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Research article

Effect of spacer on size dependent plasmonic properties of triple layered spherical core-shell nanostructure

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Abstract: In this paper, the plasmonic property of triple layered ZnO@M@Au (M = spacer) spherical core-shell nanostructures embedded in a dielectrics host medium is investigated by varying core size, spacer thickness, shell thickness and dielectrics function of the host medium within the framework of the qausi-static approximation method. The absorption coefficient of ZnO@M@Au spherical triple layered core-shell nanostructures is effectively studied by optimizing the parameters with range of nano-inclusion size mainly in between 18 and 23 nm. In this triple layered core-shell nanostructure two plasonic resonances are found associated with spacer@Au and Au@medium interfaces. The tunability of the plasmon resonances of the composite systems enables it to exhibit very interesting material properties in a variety of applications extending from near-UV to near-infrared spectral region.

Keywords: spacer; triple layered core-shell nanostructure; surface plasmon resonance; absorption coefficient; dielectrics function

1. Introduction

Based on the number of constituents, nano-materials can be classified as monolithic or composite. A monolithic material has a single constituent, while composite materials are made of two or more constituent materials having significantly different optical, plasmonic, catalytic, biological, physical, and chemical properties [1-3], that, when combined, produce a material with a characteristic different from the individual components. In a composite material, one of the constituents is a continuous matrix which is called a host matrix while the others dispersed in the host matrix are called inclusions or fillers.

Among the nano-inclusions, core-shell nanoparticles (CSNPs) combine from one core and one or more than one shell nanomaterials by using encapsulation process with different geometry and size to obtain a new material with combined and/or other unique properties neither shown by constituents [4–6]. This new or unique properties mainly originated from the interaction of plasmonic shell materials with the electromagnetic field which is greatly intensified by a phenomenon known as the surface plasmon resonance (SPR) and the interaction of plasmon of the metallic shell with plasmon/exciton/plariton of the inner material [7,8]. The plasmonic properties of the composite strongly depend on the geometry, size, composition and the dielectrics function (DF) of the host matrix [7,9,10]. Plasmonic properties and other functions such as catalytic, magnetic, and optical can be imparted to the inner materials due to coating material.

Recently, noble metal nanoparticles (NPs) (Like Ag, Au, Cu, Pt) have attracted the attention of the researchers due to their unique catalytic, electronic, plasmonic and optical properties [11] as well as high chemical stability, bio-affinity, strong absorption of light from visible to infrared (IR) spectral region [1,12,13], which are dominated by the localized surface plasmon resonance (SPR) [14]. In addition to these properties, the potential application of noble metal NPs are preferable as coating material. The coated nanoparticles are depending on the ability to control their size, shape, and the environment surrounding them.

Modifying or making new materials either by doping or making shell is the day to day activity of the researchers. Specially, core-shell nanoparticles (CSNPs) are considered to be extremely promising nanocomposite among hybrid nanostructures (alloy, doping and hetro-structure) in view of their greatly enhanced their properties and applications. As stated above, CSNPs have unique/new properties and such unique, useful and tailorable properties have also advanced CSNPs as a very important class of emerging nanocomposite for a wide range of applications, for instance, catalysis, biomedical, energy/data storage, solar cell, antibacterial, renewable energy, photonics, electronics [15-23]. With all these advantages, core-shell nanostructures (CSCNs) have been broadly investigated experimentally and/or theoretically [4,24,25] by various research groups in the past decade and applied to a wide variety of fields. CSNPs can be consist from metal, semiconductor, dielectrics or organic/inorgamnic materials one is a core and another or the same material is a shell [7,21,26,27]. Triple layered core-shell nanostructure (TLCSNS) is one of the core-shell nanoinclusion consists of three different materials. Due to the number of constituents, TLCSNSs have new/unique properties and further applications than two layered structures and also have been studied experimentally and/or theoretically [14,28,29]. However, in TLCSNS, semiconductor as a core, different materials as a spacer and noble metal as a shell (spacer can be dielectrics, semiconductor, and vacuum) hybrid arrangement is not further studied yet. In addition to this, the present investigation provides a very conceptual framework, including specific cases previously investigated. In such system, there is an interaction between exciton-exciton/polariton-plasmon in the nanoinclusion which is very interested for new properties and various applications.

In this paper, I studied the plasmonic properties of triple layered spherical ZnO@M@Au coreshell nanostructure embedded in active host matrix. The shell metal Au NP has been investigated most extensively because of its high catalytic, universal biocompatibility, optical sensitivity, facile preparation, resistance to oxidation, and surface plasmon resonance (SPR) band that can absorb and scatter visible light relative to other noble metals [30]. The plasmonic resonance of Au nanoshell tuned from visible to near-infrared spectral region. ZnO NP is wurtzite zinc oxide has wide band-gap (3.37 eV), high exciton binding energy (~60 meV) at room temperature (RT), and high

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dielectric constant [31–33], and it is reliable material for visible and near-UV applications [31,32]. ZnO NP has attractive extensive attention due to its potential application in laser diodes, solar cell, field emission displays, optoelectronics devices, gas sensor, photocatalysis, ultraviolet laser [34,35]. Due to this noble properties and applications of ZnO and Au NPs, core-shell combination of these materials is a desirable way to generate new/unique properties and enhanced applications. Indeed, ZnO@Au core-shell nanostructures have been investigated for various applications [30–37] and display improved properties. In the present work, to modify the properties as well as to enhance the applications of this combination, the third material is inserted between them.

In this theoretical work, mainly the absorption coefficient of noble metal coated triple layered core-shell NPs is effectively studied by optimizing the parameters: (i) the size of core ZnO NP, (ii) the size of composite, (iii) dielectrics function of host matrix, (iv) shell thickness, (v) spacer thickness, and (vi) dielectrics function of spacer. The spacer material includes all materials which have dielectrics function between 1 and 7 (including vacuum and water) and thickness of spacer ranging from 4 to 9 nm. For the small size nanocomposite, the quasistatic limit is an appropriate to study the plasmonic response and the dipolar mode resonance is more enhanced than the other higher order multipoles.

The paper is organized as follow: in section 2, I will discuss the basic idea of nanoinclusion with a representative model by using electrostatic approximation, followed by the derivation of formula which determine plasminic properties of nanoinclusion. In section 3, describes the plasmonic response of triple layered core-shell nanostructures: absorption coefficient by optimizing the parameters. Finally, the main result of the work is summarized in section 4.

2. Theoretical model and calculation

Consider triple layer core-shell nanoparticle consisting of a semiconductor core (ZnO) of dielectric function (DF) ε_c , spacer (DF) ε_d and a metallic shell (Au) of DF ε_s embedded in a non-absorptive host matrix having a real DF ε_h , as shown in Figure 1, i.e., cross-sectional view of the composite. The radius of core r_c , spacer r_d and shell r_s , respectively, in which the volume fraction spacer to spacer + core and shell to composite is $\beta_1 = 1 - (r_c/r_d)^3$ and $\beta_2 = 1 - (r_d/r_s)^3$, respectively. When the composite of triple layered CSNP is irradiated (placed in) with an electromagnetic radiation, an electric field is induced in the system due to polarization. The distribution of the electrostatic potential Φ associated with the induced field inside and outside of the NP can be obtained by solving the Laplace equation, $\nabla^2 \Phi = 0$ in spherical coordinates.



Figure 1. The array of triple layered spherical CSNSs embedded in the dielectrics host medium. The right figure is the resolution of nano-inclusion.

The distribution of the potentials has different expressions in four regions are take the form of the following equations from Eqs 1–4:

$$\phi_1 = A_1 r \cos\theta, \ r \le r_c \tag{1}$$

$$\phi_2 = \left(A_2 r + \frac{B_1}{r^2}\right) \cos\theta, \ r_c \le r \le r_d \tag{2}$$

$$\phi_3 = \left(A_3 r + \frac{B_2}{r^2}\right) \cos\theta, \ r_d \le r \le r_s \tag{3}$$

$$\phi_4 = \left(-E_0 r + \frac{B_3}{r^2}\right) \cos\theta, \ r \ge r_s \tag{4}$$

The above potentials ϕ_1 , ϕ_2 , ϕ_3 and ϕ_4 are calculated from inside the core, spacer, metallic shell, and host matrix, respectively. The parameters E_0 is applied external field, r is the distance from the origin (center of the composite) as well as θ is the zenith angle. At the interfaces of the composite, the coefficients A_1 , A_2 , A_3 , B_1 , B_2 and B_3 can be determined by using the appropriate boundary conditions.

It is worth noting that the second term on the right side of Eq 4, represents the induced potential outside the composite. To analysis polarazability and absorption coefficient, the author focused to calculate only the total induced field outside the concentric sphere. In other words, the author is interested on the value of the coefficients B_3 (as shown in Eq 5) rather than other coefficients, which is found to be by concurrently using the above four equations based on normal component of the electric displacement vector and the continuity condition of potential at the interfaces

$$B_{3} = r_{s}^{3} E_{0} \left(1 - \frac{27}{2} \frac{\varepsilon_{h} \left(\frac{B}{\beta_{2}} + A \right)}{X} \right)$$
(5)

where the parameters incorporated in Eq 5 are defined as follow from Eqs 6-8.

$$A = 3\varepsilon_1^2 + \varepsilon_1(\varepsilon_c - \varepsilon_1)\left(\frac{3}{2}\right)\beta_1 - 3\varepsilon_c\varepsilon_s - \varepsilon_s(\varepsilon_c - \varepsilon_1)\beta_1 \tag{6}$$

$$B = 9\varepsilon_1\varepsilon_s + 3\varepsilon_s(\varepsilon_c - \varepsilon_1)\beta_1 \tag{7}$$

$$X = C_1 \varepsilon_s^2 + C_2 \varepsilon_s + C_3 \tag{8}$$

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where C_1 , C_2 and C_3 are constants depending on the dielectrics function of core, spacer and host matrix.

The induced potential Eq 9 outside the inclusion is

$$\Phi_{ind} = \frac{B_3}{r_s^3} \cos\theta = \frac{\tilde{p}}{4\pi\varepsilon_h r_s^3} \cos\theta \tag{9}$$

where \tilde{p} is the electric dipole moment of the system.

In a uniform external electric field, a dielectric object becomes polarized. In far field, the polarized object can be approximated as an electric dipole because the higher order field components decay quickly as a function of distance [38].

The electric dipole moment Eq 10 by combining the above equations:

$$\tilde{p} = 4\pi\varepsilon_h B_3 = \varepsilon_h \alpha E_0 \tag{10}$$

where α is the polarizability of the composite given Eq 11

$$\alpha = 4\pi r_s^n \left[1 - \frac{3}{2} \frac{\varepsilon_h (B/\beta_1 + A)}{X} \right]$$
(11)

Some of the metal properties, including optical properties can be described with the simple freeelectron gas Drude–Sommerfeld model of DF. In the framework of this model, by applying an external field, the conduction electrons move freely between independent collisions occurring at the average rate of size- and frequency-dependent dielectric function ε_s predicted by the Drude– Sommerfeld model as shown in Eq 12 [9,33,39]:

$$\varepsilon(\omega, r_{eff}) = \varepsilon_{\infty} - \frac{\omega_p^2}{\omega^2 + i\omega(\gamma_{bulk} + A_{r_{eff}}^{\nu_F})}$$
(12)

where ε_{∞} is the phenomenological parameter describing the contribution of bound electrons to polarizability, ω_p is the bulk plasmon frequency, γ_{bulk} is the damping constant of the bulk material, v_F is the electron velocity at the Fermi surface, A = 0.25 [33] is an empirical paramete. For Au, $\varepsilon_{\infty} = 9.84$, $\gamma_{bulk} = 0.072 \ eV$ and $\omega_p = 9.01 \ eV$ [9,40], $v_F = 1.38 \ Mm/s$ [41]. r_{eff} is the effective mean free path of collisions and can be calculated as Eq 13 [33,41]

$$r_{eff} = 0.5 \left(((r_s - r_d)(r_s^2 - r_d^2))^{1/3} \right)$$
(13)

3. Numerical result and discussion

For the dimension of composite less than the wavelength of incident light, the quasi-static approach method is appropriate for the calculation of polarizability and then the absorption coefficient of the composite. For small size composite, the incident electric field may be regarded as being spatially uniform over the entire of the particle; so that the particle can be replaced by an oscillating dipole and this is referred to as the quasi-static approximation. In this study, the spectra of triple layered core-shell nanostructures extended in to from visible to the infrared (NIR) spectral region (450–850 nm) were observed.

By considering the quasi-static limit approximation method, the absorption (δ_{ac}) and scattering coefficient (δ_{sc}) as explained from Eqs 14–15, respectively, can be modeled as the optical response,

$$\delta_{ac} = \frac{k}{\pi r_s^2} Im[\alpha] \tag{14}$$

$$\delta_{sc} = \frac{k^4}{6\pi^2 r_s^2} |\alpha|^2 \tag{15}$$

where $k = 2\pi \sqrt{\varepsilon_h} / \lambda$ [42] is a wavevector.

One of the parameter which affects the optical properties of nanocomposite is the size. Herein, in order to compere the optical response of two different size of nano-inclusions by keeping the concentration of spacer and shell remain the same and using Eq 14, the absorption coefficient of the composite is depicted in Figure 2: for the radius of composite 11 nm and 18 nm for Figure 2a,b, respectively. The concentration of spacer and shell for Figure 2a is 64.85% and 53.86% as well as for Figure 2b is 63.56% and 52.95%, respectively. Two resonances are observed associated with the interfaces of metallic shell: the first resonances corresponding to inner interface and the second resonances associated with outer interface. As shown in the figure, the enhancement of absorption coefficient depends on the size of composite, the resonances are enhanced for larger size Figure 2b. Absorption coefficient more sensitive to lower DF of spacer, after $\varepsilon_d = 4$, the optical resonances are no longer sensitive to the increasing of DF in second plasmon resonance-absorption coefficient slightly increased but shifted to higher wavelength. For both configurations, the position of both plasmon resonances' peak is remain the same, i.e., the position of peaks is not depending on the size of nanocomposite.

The dipolar modes accompanied by different number of higher order multipoles, and surprisingly larger absorption coefficient can be achieved via thicker shells. The absorption coefficient of ZnO@M@Au spherical core-shell NP is plotted by further increasing the size of composite to 23 nm as shown in Figure 3. The enhancement and the tunability of absorption coefficient is vary with thickness of spacer and shell. When the shell thickness increase from 4 nm Figure 2b to 9 nm Figure 3a, i.e., the concentration of spacer and shell is 63.56% and 77.45%, respectively: (i) the resonance associated with outer interface more enhanced and slightly shifted to higher wavelength and (ii) the amplitude of resonance associated with inner interface is decreased and red-shifted. Due to the strong interaction of the two plasmon resonance for small shell thickness, the two resonances are move apart [43]. However, when the shell thickness increased, the interaction between the two resonances is weak and the property is becoming dominated by shell material. So, the two resonances shifted to higher wavelength but the amplitude is different. The separated electric charges on the outer surface of shell are freely oscillated. Due to this, the plasmonic resonance is more enhanced for outer interface. The absorption efficiency is also depending on the thickness of spacer for the same size of nano-inclusion. As depicted in Figure 3b, when the spacer thickness increase from 4 nm Figure 2b to 9 nm Figure 3b, i.e., the concentration of spacer and shell is 85.42% and 43.63%, respectively, the first resonances are more enhanced than the second resonances. The resonances associated with the outer interface are tuned to infrared spectral region and the resonance corresponding to vacuum ($\varepsilon_d = 1$) is almost vanish. The resonance with high tunability of nanocomposite from near UV to near IR spectral region is used for drug delivery, photo-catalysis, biomedical, gas-senser application [44–48].



Figure 2. (Color online) Absorption coefficient in different size (a) $r_c/r_d/r_s = 6/8.5/11$ and (b) $r_c/r_d/r_s = 10/14/18$ embedded in silica host matrix.

Extinction coefficient (σ_{ec}) is the summation of absorption coefficient Eq 14 and scattering coefficient Eq 15. The extinction coefficient of ZnO@M@Au nanocomposite is plotted in Figure 3c,d by increasing core size and shell thickness, respectively, as shown in the legend. As shown in Figure 3c, when the size core ZnO NPs is increased for a fixed size of composite 23 nm, extinction coefficient decreased and red-shifted in the second resonance. For a fixed size of composite, when the core size is increased simultaneously the shell thickness decreased. From the result shown in the figure, for the gold's shell thickness was fixed at $r_s = 23 nm$ and when the radius of core was increased from $r_c = 10 nm$ to $r_c = 15 nm$, the extinction coefficient peak of ZnO@M@Au hybrid nanoparticles shifted gradually to the peak of core material. Correspondingly, the concentration of Au shell is 77.45%, 72.26%, 66.34%, 59.62%, 52.07% and 43.63% as well as the concentration of spacer material is 63.56%, 60.56%, 57.81%, 55.28%, 52.95% and 50.80%. On the other hand, by increasing the shell thickness from 4 to 9 nm for a fixed core size 10 nm and spacer thickness 4 nm (see Figure 3d), the extinction coefficient is increased and the peak position is red-shifted and blueshifted, respectively, for the first and the second resonance. When the concentration of metallic shell is increased (52.95%, 59.99%, 65.70%, 70.37%, 74.23% and 77.45%), the properties of the extinction spectra of hybrid core-shell nanoparticles were dominated by the properties of the shell metal. If the concentration of Au is increase to $\sim 100\%$, the two interface's resonances have to merge each other, i.e., a single resonance only for nano-spherical Au.

One the most important factor that affect the optical properties of nanocomposite is dielectrics function of the host matrix. In addition to DF and thickness of spacer, shell thickness as well as size of nano-inclusion; dielectric constant of the surrounding medium is also play a predominant role in the position of the plasmonic peak and its intensity. The author has plotted the effect of DF of the host medium for two different size of nano-inclusion 18 nm and 23 nm. As shown in Figure 4, the absorption coefficient is enhanced and red-shifted specially for the second resonance when the DF of host medium is increased. The first resonance in Figure 4a is enhanced without shifting when the DF of the host medium increased, but, it in Figure 4b is enhanced with red-shift. In general, increase of the dielectrics function of the host medium promotes the appearance of higher order multipolar modes at larger wavelengths and red-shifts the already existing plasmon resonances.



Figure 3. (Color online) Effect of spacer thickness and shell thickness on the absorption coefficient of size (a) $r_c/r_d/r_s = 10/14/23$ and (b) $r_c/r_d/r_s = 10/19/23$. The effect of core size and shell thickness on extinction coefficient (c) and (d) for DF of spacer ($\varepsilon_d = 2.25$). All inclusions are embedded in silica host matrix.



Figure 4. (Color online) The effect of dielectrics function of host medium on the absorption coefficient of different size (a) $r_c/r_d/r_s = 10/14/18$ and (B) $r_c/r_d/r_s = 10/14/23$ for DF of spacer $\varepsilon_d = 1.5$.

For triple layered core-shell nanostructures, the optical property is also affected by core size and spacer thickness for the same size of nano-inclusion. For size of nano-inclusion 23 nm (or 4 nm shell thickness or 43.63% shell concentration), the optical response of nanocomposite for different core

size is depicted in Figure 5. As shown in Figure 5a, the first resonance associated with inner interface is slightly enhanced without shifting, but, the resonance associated with outer interface is more enhanced and red-shifted (shifted to IR spectral region) for the radius of core ZnO NPs 15 nm. However, in Figure 5b for the radius of core 10 nm, the first resonance is more enhanced than the second resonance without shifting and the second resonance is red-shifted. The result indicates that the plasmon band of ZnO@M@Au core-shell nanostructure is sensitive to the big nano-spheres with (i) a big core for the second plasmon resonance and (ii) large spacer thickness for the first plasmon resonance. This result directly related to the interaction of plasmon of the shell material with exciton/polariton of the spacer as well as beyond to the spacer with polariton of the core material. When the size of core material is becoming bigger for the same size of nanocomposite, the interaction plasmon with core material is stronger.



Figure 5. (Color online) The effect of DF of host medium on the absorption coefficient of different core size and spacer thickness (a) $r_c/r_d/r_s = 15/19/23$ and (b) $r_c/r_d/r_s = 10/19/23$ for DF of spacer $\varepsilon_d = 1.5$.

4. Conclusion

In this work, the author studied the effect of core size, spacer thickness, dielectrics function of spacer, shell thickness, size of nano-inclusion and dielectrics function of the host medium on the optical as well as plasmonic properties of triple layered ZnO@M@Au spherical core-shell nanostructure embedded in active host matrix. The absorption coefficient of ZnO@M@Au strongly depends on the thickness of noble metal Au. Absorption coefficient and extinction coefficient as a function of wavelength of incident possess two plasmon resonances with the peaks extending from near UV to IR spectral region. These resonances correspond to the spacer@Au and Au@hostmedium interfaces. The enhancement of absorption coefficient is depending on thickness or concentration of Au. The plasmon resonance associated with inner interface is more enhanced for large spacer thickness as well as plasmon resonance associated with outer interface is more enhanced for large size of core ZnO for a fixed size of nano-composite. The position of resonance peaks was strongly depending on the concentration spacer and shell not on the size of nano-inclusion. For the same size of nanoinclusion, the second resonance is red-shifted when (i) dielectrics function of spacer increased, (ii) shell thickness decreased, (iii) dielectrics function of host-medium increased, and (iv) core size increased and this resonance enhanced when all of these parameters are increased. The first resonance is more enhanced when the concentration of spacer material is increased. Note

that the results shown, triple layered core-shell nanostructures are composed of a semiconductor core of ZnO coated by thin Au NP with different spacer material can be ideal candidate for enhancing biological, solar-cell, catalysis application. Important remark can be pulled in this theoretical work, ZnO@M@Au core-shell nanostructures with large core absorbed more light.

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Conflicts of interests

The author declares no conflict of interest.

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