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Optical Properties of Nano-structured Silica Coated Silver Particles

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Abstract

Resonance frequency of silver nanoshells was investigated by numerical calculations based on the Drude correlation model to the Mie approximation. In a system of Ag atom coated with silica material. The optical properties of nanostructured architectures are highly sensitive to their composition, structures, dimensions, geometries and embedding mediums. Then the simulated absorption spectra of single-component metal nanoparticles and Ag@SiO_2 nanoshell were calculated using both Mie and Drude model. We describe the benefits of plasmon resonance shaping by means of a simulation consisting in optimizing the plasmon frequency of multifunctional hybrid nanomaterials of the core/shell type. The discussion concerns the maximum absorption of the Ag/SiO_2 system identified at about 430 nm if the material is immersed in water. In the second phase, we modulated the SiO_2/Ag ratio to adapt it to the biological window. Thus, the various tests allowed us to conclude that to satisfy the question, the ratio of core radius (≈SiO_2) to shell thickness (≈Ag) must be significantly greater than 5. This has resulted in considerable insight concerning the variation of plasmon wavelength with nanoparticle size, shape and dielectric environment, as well as the use of these particles for optical sensing applications.

Keywords

Drude Model, Mie Theory, Silver Nanoparticle, Silver Nanoshell, Surface Plasmon Resonance, Silica

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1. Introduction

In the nanometric size range, the optical properties of unique gold and silver particles are those of the solid modified by dielectric confinement: in particular Surface Plasmon Resonances. Noble metals have fascinated humanity for several millennia. Such fascination is not only the result of ancestral customs or social conventions. Scientific knowledge provides some explanations for this extract. Indeed, the advent of nanoscience has opened up new opportunities. Gold and silver nanoparticles have been studied and used in several fields of high-tech and biotechnological applications, such as sensory probes [1], medical diagnosis [2], catalytics [3, 4], nanoelectronics [5, 6], etc. This extraordinary interest is reflected in the enormous increase in original published work and the large number of journals dealing with preparation, electronic structure and optical properties in combination with the potential applications of gold and silver nanoparticles [7, 8]. However, these nanoparticles are widely explored for biomedical applications because of their advantages of easy synthesis and surface functionalization. For example, gold nanoparticles are of great importance for electronic and biomedical applications that want to use gold and silica nanoparticles for the selective destruction of cancer cells. The particularity of the nanoparticles of noble metals (gold [Xe] $4f^{14}5d^{10}6s^1$, silver [Kr] $4d^{10}5s^1$ and copper [Ar] $3d^{10}4s^1$),

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exhibit a strong absorption band in the visible range. This band is called Surface Plasmon Resonance (SPR). This physical phenomenon changes according to several parameters such as the nature of the metal [9], the size [10, 6], the surrounding environment [11, 6], the shape [8, 12, 6]. In 1908, Mie [13] was the first to explain this phenomenon theoretically by solving Maxwell's equations for an electromagnetic radiation field interacting with a spherical metal particle under appropriate boundary conditions. Using electrodynamics, he applied an exact electromagnetic theory that describes the absorption and scattering of an electromagnetic wave by spherical particles. When the particle is subjected to an electromagnetic field with a very large wavelength in relation to the size of the particles, the electrons in the conduction band have a collective oscillation movement (Figure 1).

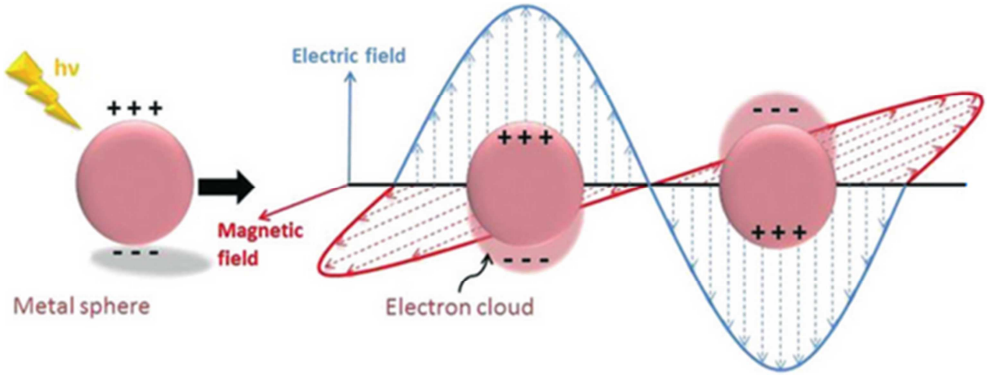

Figure 1. Schematic illustration of a localized surface plasmon resonance.

When the two frequencies (frequency of the electromagnetic wave and the natural oscillation frequency) are equal, a resonance phenomenon called surface plasmon resonance (RPS) is observed. This physical phenomenon, which results from the collective vibration of the electron gas on the surface of the nanoparticle, involves the electrons 6s of the gold conduction band [14, 15]. As shown in Figure 1, nanoparticles of noble metals are characterized by a single resonance band located in the visible. For example, C. E. Rayford II et al [10], studied spherical gold nanoparticles in aqueous solution with respective radius of 10nm, 25nm and 50nm, the observed resonance peak is 516nm, 524nm and 552nm respectively. Concerning the absorption peak of the silver metal studied experimentally, is recorded at 396nm [16] for a spherical size of 30±6.8nm, at 400nm [17] for a size of 16nm. The morphology of nanoparticles seems to have a greater influence on the position and width of the plasmon band. Unlike spherical nanoparticles, ellipsoidal or nano rods nanoparticles are characterized by the appearance of two plasmonic bands on either side of the RPS position of a nanosphere of the same volume. These two resonances are related to the orientation of the polarization: we have the transverse component also called the transverse mode, observed when the polarization is along the minor axis and the longitudinal component also called the longitudinal mode, when the polarization follows along the major axis of the ellipsoid [18]. This hypothesis is based on Gans' postulate, known as the Gans theory [18, 19]. Thus, in 1912 Richard Gans extended Mie's theory by considering spheroidal particles and showed that non-spherical particles have an absorbance at higher wavelengths than spherical particles of comparable size. In order to improve the optical properties of nanoparticles (≈surface plasmon resonance), several authors [20-22] have developed compound structures. These are the nanostructures called "core/shell" generally known as nanoshells. Thus, nanostructured two-layer systems, in two-phase form, offer a wide range of new properties, potentially extending the already numerous applications of metal nanoparticles. In particular, core-shell nanostructures have tunable properties mainly based on the two compounds, core size and shell thickness. It then becomes possible to modify and control, for example, the surface plasmon resonances of such particles.

Very recently, it has been shown that it is possible to precisely control the size of gold noanoshell with silica, a particularly inert material, can be used in biomedicine. This illustrates once again the change in optical properties induced by the reduction or increase in the size of the material. Like the gold nanoparticles already published, we present in this study an optimization of Ag/SiO_2 nanoshells for biomedical applications. Thus, for this application, it is recommended from several articles that these types of materials must absorb in the near infrared spectral region (650-900nm) [23, 24]. This is the region of the spectrum for which photon penetration is deeper, thus allowing a cancerous region to be locally heated and destroyed without destroying healthy tissues near the tumor, as are the disadvantages of various methods involving a laser, microwaves, radio frequencies and ultrasound.

Spherical silver nanoparticles have useful attributes such as optoelectronic properties related to size and shape, a high

surface-to-volume ratio, excellent biocompatibility and this is due to their excellent conductivity, chemical stability and their use as a catalyst [25]. These properties make silver nanoparticles an important tool in medical science [6].

2. Method and Materials

In this section is detailed the Drude model and the values of the optical parameters of the Drude model. The Drude model explains the electrodynamic properties of metals. The simple approach is to regard the conduction band electrons as non-interacting electron gas and yields a fairly accurate description of metals like silver, gold or aluminum. Several experimental optical done are listed in table 1. Mie's theory, used by many authors, is claimed to be valid only for spherical particles and concerns the scattering of light by small particles in front of the wavelength. Thus, to determine the size dependence of the dielectric function of metal particles, it is first necessary to decompose the dielectric function into two contributions: an interband contribution (explaining the response of 5d electrons) and an intra-band contribution for free electrons (Drude). From this the equation of the dielectric function ε (ω) of the metal is written [26, 27]:

$$\varepsilon(\omega)=\varepsilon^{inter}(\omega)+\varepsilon^{intra}(\omega) \tag{1}$$

The authors stated that the Drude model is used to describe the behaviour of conductivity electrons and thus to determine the intra-band dielectric function. And from this, the intra-band dielectric function of the metal is as follows:

$$\varepsilon^{intra}(\omega)=1-\frac{\omega_p^2}{\omega^2+i\omega\Gamma} \tag{2}$$

Equation (2) can be rewritten by separating the real part ε_r from the imaginary part ε_i.

$$\varepsilon^{intra}(\omega)=1-\frac{\omega_p^2}{\omega^2+\Gamma^2}+i\frac{\omega_p^2\Gamma}{\omega(\omega^2+\Gamma)}=\varepsilon_r^{intra}(\omega)+i\varepsilon_i^{intra}(\omega) \tag{3}$$

For optical frequencies, Γ« ω. This amounts to rewriting equation (3) and as a function of λ as follows:

$$\varepsilon_r^{intra}(\lambda)\approx 1-\frac{\lambda^2}{\lambda_p^2} \tag{4}$$

$$\varepsilon_i^{intra}(\lambda)\approx\frac{\lambda^3\Gamma}{\lambda_p^2} \tag{5}$$

Table 1. Optical parameters of gold and silver of the Drude model.

Materials	Contribution interbande		λp (nm)	Γ=1/τ (sec)
Au	Real: 6,9 [28, 29]	Imaginary: 2,45 [30]	145 [31]	$175e^{-7}$ [28]
Ag	Real: 3,7 [28, 32]	Imaginary: 3,9 [28, 32]	139,60 [31]	$31e^{-15}$ [28, 32]

By always placing oneself in the quasi-static approximation and starting from Laplace's equations, we obtain the expression of the polarizability α of metallic nanoshells [33]:

$$\alpha=4\pi R_2^3\varepsilon_0\left\{1-\frac{3\left[(\varepsilon_2-\varepsilon_1)R_1^3+(2\varepsilon_2+\varepsilon_1)R_2^3\right]\varepsilon_m}{2(\varepsilon_2-\varepsilon_1)(\varepsilon_m-\varepsilon_2)R_1^3+(2\varepsilon_2+\varepsilon_1)(2\varepsilon_m+\varepsilon_2)R_2^3}\right\} \tag{6}$$

In the case of a multilayer sphere (particularly a core-shell system), the expression of the effective absorption cross-section as a function of the dielectric functions of the three media (core ε_1, shell ε_2 and surrounding medium ε_m) or simply of the polarisability α is given by [33]:

$$\sigma_{abs}=\frac{2\pi}{\lambda\varepsilon_0}\mathrm{Im}(\alpha) \tag{7}$$

On the other hand, there must be equality of the wave vector of incident light and that of the surface plasmon, this is how several authors have defined the relationship opposite [28]:

$$\varepsilon=\varepsilon_1(\omega)+i\varepsilon_2(\omega)=(n+ik)^2 \tag{8}$$

Of this equation, n denotes the refractive index describing the phase velocity of the wave and k denotes the extinction index describing the absorption of the wave during its propagation in the material. These indices are related to the real and imaginary parts by the following relationships $\varepsilon_1(\omega)=n^2-k^2$ and $\varepsilon_2(\omega)=2nk$. Numerically, we used the numerical values n and k of SiO_2 established by E. Palik et al [34] and for metals we adapted them to the Drude model explained above.

3. Results and Discussion

We conducted numerical analyses to determine the efficacy of nanocomposites containing silver nanoparticles for use as active targeting (therapeutic strategy). The materials considered, silver and silica, were first adapted to the nanoscale and during the entire stimulation process our materials are considered studied in water (n=1,333) as a surrounding environment. This work is based on the optimization of a few parameters such as the nature of the metal, size and composition. We are committed to making a comparative study of the quantitative results obtained by our digital program and the one available in the literature.

3.1. Efficient Absorption Section for Gold and Silver Nanoparticle

Figure 2 shows the surface plasmon resonance gain corresponding to the effective absorption cross-section for spherical gold and silver nanoparticles with a radius R=30nm. It is from the expressions given in the equations from the Drude model that we have simulated the optical response of these two metallic nanospheres. First, we note the appearance of a single resonance mode, which characterizes the surface plasmon resonance (RPS) of the nanospheres of noble metals.

In the specific case of 30nm gold nanoparticles, this resonance is detected around 516nm while the plasmon resonance of silver nanoparticles is located at 408nm. Under the same study conditions, there is agreement with the data in the literature for objects of similar size, which confirms our model and validates our study code. For example, Y. Sun et al [35], obtained an absorption maximum at 516nm and 405nm for gold and silver nanoparticles respectively. Indeed, this remarkable optical property, which is characterized by the appearance of an absorption band, depends on the size as well as the shape of the nanoparticles as illustrated in previous publications [36, 37].

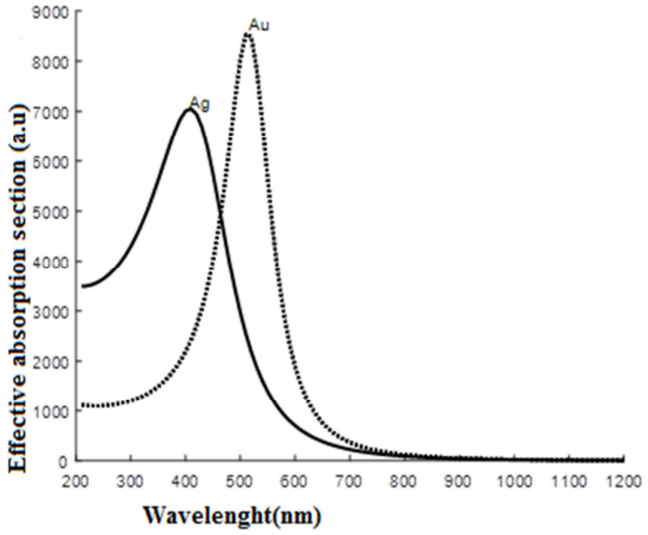

Figure 2. Absorption spectrum of gold and silver nanoparticles.

3.2. Modulation of the Optical Properties of Nanoshells

This particular process can therefore lead to systems with different optical properties depending on the state of structure of the nanoshells. In this section we then analyzed two types of configuration, namely Ag/SiO_2 and SiO_2/Ag. The interest here is therefore to follow the formation mechanism since silver nanostructures are distinguished by surface plasmon resonances totally dependent on their shape and structure. The desired objective was to have an absorption of silver nanoshells around 800nm to later evaluate their photothermal properties. For an application in the field of nanomedicine, the silicic shell would provide an additional function: it would allow, a priori, the encapsulation, transport and release of an active principle [38, 39].

3.2.1. Effective Absorption Section of the Nanoshell Ag/SiO_2

Figure 3 below shows the plasmonic evolution of Ag/SiO_2 nanosystems. The core material which is the silver nanosphere at a radius R=30nm and the silica shell varies from 5nm to 50nm. The evolution of the spectrum is mainly related to the growth of nanoparticles. However, the greater the size of the silica thickness, the greater the absorption frequency range of nanoparticles. In addition, the width of the peaks is relatively large with the size of the material. The maximum absorption spectrum first appears at 408nm (shown in blue on the spectrum in Figure 3) and corresponds to the polar component of the plasmon resonance of spherical silver nanoparticles. As particle size increases, this resonance intensifies and moves very slightly towards long wavelengths. When the nanoparticles reach a SiO_2 size of about 30nm, the resonance frequency is positioned at 430nm. After multiple tests, it is found that this position corresponds to the maximum absorption of the studied system (i.e. Ag/SiO_2) is constant at the absorption frequency λ_{abs}=430nm (i.e. a maximum offset of ≈22nm with the addition of SiO_2 on silver). From these observations, we can say that the absorption of nanoparticles was almost unchanged by the formation of the silicic shell.

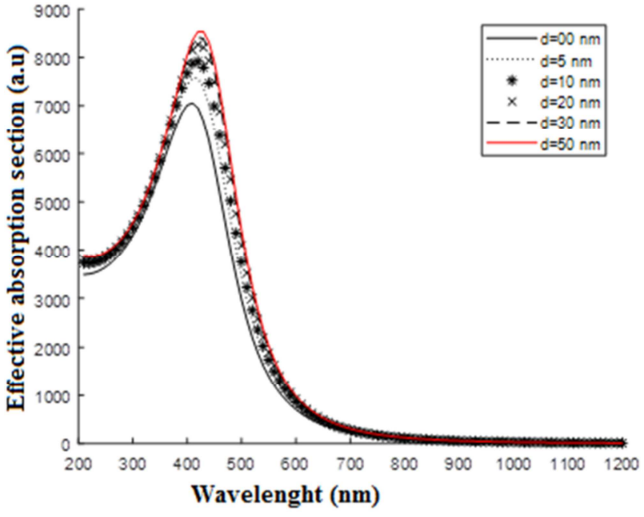

Figure 3. Absorption spectrum of Ag/SiO_2 nanoshells.

3.2.2. Effective Absorption Section of the SiO_2/Ag Nanoshell

Figure 4 shows the optical properties for silica/silver core/shell nanoparticles with different sizes. The silica nanoparticles radius is kept constant at 25nm. She translates plasmon resonance evolution as a function of the thickness of

the silver layer deposited on a silica nanosphere. First of all, it is important to note that silica has no absorption band as shown in Figure 4 on the left. As the shell thickness is increased, the peaks shift to the blue and become more intense. The absorption peak appears to be well differentiated according to the relative thickness of silver since it moves from a wave frequency of 640nm (low thickness ≈5nm) to 460nm (higher thickness ≈20nm). The morphology of nanomaterials seems to have very little influence on the width at mid-height of the resonance bands, but significantly on the position of the plasmon band. Thus, in the case of silver nanoshells, which consist of a dielectric core (SiO_2) and a metal shell (silver), the latter is considerably displaced towards red compared to silver nanospheres of the same size.

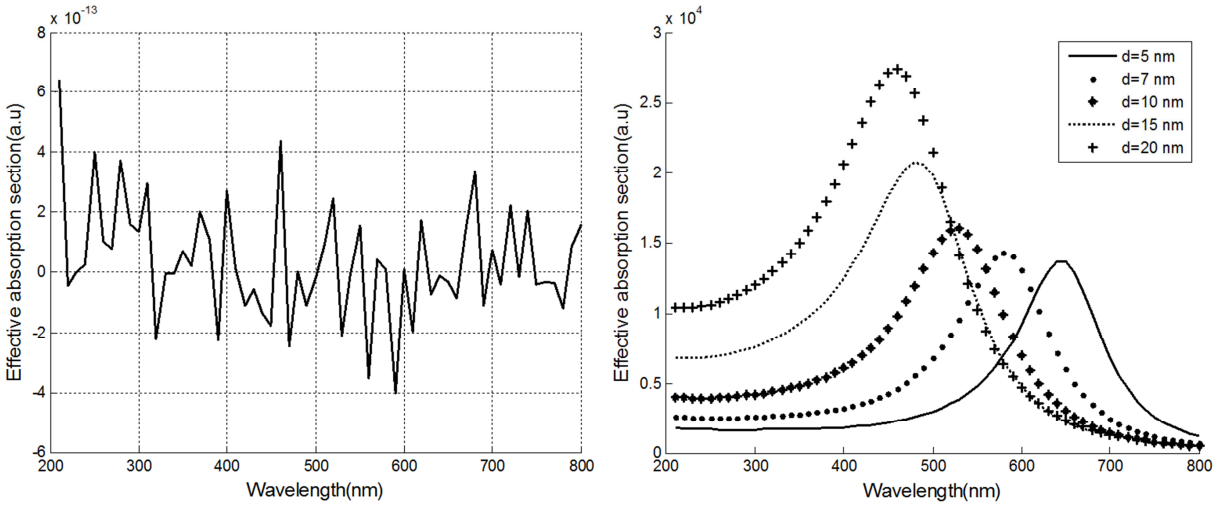

Figure 4. Influence of silver nanoparticle size on silica nanosphere: On the left silica nanosphere and on the right silver nanoshell SiO_2/Ag.

It appears that when a small silver layer (5nm) is added to the silica nanosphere, the absorption is located at the long wavelengths (λ_{abs}=640nm). For a thickness of 7nm of silver, the absorption is shifted to short wavelengths (λ_{abs}=580nm). After a 10nm deposition, the absorption band is further displaced towards red. The changes from a spectral point of view show that when the deposited silver layer is thinner than 5nm, the nanoshell can lead to an absorption around 650-900nm. Finally, the study shows that when the added thickness is too great, the absorption peak will almost disappear from the near infrared region. Through the spectra above, we were able to demonstrate that it is possible to visualize the optical behavior of silver nanoshells via the silver precursor size used. However, it is quite difficult, from one test to the next, to accurately draw the correct core/shell ratio for the intended application. In order to preserve optical activity around the near infrared, we can probably retain a material developed on the basis of a theoretical ratio greater than 5. The optical properties of spherical silver and silica/silver nanoparticles can be tuned by adjusting the physical dimensions. The dielectric properties of the material are extremely important and play a large role in the intensity and placement of the plasmon resonances.

3.2.3. Influence of SiO_2 Core Size on Plasmon Resonance

The digital optical absorption spectra highlighting plasmon resonances for different core diameters are shown in Figure 5.

The size of the silica nucleus varies from 10nm to 30nm and the silver shell of thickness equal to 5nm fixed for all six systems. They also reveal a significant change in the structure of the nanoshells as the size of SiO_2 increases (i.e. when the theoretical SiO_2/Ag ratio increases). As the size of SiO_2 increases, a significant shift of the plasmon resonance band towards red is induced, in other words, absorption spectrum shifted to longer wavelengths with, as illustrated in Figure 5 below. From a qualitative point of view, the analysis of the SiO_2/Ag characteristic peak reveals the results opposite: 500nm, 550nm, 645nm, 690nm, 760nm and 830nm corresponding respectively to the SiO_2/Ag nanoshell with a 10/5, 15/5, 25/5, 30/5, 40/5 and 50/5 ratio. This large study confirms the previous study. From these observations, we can therefore affirm that the theoretical ratio between the radius of the core (SiO_2) and the thickness of the shell (Ag) allowing to observe a resonance in the near infrared must be greater than 5. In the continuity of these results, the hybrid architecture of the SiO_2/Ag core/shell system, the size of the core would be larger than that of the shell.

The above paragraphs provide the full description of the silver nanoshells of the Drude model. This same process and code was used in our recent article [36] for gold nanoshells. Although many theoretical calculations of the optical properties of noble metal nanoparticles have not used this model, our results are very much in agreement with the literature as confirmed in the first part of the results. This is not a comparative study, according to ours calculations, the

maximum absorption difference is about 100nm in favour of the Au/SiO_2 system. In other words, gold nanoshells would have more interesting optical results (surface plasmon resonance) than silver nanoshells. This difference may be due to the difference in plasmon energy volume of the gold metal (8.98eV) and the silver metal (8.85eV) [40]. In summary, for spherical silver nanoparticles, associated with a silica shell, the surface plasmon resonance localizes to a maximum absorption band at ~430nm. However, in the case where silver particles are deposited on silica, the signal measured in absorption depends strongly on the size of the silica. Metal shells have a resonance that varies very quickly with the thickness or size of the core. Figure 6 shows the schematic configuration of the type of spherical nanoshell studied in sections.

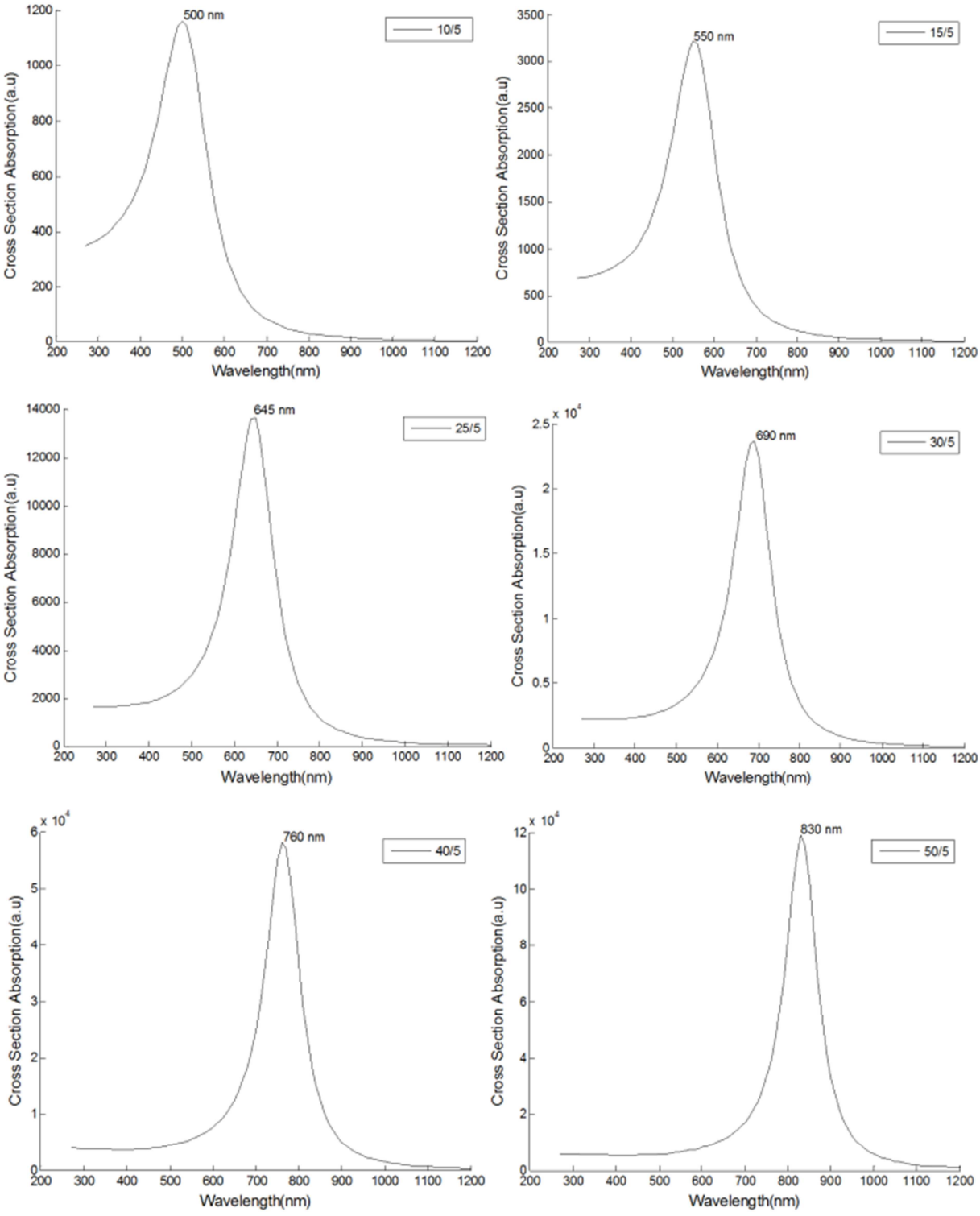

Figure 5. Absorption peak for the different SiO_2/Ag compositions.

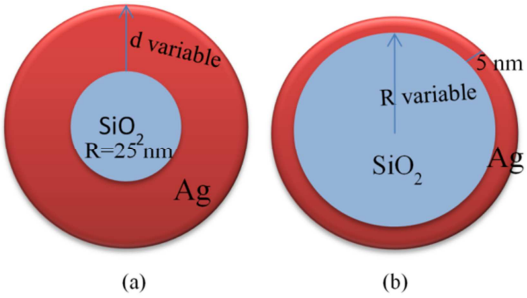

Figure 6. Schematic structure of silver nanoshells SiO_2/Ag.

4. Conclusion

In this study, Mie and Matlab simulations have been applied to illustrate the optical properties of typical polyhedral, both single-component structures and Ag@SiO_2 core-shell configurations. The polyhedral, we have investigated spherical nanoparticles, most of the simulation have been compared with the UV-Vis spectra from previously published works. Drude model has been presented in this work to examine the influence of composition and core sizes on the absorption curve. We performed consistent numerical calculations of the surface plasmon bands from the Drude model. Our application of Mie theory and the Drude model demonstrate an almost perfect agreement of our results with the corresponding experimental data. Thus, the interest of this work is the optimization of a core/shell nanomaterial, hybrid Ag/SiO_2 systems with a more biocompatible character. This result therefore highlights the influence of the growth of nano-objects on the phenomenon of surface plasmon resonance. Finally, we obtained nanoshells with sizes suitable for the formation of silica or silver shells whose optical properties meet the desired properties. It is concluded from this study that silver nanospheres must have a theoretical ratio of radius (silica) to thickness (Ag) significantly greater than 5 for the resonance band to appear in the biological window for applications in the expected nanomedicine domain.

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