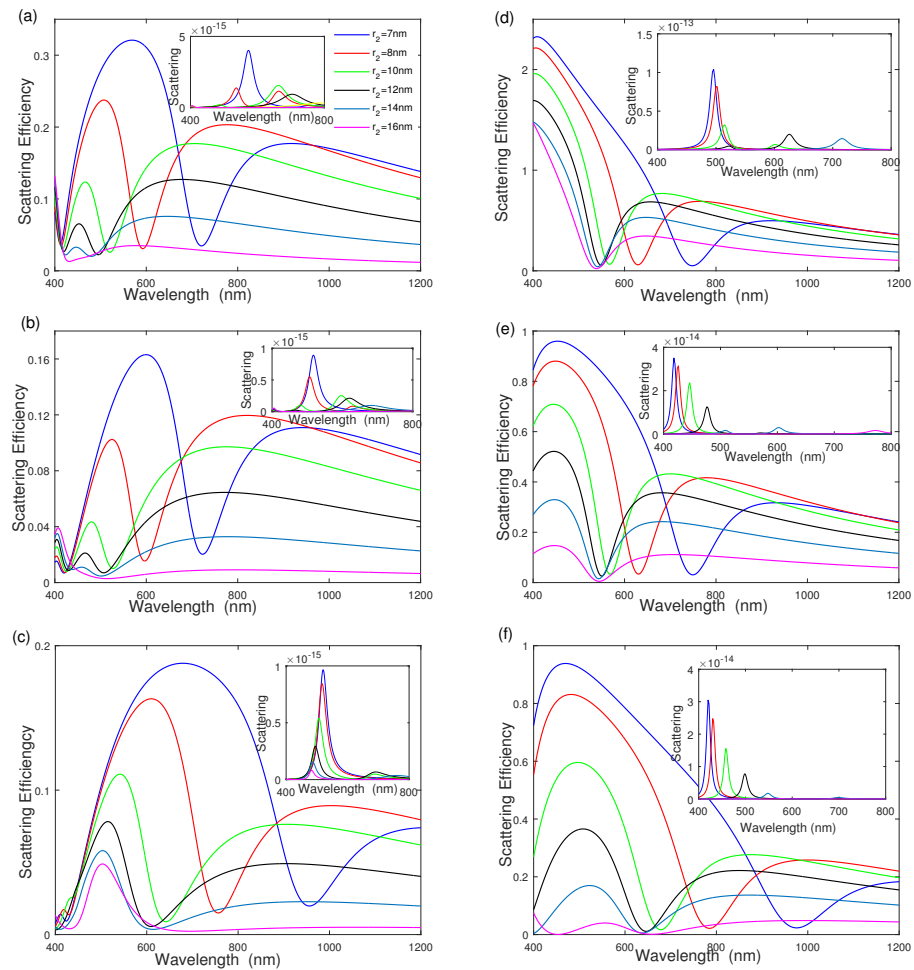


Figure 4. The effect of middle dielectric layer thickness $r_2 - r_1$ on scattering efficiency and scattering. (a–c) Silver–dielectric–gold nanoshell with $[\varepsilon_2, \varepsilon_4] = [4.0, 2.0]$, $[\varepsilon_2, \varepsilon_4] = [2.0, 2.0]$, and $[\varepsilon_2, \varepsilon_4] = [2.0, 4.0]$. (d–f) Gold–dielectric–silver nanoshell with $[\varepsilon_2, \varepsilon_4] = [4.0, 2.0]$, $[\varepsilon_2, \varepsilon_4] = [2.0, 2.0]$, and $[\varepsilon_2, \varepsilon_4] = [2.0, 4.0]$, respectively.

3.3. The Effect of Middle Dielectric Function on Scattering Efficiency

Since the dielectric function of the separated dielectric layer affects the electron oscillation, the scattering cross section and scattering efficiency depend on the dielectric function of the middle dielectric layer ε_2 . Figure 5 shows the influences of the middle dielectric function ε_2 on the scattering efficiency with geometric parameter $[r_1, r_2, r_3] = [6 \text{ nm}, 14 \text{ nm}, 20 \text{ nm}]$ and surrounding medium function $\varepsilon_4 = 4.0$, while varying the middle dielectric function ε_2 from $\varepsilon_2 = 1.0$ to $\varepsilon_2 = 6.0$. As seen from Figure 5a, the $|\omega_-^>$ mode has a redshift, and its intensity reduces quickly. $|\omega_+^>$ becomes larger quickly and then remains at a constant by increasing the middle dielectric layer ε_2 . According to the plasmon hybridization theory [14,28], the cavity plasmon is affected by the cavity dielectric function and is reduced to lower energy by increasing the dielectric function ε_2 . The coupling between the bonding shell plasmon mode and the inner core is reduced, in good agreement with the scattering cross-section in the inset of Figure 5a. Figure 5b examines the effect of ε_2 on the scattering efficiency of gold–dielectric–silver nanoshell. Both the intensity of $|\omega_-^>$ mode and $|\omega_+^>$ mode reduce quickly. The scattering-efficiency intensity of the gold–dielectric–silver nanoshell is much stronger than that of the silver–dielectric–gold nanoshell.

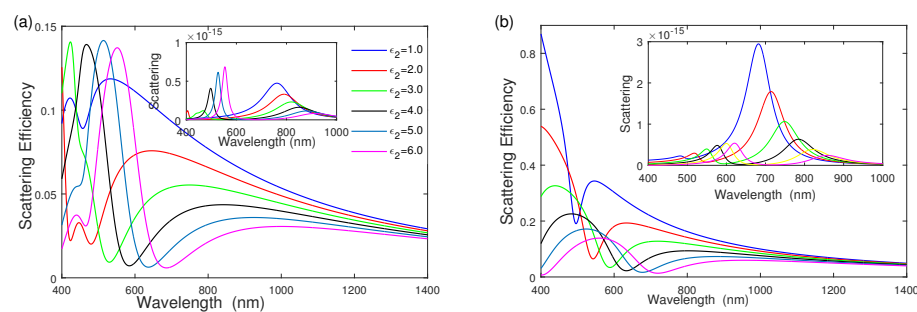


Figure 5. The effect of middle dielectric function ϵ_2 on scattering efficiency and scattering. (a) Silver-dielectric-gold nanoshell with $\epsilon_4 = 4.0$, (b) gold-dielectric-silver nanoshell with $\epsilon_4 = 4.0$, respectively.

3.4. The Effect of Surrounding Dielectric Function on Scattering Efficiency

The surrounding medium dielectric function ϵ_4 also influences the scattering efficiency of bimetallic three-layered nanoshells; we explore the effect of surrounding medium dielectric function ϵ_4 on the scattering efficiency of bimetallic three-layered nanoshells, as shown in Figure 6, where the geometric parameter $[r_1, r_2, r_3] = [5 \text{ nm}, 15 \text{ nm}, 20 \text{ nm}]$ and middle dielectric function ϵ_2 are fixed at $\epsilon_2 = 2.0$. Figure 6a shows that the scattering efficiency of silver-dielectric-gold nanoshell increases by increasing the surrounding medium function ϵ_4 from $\epsilon_4 = 1.0$ to $\epsilon_4 = 6.0$, where $|\omega^+ \rangle$ becomes intense quickly and $|\omega^- \rangle$ increases slowly, unlike that of Figure 5. The inset of scattering cross section is shown in Figure 6a, where $|\omega^- \rangle$ causes a distinct red-shift with increasing surrounding medium function ϵ_4 [15,19]. Due to the dielectric screening with a high refractive index, the induced charge on the outer gold nanoshell surface decreases, and then the induced charge on the inner surface of the silver core follows. Therefore, the electric repulsion force between the core and the outer nanoshell reduces. Thus, $|\omega^- \rangle$ mode exhibits a red shift due to the dielectric screening. Figure 6b also provides the scattering efficiency of the gold-dielectric-silver nanoshell, compared with Figure 6a. Figure 6b shows that scattering efficiency is similar to that in Figure 6a, where both $|\omega^+ \rangle$ and $|\omega^- \rangle$ become intense quickly. The scattering efficiency of the silver-dielectric-gold nanoshell is much stronger than that of the gold-dielectric-silver nanoshell.

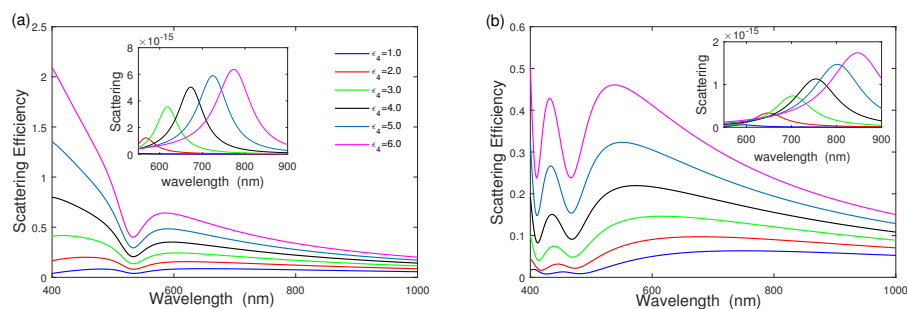


Figure 6. The effect of surrounding medium dielectric function ϵ_4 on scattering efficiency and scattering. (a) Silver-dielectric-gold nanoshell with $\epsilon_2 = 2.0$, (b) gold-dielectric-silver nanoshell with $\epsilon_2 = 2.0$, respectively.

3.5. The Local Electric Field Intensity Distribution of Gold–Dielectric–Silver Nanoshells

Finally, in order to find the effect of geometrical parameters on scattering efficiency of bimetallic three-layered nanoshells, we calculate the local electric field intensity distributions of gold-dielectric-silver three-layered nanoshells with different geometrical parameters, middle dielectric layer function ϵ_2 , and surrounding medium ϵ_4 , where the direction of the incident electric field is parallel to the x-axis. Figure 7a,b show local electric field intensity distributions of gold-dielectric-silver nanoshell with LSPR wavelength $\lambda = 444 \text{ nm}$ and $\lambda = 722 \text{ nm}$, respectively, where the geometrical parameters are

$[r_1, r_2, r_3] = [10 \text{ nm}, 15 \text{ nm}, 25 \text{ nm}]$ and $[\varepsilon_2, \varepsilon_4] = [4.0, 2.0]$. As shown in Figure 7a, we can see that the local electric field is concentrated in the inner gold core, while the local electric field intensity of silver nanoshell is very weaker at wavelength $\lambda = 444 \text{ nm}$ denotes as $|\omega^+_{-}\rangle$ mode. For $|\omega^+_{-}\rangle$ mode, the same kind of charges signed on the inner gold core and out of the silver nanoshell, which repulses in $|\omega^+_{-}\rangle$ mode. Figure 7b shows the local electric field focused in the middle dielectric layer, whereas the inner gold core and outer silver nanoshell are much weaker at wavelength $\lambda = 722 \text{ nm}$. There are different kinds of charges signed on the inner gold core and inner surface of the outer silver nanoshell, which attracts in $|\omega^+_{-}\rangle$ mode [15,19]. The scattering efficiency can be well improved with the larger inner core radius r_1 , smaller middle dielectric layer function ε_2 , and larger surrounding medium ε_4 . Figure 7c,d provide local electric field intensity distributions of gold–dielectric–silver nanoshell with $[r_1, r_2, r_3] = [15 \text{ nm}, 18 \text{ nm}, 30 \text{ nm}]$ and $[\varepsilon_2, \varepsilon_4] = [1.0, 5.0]$. Figure 7c,d show that the local electric field intensity is much stronger than that of Figure 7a,b; the decreasing middle dielectric layer leads to the surface induced charges increasing, and then the attraction leads to the local field intensity greatly increasing.

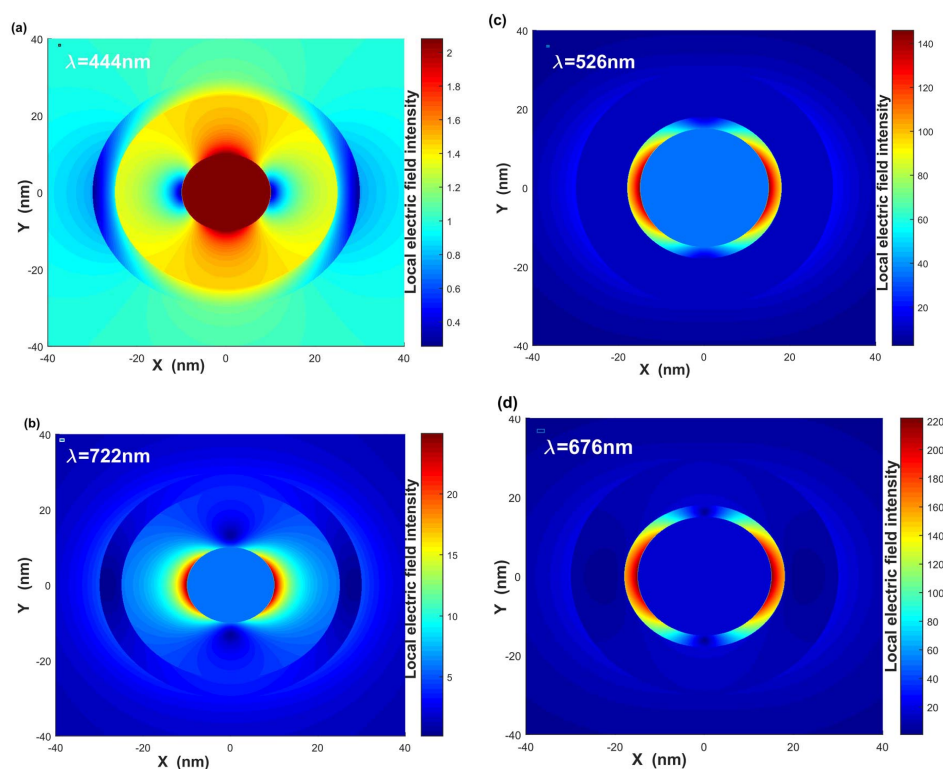


Figure 7. Local electric field intensity distributions of gold–dielectric–silver nanoshell with each LSPR wavelength. (a,b) $[r_1, r_2, r_3] = [10 \text{ nm}, 15 \text{ nm}, 20 \text{ nm}]$, $[\varepsilon_2, \varepsilon_4] = [4.0, 2.0]$, and the local surface plasmon resonance wavelengths are at $\lambda = 444 \text{ nm}$ and $\lambda = 722 \text{ nm}$, respectively. (c,d) $[r_1, r_2, r_3] = [15 \text{ nm}, 18 \text{ nm}, 30 \text{ nm}]$, $[\varepsilon_2, \varepsilon_4] = [1.0, 5.0]$, and the local surface plasmon resonance wavelengths are at $\lambda = 526 \text{ nm}$ and $\lambda = 676 \text{ nm}$.

4. Conclusions

In conclusion, we have examined how the geometrical parameters, including the inner radius, the middle dielectric layer thickness, the middle dielectric function, and the surrounding medium dielectric function, affect the scattering efficiency of three-layered nanoshells by using the quasi-static theory. Our results have demonstrated that the scattering efficiency strongly depends on those geometric parameters. The intensity of the scattering efficiency can be improved by reducing the middle layer dielectric function ε_2 and increasing the surrounding medium function ε_4 . As a result, a more robust scattering efficiency can be achieved by controlling the geometrical parameters and surrounding di-

electric function. Since the scattering cross section of the homogeneous spherical core-shell particles is calculated analytically, it opens a way to understand the underlying physics by comparing the results with that by using the Mie scattering theory [30,34].

Author Contributions: J.-B.H., Y.-L.C. and J.L. performed the numerical simulations and wrote the manuscript, which all of the authors discussed. Y.-W.M. and C.-C.S. reviewed and revised the manuscript. All authors have read and agreed to the published version of the manuscript.

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References

- Ding, S.J.; Ma, L.; Feng, J.R.; Chen, Y.L.; Yang, D.J.; Wang, Q.Q. Surface-roughness-adjustable Au nanorods with strong plasmon absorption and abundant hotspots for improved SERS and photothermal performances. *Nano Res.* **2022**, *15*, 2715–2721. [\[CrossRef\]](#)
- Li, Q.J.; Wang, J.G.; Ding, Q.J.; Chen, M.D.; Ma, F.C. Coupling effect on charge-transfer mechanism of surface-enhanced resonance Raman scattering. *J. Raman Spectrosc.* **2017**, *48*, 560–569. [\[CrossRef\]](#)
- Li, Q.; Shi, Z.H.; Wu, L.J.; Wei, H. Resonance scattering-enhanced photothermal microscopy. *Nanoscale* **2020**, *12*, 8397–8403. [\[CrossRef\]](#) [\[PubMed\]](#)
- Maier, S.A. *Plasmonics: Fundamentals and Application*; Springer: Berlin, Germany, 2007.
- Zhao, F.; Wang, X.; Zhang, Y.; Lu, X. In situ monitoring of silver adsorption on assembled gold nanorods by surface-enhanced Raman scattering application. *Nanotechnology* **2020**, *31*, 295601. [\[CrossRef\]](#) [\[PubMed\]](#)
- Zhu, J.; Li, X.; Li, J.J.; Zhao, J.W. Enlarge the biologic coating-induced absorbance enhancement of Au-Ag bimetallic nanoshells by tuning the metal composition. *Spectrochim. Acta-Part A* **2018**, *189*, 571–577. [\[CrossRef\]](#)
- Qi, Y.; Shu, C.C.; Dong, D.; Petersen, I.R.; Jacobs, K.; Gong, S.Q. Fast quantum state transfer in hybrid quantum dot-metal nanoparticle systems by shaping ultrafast laser pulses. *J. Phys. D Appl. Phys.* **2019**, *52*, 425101. [\[CrossRef\]](#)
- Hwang, Y.; Koo, D.J.; Ferhan, A.R.; Sut, T.N.; Yoon, B.K.; Cho, N.; Jackman, J.A. Optimizing Plasmonic Gold Nanorod Deposition on Glass Surfaces for High-Sensitivity Refractometric Biosensing. *Nanomaterials* **2022**, *12*, 3432. [\[CrossRef\]](#)
- Wu, Z.W.; Ma, Y.W.; Jiang, Y.Y.; Li, J.; Yin, X.C.; Zhang, L.H.; Yi, M.F. Theoretical study of optimizing the refractive index sensitivity of dielectric-metal nanotube by tuning the geometrical parameters. *Opt. Commun.* **2019**, *449*, 57–62. [\[CrossRef\]](#)
- Wang, X.H.; Zhang, C.Y.; Zhou, X.L.; Fu, Z.K.; Yan, L.; Li, J.P.; Zhang, Z.L.; Zheng, H.R. Plasmonic Effect of Ag/Au Composite Structures on the Material Transition. *Nanomaterials* **2022**, *12*, 2927. [\[CrossRef\]](#)
- Kang, H.S.; Long, M.Y.; Zhou, T.; Liu, J.C.; Duan, Y.T.; Ma, L.; Chen, X.B.; Ding, S.J.; Zhou, L. High-index facets and multidimensional hotspots in Au-decorated 24-faceted PbS for ultrasensitive and recyclable SERS substrates. *J. Mater. Chem. C* **2022**, *10*, 958–968. [\[CrossRef\]](#)
- Farooq, S.; Rativa, D.; Araujo, R.E. Near-Infrared Colloidal Gold Nanostructures. *Plasmonics* **2020**, *15*, 1507–1515.
- Ding, S.J.; Zhang, H.; Yang, D.J.; Qiu, Y.H.; Nan, F.; Yang, Z.J.; Wang, J.; Wang, Q.Q.; Lin, H.Q. Magnetic Plasmon-Enhanced Second-Harmonic Generation on Colloidal Gold Nanocups. *Nano Lett.* **2019**, *19*, 2005–2011. [\[CrossRef\]](#)
- Prodan, E.; Radloff, C.; Halas, N.J. A hybridization model for the plasmon response of complex nanostructures. *Science* **2003**, *302*, 419–422. [\[CrossRef\]](#)
- Prodan, E.; Nordlander, P. Plasmon hybridization in spherical nanoparticles. *J. Chem. Phys.* **2004**, *120*, 5444–5454. [\[CrossRef\]](#)
- Xia, X.H.; Liu, Y.; Ameer, G.A. Engineering sub-100 nm multi-layer nanoshells. *Nanotechnology* **2006**, *17*, 5435–5440. [\[CrossRef\]](#)
- Khosravi, H.; Daneshfar, N.; Bahari, A. Theoretical study of the light scattering from two alternating concentric double silica-gold nanoshell. *Phys. Plasmas* **2010**, *17*, 053302. [\[CrossRef\]](#)
- Shirzaditabar, F.; Saliminasab, M. Geometrical parameters effects on local electric field enhancement of silver-dielectric-silver multilayer nanoshell. *Phys. Plasmas* **2013**, *20*, 052109. [\[CrossRef\]](#)
- Shirzaditabar, F.; Saliminasab, M. Optimization of silver-dielectric-silver nanoshell for sensing applications. *Phys. Plasmas* **2013**, *20*, 668. [\[CrossRef\]](#)
- Campbell, S.D.; Ziolkowski, R.W. Impact of strong localization of the incident power density on the nano-amplifier characteristics of active coated nano-particles. *Opt. Commun.* **2012**, *285*, 3341–3352. [\[CrossRef\]](#)

21. Arslanagic, S.; Ziolkowski, R.W.; Breinbjerg, O. Analytical and numerical investigation of the radiation from concentric metamaterial spheres excited by an electric Hertzian dipole. *Radio Sci.* **2007**, *42*, RS6S16. [[CrossRef](#)]
22. Campbell, S.D.; Ziolkowski, R.W. Simultaneous Excitation of Electric and Magnetic Dipole Modes in a Resonant Core-Shell Particle at Infrared Frequencies to Achieve Minimal Backscattering. *IEEE J. Sel. Top. Quantum* **2013**, *19*, 4700209. [[CrossRef](#)]
23. Gordon, J.A.; Ziolkowski, R.W. The design and simulated performance of a coated nano-particle laser. *Opt. Express* **2007**, *15*, 2622–2653. [[CrossRef](#)]
24. Zouros, G.P.; Kolezas, G.D.; Almpanis, E.; Tsakmakidis, K.L. Three-Dimensional Giant Invisibility to Superscattering Enhancement Induced by Zeeman-Split Modes. *ACS Photonics* **2021**, *8*, 1407–1412. [[CrossRef](#)]
25. Zouros, G.P.; Kolezas, G.D.; Almpanis, E.; Baskourellos, K.; Stefański, T.P.; Tsakmakidis, K.L. Magnetic switching of Kerker scattering in spherical microresonators. *Nanophotonics* **2020**, *9*, 4033–4041. [[CrossRef](#)]
26. Daneshfar, N. The study of scattering-to-absorption ratio in plasmonic nanoparticles for photovoltaic cells and sensor applications. *Plasmonics* **2021**, *16*, 2017–2023. [[CrossRef](#)]
27. Bansal, A.; Verma, S.S. Tailoring LSPR-based absorption and scattering efficiencies of semiconductor-coated Au nanoshell. *Plasmonics* **2014**, *9*, 335–341.
28. Bansal, A.; Seckhon, J.S.; Verma, S.S. Scattering efficiencies and LSPR tunability of bimetallic Ag, Au and Cu nanoparticles. *Plasmonics* **2014**, *9*, 143–150. [[CrossRef](#)]
29. Zhu, J.; Zhao, S.M. Improve the effective resonance light scattering of three-layered plasmonic Au@Dielectric@Ag bimetallic nanoshells. *Plasmonics* **2022**, *17*, 173–180. [[CrossRef](#)]
30. Averitt, R.D.; Westcott, S.L.; Halas, N.J. Linear optical properties of gold nanoshells. *J. Opt. Soc. Am. B* **1999**, *16*, 1824–1832.
31. Jackson, J.D. *Classical Electrodynamics*, 3rd ed.; Wiley: Hoboken, NJ, USA, 1998. [[CrossRef](#)]
32. Bohren, C.F.; Huffman, D.R. *Absorption and Scattering of Light by Small Particles*; Wiley: New York, NY, USA, 2000.
33. Johnson, P.B.; Christy, R.W. Optical Constants of the Noble Metals. *Phys. Rev. B* **1972**, *12*, 4370–4379. [[CrossRef](#)]
34. Wu, Z.S.; Wang, Y.P. Electromagnetic scattering for multilayered sphere: Recursive algorithms. *Radio Sci.* **1991**, *26*, 1393–1401. [[CrossRef](#)]