Nanoscience and Nanotechnology Research, 2017, Vol. 4, No. 1, 1-6
Available online at http://pubs.sciepub.com/nnr/4/1/1
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DOI:10.12691/nnr-4-1-1

Control of the Surface Plasmon Resonance of Two Configurations of Nanoparticles: Simple Gold Nanorod and Gold/Silica Core/Shell

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Abstract We report on the study of optical properties of ellipsoidal nanoparticles forms using Gans's theory and Drude model; we introduce important parameters such as surrounding medium, shell thickness. Nanorods shaped nanoparticles are distinguished of others forms by the appearance of two peaks corresponding to their plasmonic bands: transverse mode placed in the visible region and the longitudinal mode located toward longer wavelength. The simulation results show a strong dependence of core/shell ratio and surrounding medium on longitudinal resonance. Nano shell nanoparticles composed with a big core (gold) aspect ratio are more shifted with increasing of silica layer.

Keywords: *nanorods particles, core/shell particles, surface plasmon resonance*

Cite This Article: A Sambou, P. D. Tall, Kh Talla, O. Sakho B D Ngom, and A C Beye, "Control of the Surface Plasmon Resonance of Two Configurations of Nanoparticles: Simple Gold Nanorod and Gold/Silica Core/Shell." *Nanoscience and Nanotechnology Research*, vol. 4, no. 1 (2017): 1-6. doi: 10.12691/nnr-4-1-1.

1. Introduction

Metal nanoparticles; in particular gold nanorods are very promising for biomedical and detection applications because of their optical absorption properties can be modulated in the visible and the near infrared region by a simple modification of the aspect ratio of the core/shell.

The aspect ratio is the value of the long axis (length) divided by the short axis (width) of a cylindrical or rod-shaped particle. Chemical and biological sensors have been extensively studied due to their wide applications in contamination assessment, medical diagnostics and other areas. Surface Plasmon Resonance (SPR) methods have made an important contribution to the observation of bimolecular interaction [1].

For this type of sensor taking advantage of the nanoparticles of noble metals can be used a relatively simple approach. For noble metals nanoparticles, the plasmon resonance band can be changed by changing certain physical characteristics such as the size, shape, composition and refractive index of the surrounding medium [2,3,4].

Surface Plasmon resonance is collective excitations of the electrons at the interface between a conductor and an insulator and it is described by evanescent electromagnetic waves that are not necessarily located at the interface [5,6]. For in vivo therapies, gold nanospheres are not competent because of optical properties (i.e. absorption, extinction) outside the near-infrared window in biological tissues (650 nm to 900 nm). Instead, gold nanoshells coated on dielectric core [7] and nanorods with high aspect ratio [8,9] are widely employed due to tunable peak absorption from 550 nm up 1 μm.

Exploring the structure of the gold nanoparticles or their form open perspectives of a new therapeutic tool. According to [10] an injection of core shell nanoparticles (55 nm of SiO_2 / 10 nm of Au, corresponding to a plasmonic resonance at 800 nm) in a mammary tumor or a mousse, followed of a laser irradiation of 820 nm with low dose of 4 W/cm^2 during 4 to 6 minutes, allow evolution of temperature at 37±7°C and local destruction of cancerous tissues.

In elliptical shaped or rod shaped gold particles, two separate absorption bands are observed due to transverse and longitudinal structures of the particle, as illustrated in the references [11,12]. The transversal resonance is located in the green spectral region (≈480 nm to 600 nm) and the longitudinal shifted to the infrared region (≈650 nm to 1100 nm). In addition, the longitudinal band of gold nanorods is strongly desired to be in the spectral range of 650-900 nm. Light irradiation in this region can penetrate deeper in tissues and cause less photodamage than UV-visible irradiation.

For gold nanorods, transversal mode, weaker band localization is similar to the gold nanosphere. The two resonance modes, transversal mode and longitudinal mode, correspond to electron oscillation associated with the short and long axis of the nanorod, respectively.

The transverse plasmon band of gold nanoparticles has been found to be insensitive to the changes in the size of the nanorod particle and the surrounding refractive index, whereas the longitudinal plasmon band show à red-shift

with the increase of aspect ratio of nanorod particle and this is very sensitive to any change of the refractive index [13,14].

There are many literature reports about how to control the optical properties of these particles through adjusting their size and shape during wet chemical nanoparticle growth. Based on the specific interaction of the nanoparticles with light, applications in the field of cancer therapy [15], sensor applications [16] are envisaged. In this study, we use core/shell configuration of nanorods in a dielectric environment using Gans theory and Drude model for the simulation of the optical properties (i.e. absorption, scattering and extinction cross section) of silica coated gold nanorod particle.

MATLAB simulations program are used to study and discuss the factors that govern the plasmon resonance (for example the longitudinal resonance) of dielectric shell coated gold nanorods. Matlab is a software and can serve as a tool in developing the adequate mathematical model. We have used to calculate absorption, scattering and extinction cross-section for different structure (aspect ratio). It is a program that can be used also to study the influence of coated dielectric shell and the surrounding medium. To observe resonance phenomenon, we have used Drude model and Gans's theory.

2. Material and Methods

2.1. Gans's Theory

It is in 1912 that Richard Gans modified Mie's theory (exclusively applicable to the spherical nanoparticles) by including ellipsoidal particles [17]. Gans showed that non spherical particles have absorption with wavelengths longer than those of the spherical particles of comparable size. Due to the ellipsoidal shape, the surface plasmon mode would split into two distinct modes. The first is along the short axis of the ellipsoid (axis b and c), while the second is rather along the long axis (axis a) as shown in Figure 1. The two oscillations induce a load separation and thus localized a resonance plasmon in the particle. In this work, we consider nanorods of prolate form as schematized in follow Figure 1.

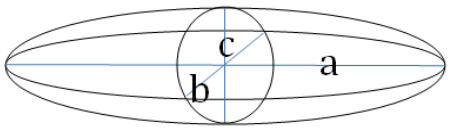

Figure 1. Schematic of an ellipsoidal particle with dimensions (a, b, c), such as a>b=c (prolate form)

To further understand theoretically how we get the single and dual modes, we first need to understand the concept of extinction. According to references [11,13,18], the extinction cross section of the single prolate spheroid is then expressed on the following equations (1).

$$\sigma_{ext} = \frac{2\pi NV}{3\lambda}\varepsilon_m^{3/2}\sum_j \frac{\left(1/L_j^2\right)\varepsilon_i}{\left(\varepsilon_r + \frac{1-L_j}{L_j}\varepsilon_m\right)^2 + \varepsilon_i^2} \quad (1)$$

Where N is the number of particles per unit volume; ε_r and ε_i are respectively the real and imaginary parts of the complex dielectric function of the particle and $\varepsilon_m=n^2$ (n is the refraction index of the matrix in which the particle is encapsulated) is the dielectric constant of the surrounding medium; $V=4*ab^2/3$, is the particle volume (with the condition a>b=c) and L_j (j=a, b, c) is the geometrical depolarization factors describe the shape of the particle given by (2) and (3), ł λ the wavelength of the light in vacuum.

$$L_a = \frac{1-e^2}{e^2}\left[\frac{1}{2e}\ln\left(\frac{1+e}{1-e}\right)-1\right] \quad (2)$$

$$L_b = L_c = \frac{1-L_a}{2} \quad (3)$$

Where e is the ellipticity, given by (4)

$$e = \sqrt{1-\left(b/a\right)^2} \quad (4)$$

The aspect ratio £ of the nanorod is thus defined as £=a/b.

In our simulations, constants values and protocol are introduced in article [7] such as Drude model.

In this study, first material is considered as prolate core form with semiaxis $a_1>b_1=c_1$ coated with a second material shell with other semiaxis $a_2>b_2=c_2$. The polarizabilities depend of the function of core ε_a, shell ε_b and matrix dielectric constants ε_m. The polarizabilities α_j for core/shell nanorods particles can be calculated as fallows [19,20].

$$\alpha_j = \frac{V\left\{\begin{array}{l}\left(\varepsilon_b-\varepsilon_m\right)\left[\varepsilon_b+\left(\varepsilon_a-\varepsilon_b\right)\left(L_j^{(1)}-fL_j^{(2)}\right)\right]\\ +f\varepsilon_b\left(\varepsilon_a-\varepsilon_b\right)\end{array}\right\}}{\left\{\begin{array}{l}\left[\varepsilon_b+\left(\varepsilon_a-\varepsilon_b\right)\left(L_j^{(1)}-fL_j^{(2)}\right)\right]\\ \times\left[\varepsilon_m+\left(\varepsilon_b-\varepsilon_m\right)L_j^{(2)}\right]\\ +fL_j^{(2)}\varepsilon_b\left(\varepsilon_a-\varepsilon_b\right)\end{array}\right\}} \quad (5)$$

Where $V=4a_2b_2c_2/3$, is the volume of the particle; $f=a_1b_1c_1/a_2b_2c_2$, is the volumic fraction of the core; L_j, is the depolarization factors (j=1, 2, 3) refer to the longitudinal (axis a) and transverse modes (axis b and c), (k=1, 2) refer to the core and shell given respectively by the relation (6) and (7).

$$L_1^{(k)} = \frac{1-e_k^2}{e_k^2}\left(-1+\frac{1}{2e_k}\ln\frac{1+e_k}{1-e_k}\right) \quad (6)$$

$$L_2^{(k)} = L_3^{(k)} = \frac{1}{2}\left(1-L_1^{(k)}\right) \quad (7)$$

Where e_k, given by:

$$e_k^2 = 1-\frac{b_k^2}{a_k^2}. \quad (8)$$

Thus, the extinction σ_{ext} can be modeled as the optical response and we have the following relation.

$$\sigma_{ext} = \sigma_{abs} + \sigma_{sca} \quad (9)$$

Where σ_{abs} and σ_{sca} are the absorption and scattering cross sections and are given respectively by the equations (10) and (11).

$$\sigma_{abs}(\lambda) = \frac{k}{3}\sum_{i=1}^{3} \mathrm{Im}\,\alpha_j(\lambda) \tag{10}$$

$$\sigma_{sca}(\lambda) = \frac{k^4}{18\pi}\sum_{i=1}^{3} \left|\alpha_j(\lambda)\right|^2 \tag{11}$$

Where k is a coefficient, which depend on wavelength and given by equation (12)

$$k = \frac{2\pi}{\lambda}\varepsilon_m^{1/2}. \tag{12}$$

3. Results and Discussion

3.1. Results Related of the Single Gold Nanorod

The spectra (in arbitrary units) of the gold nanorods, gold/silica core/shell nanorods with different aspect ratio and surrounding medium are calculated in the wavelength region 200-1200 nm. The simulated optical properties of the gold nanorods are shown in Figure 2. In this first part, we study the influence of the aspect ratio (Figure 2.a) and the dielectric constant of the surrounding medium (Figure 2.b) on the position of the band resonance. Figure 2.b, shows the calculated extinction cross sections for the gold nanorod with 3.55 aspect ratio, a series of simulation were performed for different refraction indexes of surrounding medium: water n=1.333, ethanol n=1.359 and benzyl alcohol n=1.538. Results of the numerical simulations indicate that the extinction spectrum of the gold nanorods is very sensitive to the refractive index of the surrounding bulk medium.

The extinction spectra of gold nanorods are characterized by two peaks; the dominant band, at longer wavelength and a much weaker band, at shorter wavelength. The first band located in the green spectral region corresponds to the oscillation of the electrons perpendicularly to the major (long) rod axis, and it is referred to as the transverse plasmon due the polarization of the incident light along the short axis.

The transverse band is relatively insensitive to the nanorod aspect ratio (only decrease of the intensity with increasing of aspect ratio) (Figure 2.a), but is sensitive to the surrounding medium and the refractive index change in the medium (Figure 2.b). The other band occurs at longer wavelength and the infrared region. This is caused by the oscillation of the electrons in parallel to the major (long) rod axis, and is known as the longitudinal surface plasmon. This band is very sensitive to the aspect ratio and by increasing the aspect ratio (length divided by width), the longitudinal band maximum shifts to longer wavelength with a relative decrease in the peak intensity, seen Figure 2.a. For example, we can see that an increase of the gold nanorods aspect ratio, the longitudinal band red-shift from 672 nm to 694 nm for aspect ratio from 2.66 to 2.88. Thus, for the aspect ratio, £=2.66, £=2.88, £=3.11, £=3.33, £=3.55 and £=4.00, the longitudinal band is observed at 672 nm, 694 nm, 716 nm, 738 nm, 761 nm and 805 nm. As the aspect ratio increases, the energy separation between the two bands of the plasmon resonance increases. This means that the longitudinal band can be red shifted with increasing the length of the nanorod particle, as envisaged by Gans.

According to the results reported by H. Y. Wu and co-authors [21], our simulated spectra are in reasonably in agreement. By comparison, there is relatively good agreement for the results presented by Boris Khlebtsov and co-authors [22] where they have established with a length of gold nanorod equal to 40 nm and width of 14 nm (aspect ratio, £=2.85), they have observed the resonance at a wavelength around 696 nm in surrounding medium at dielectric constant ε_m=1.776. With same size and medium, we shifted at 692 nm (either ≈4 nm of difference). It is well known through that the plasmon resonance, principally longitudinal mode is very sensitive to change in the refraction index of the surrounding dielectric medium. The maximum of the longitudinal band shift to the longer wavelengths with increasing refraction index, as illustrated in Figure 2.b. We found the following values: 760 nm, 770 nm and 840 nm for n=1.333, n=1.359 and n=1.538, respectively. The both band, transversal and longitudinal increases in intensity.

In summary, the transverse plasmon resonance peak is not quite as sensitive to the change of aspect ratio, as the longitudinal peak, which shows noticeable shifts in the aspect ratio as seen in Figure 2.a and is illustrated in Table 1. The result indicates the longitudinal plasmon band of gold nanorods is more sensitive to the surrounding medium.

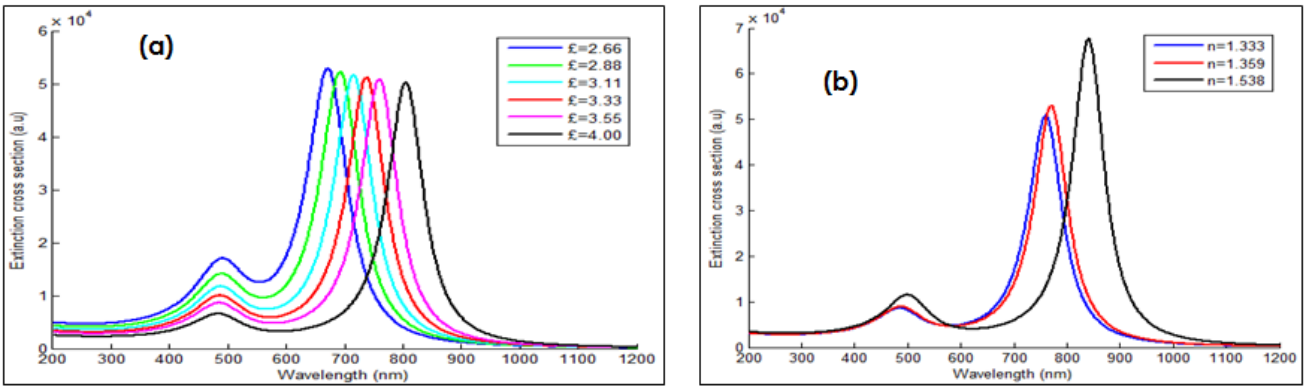

Figure 2. Spectral distribution of the extinction cross section based (a) aspect ratio and (b) surrounding medium

Table 1. Wavelength of the longitudinal band according with the surrounding medium and aspect ratio of gold nanorods

Longitudinal resonant wavelength (nm)			
Aspect ratio	n=1.333	n=1.359	n=1.538
1.55	555	560	597
1.75	580	585	623
2.00	605	612	653
2.33	639	646	693
2.50	655	663	714
2.80	685	693	750
3.50	755	765	834
3.88	794	805	882
4.66	873	886	976
5.38	946	961	1063

For the gold nanorod nanoparticles, for aspect ratio superior or equal at 2.50, the spectra exhibit a maximum of longitudinal band in the near infrared region in water. We not that the longitudinal plasmon mode of gold nanorod in ethanol and benzyl alcohol are located at wavelength longer than 650 nm when aspect ratio is approximately superior at 2.33 and 2.00, respectively. To test the performance of the theoretical result, we have compared the resonance wavelengths obtained by equation (1) with that of results obtained by the experimental methods. According to the work by N. G. Khlebtsov [23] and C. Gautier and co-authors [24], our results are in good agreement with the literature. By example, these authors have found a longitudinal plasmon band at 600 nm for an aspect ratio of 2.00 and 820 nm for aspect ratio of 4.2±0.9, respectively. All nanoparticles are emerged in water. For the same parameters and conditions, we have found the plasmon band at 605 nm and 824 nm.

The optical spectra (absorption, scattering and extinction) measured for the gold nanorods with the aspect ratios 1.55, 2.00, 3.50 and 4.66 are shown in Figure 3.

According to the Figure 3, these results reveal two remarks, we found that for the small nanoparticles (i.e. aspect ratio important), absorption dominates scattering, whereas for large nanoparticles scattering dominates absorption. These results are reasonable because the absorption cross section equation is proportional at $1/\lambda$ whereas scattering is proportional at $1/\lambda^4$.

In between the two extremes, the contribution of absorption and scattering to total extinction can be made approximately equal. These observations open the way to the study of the size distribution of gold nanorods. All these simulation results confirm the equation (9).

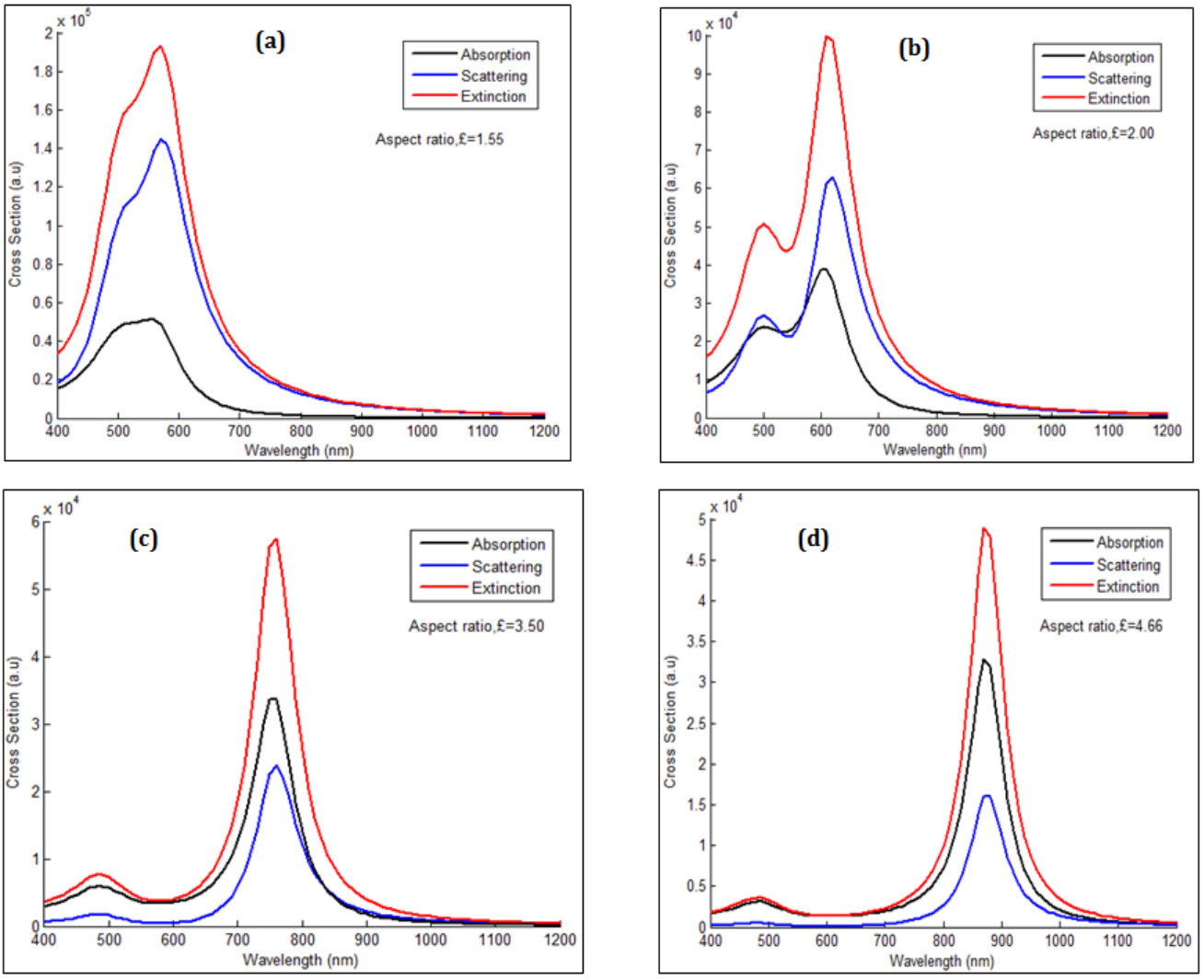

Figure 3. Calculated absorption, scattering and extinction for various aspect ratio of gold nanorod in water

3.2. Optical Response of a Composite Gold/Silica Nanorod

Silica (SiO_2) is a particularly attractive candidate material for it's role as active dielectric in a nanorod coating system. Silica shell is known to diminishe the bulk metal conductivity and improves the chemical and physical stability of the core component [25]. Silica can be used to block the core surface from making contact with a biological environment and also silica particles produced via the Stöber Method have been shown to be chemically biocompatible [26].

In this simulation part, for metals (i.e. gold), ε is usually modeled using a Drude approach and for silica material, the optical parameters (n, k) used are taken from the values measured by E. Palik [27]. Cores of gold nanorod is considered to be a prolate spheroid with dielectric function ε_a and silica shell is considered to be a spheroid shell confocal to the core with dielectric function ε_b.

When investigating the optical properties of the core/shell nanorods using simulation method, both the plasmonic properties of the gold nanorod cores and the effect of the different shell thickness on extinction spectra can be observed, as illustrated in Figure 4. Figure 4 (a) reveals the results of gold nanorods; the rods have a length and width of 70 nm and 28 nm, respectively and are encapsulated in a 4-22 nm thick silica layer. These nanorods nanoparticles have an aspect ratio of 2.50. Figure 4(b) shows the variation of the peak longitudinal resonance plasmon of Au/SiO_2 as a function of aspect ratio of core and shell thickness: Curve blue, green, red and black corresponding of aspect ratio of core equal at 3.50, 4.11, 5.00 and 5.83, respectively. In this simulation, we have used in Figure 4 (b) the same shell thickness (ts) for the four colours ts=0 nm, ts=4 nm, ts=8 nm and ts=12 nm.

In this way, Figure 4(a), for gold nanorod nanoparticles, curve bleu, the spectrum exhibits two maximums, around 496 nm with less intense peak and another maximum at 660 nm, these bands are attributed to transverse and longitudinal modes, respectively. For nanoshell, i.e. when we coated gold nanorod with silica (with the thickness of 4 to 22 nm), the transverse plasmon peak does not shift significantly, and may be readily tuned between 496 nm and 510 nm (polarization perpendicular to the long axis). The spectra exhibit a strong band in the red shift with the increase of the layer of silica added. The peak longitudinal has shifted to slightly longer wavelength (i.e. $\lambda_{max} \approx$ 660-685 nm) for shell thickness of 0 to 22 nm. According to the work of C. Gautier and co-workers [24] and Q. Zhan and co-workers [28], the simulation results for Au/SiO_2 nanorods found in this work closely resemble the experimental spectrum and we can be valeted our results.

In Figure 4 (b), the evolution of longitudinal band resonance is evidenced and the corresponding distributions of aspect ratios. As illustrated in figure, an increase the aspect ratio, 3.50, 4.11, 5.00 and 5.83, the results without coated of silica (solid curve) given in the red shift of the longitudinal bands at 755 nm, 817 nm, 907 nm and 992 nm, respectively. The dielectric shell (silica) coated gold nanorod with four values, ts= 0 nm, 4 nm, 8 nm and 12 nm, as shown in Figure 4 (b). We can see that, with the increasing shell thickness, the wavelength maximum increase rapidly toward the longer wavelength and also increase in intensity.

This result shows the following important remarks: Firstly, an increasing the silica layer, the full width at half maximum (FWHM) decrease, by consequent this band tends to be constant, as illustrated in Figure 4 (a). Secondary, for different size of core gold nanorods with a fixed shell thickness, the longitudinal resonance has moving most long with increasing aspect ratio. By example, for the four aspect ratio 3.50, 4.11, 5.00 and 5.83, a wavelength shift of 10 nm (755-765 nm), 13 nm (817-830 nm), 13 nm (907-920 nm) and 18 nm (992-1010 nm) for 4 nm of thick of silica. Can be deduced from figure 4 (b) that the size of core makes one basic parameter in the position of resonance wavelength. But also, we can probably say that for same thickness of silica, gold/silica core/shell resonance is most moved with increasing of core aspect ratio.

Summary, for single gold nanorods, change of size or surrounding medium gives a little variation in the transverse mode contrary to the longitudinal mode. For the nanoshell nanorods, at a given shell thickness, the position of the longitudinal resonance peaks in the extinction spectrum are strongly dependent of the size (i.e. aspect ratio) of the core. This study shows that the core size, is the important factor which controls the size and to imagine longitudinal band position.

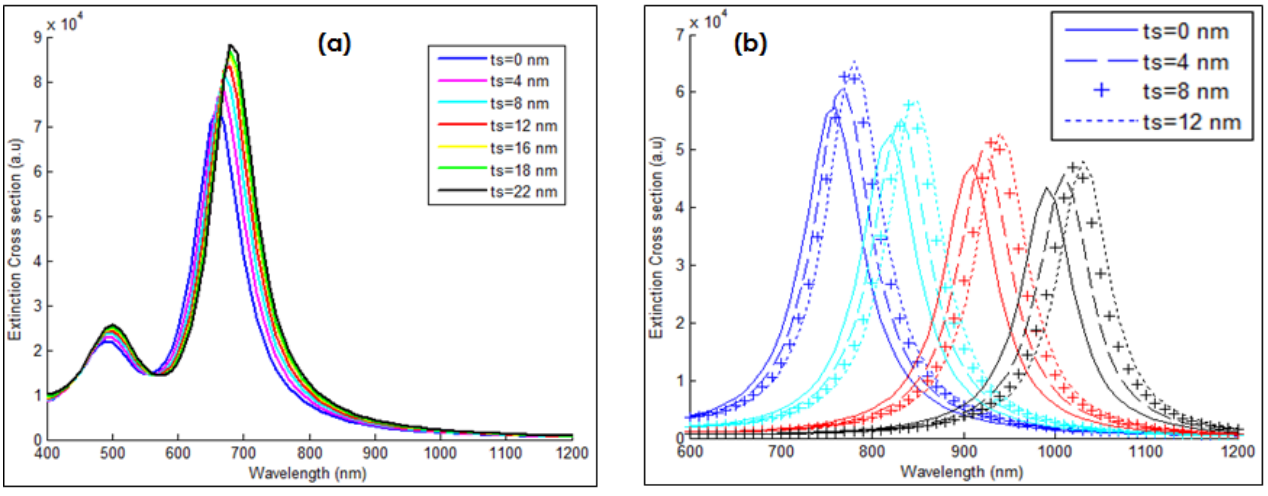

Figure 4. Simulation spectra of gold/silica nanorod nanoshell at different values of Au/SiO_2 emerged in water, a refractive index of 1.333

4. Conclusion

In this work, we have studied two configurations of nanoparticles: simple gold nanorod and gold/silica core/shell. Our objective is to control the plasmon resonance. We have studied the influences of size, surrounding medium, and size of core and shell thickness on the plasmon resonance of the Au and Au/SiO_2. Our simulation results have been compared with the one of the literature. The results indicate that gold nanorods present two plasmon resonances, one in visible spectrum located around 496-510 nm (transverse mode) and one in near infrared or infrared region (longitudinal mode). However, the longitudinal was strongly depending on the aspect ratio and the surrounding medium. For gold/silica nanoshell, it is demonstrated clearly from the results that for the same silica layers, nanoshell, which have a bigger core aspect ratio, induced the formation of longitudinal resonance, which is redshifted. We can notice that the influence of shell thickness depending of core size. These results offer real prospects for research and applications through suitable medical. Only a careful control of the nanoshell size before it can be used for biomedical applications. Our study recommends the material with core aspect ratio superior or equal at 2.50 if the material is emerged in water.

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