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RESEARCH ARTICLE

SIMULATED OPTICAL PROPERTIES OF GOLD-SILVER ALLOY NANOSHELL WITH DIFFERENT COMPOSITION.

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Abstract

In this paper the optical characteristics of the core-shell nanoparticles based on Au-Ag nanoshell core and materials silica shells are presented. Theoretical results on the optical characteristics gold-silver, as well as the measured properties of (Au-Ag)@SiO₂ are investigated considering the surface plasmon resonance phenomenon associated with the surfaces of metals. This study allows us to determine the influences of the metal rate and silica thickness on the resonance plasmon surface. The profile of the resonance bands of a gold nanoparticle in contact with a silver metal is determined by the composition distribution of each metal. Discussion on the influence of various parameters (composition, thickness of shell...) on optical properties of nanoshell nanoparticles. The numerical simulations showed that the resonance band is in visible region after silica coating on Au-Ag nanoparticles. A semiclassical theory (Mie theory) is used to study the optical behaviour of nanoshell nanoparticles. This theory is useful to improve resonance frequency to size ratio study at different percentage of gold and silver. However, the resonance frequency shows an evolution toward weak wavelength and also according (Au-Ag)@SiO₂ nanospheres, results for these structures demonstrated potential applications in windows optical, because of high absorption cross section occurring in the wide band of visible spectrum.

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Introduction:-

Nanoparticles have unique chemical and physical properties as compared to their solid bulk materials because of their high surface area and electronic properties. Small particles exhibit complex optical and physical properties. Their small sizes (<100 nm) cause strong confinement of the electrons, giving rise to fascinating effects not observed in the bulk material. The most striking phenomenon encountered in metal nanoparticles is electromagnetic resonances due to the collective oscillation of the conduction electrons. These so-called localized surface plasmon resonances depend on the local environment, shape, size and composition of the particle (Mulvaney et al., 1996; Brzobohatý et al., 2015).

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Metal nanoparticles provide magnetic (Cherukuri et al., 2010) such as iron oxide nanoparticles (Fe_3O_4), optical (Chen et al., 2005) and electronic (Adams et al., 2003) properties which are different from the corresponding bulk metal materials, leading therapeutic (Loo et al., 2004), photothermal (Wu et al., 2011) and electronic (Adams et al., 2003) device applications, respectively. Recent research efforts have explored metal nanoparticles having complex compositions with controllable sizes and morphologies (Sambou et al., 2016; Sambou et al., 2017).

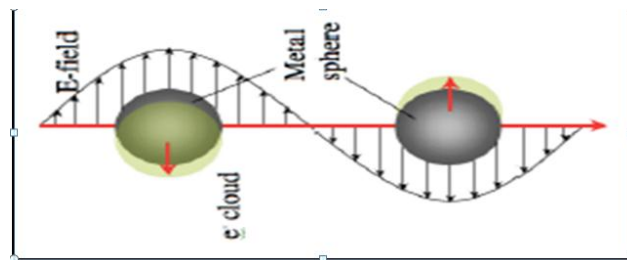


Fig.1:-Schematic of the coherent oscillations of the surface conduction band electrons induced by the oscillating electric field (Kelly et al., 2003)

Special attention has been given to metal nanoparticles for use as optical materials for example silver (Sambou et al., 2016; Soulé et al., 2013) and particularly gold (Soulé et al., 2013; Feis et al., 2014). Gold (Au) and silver (Ag) nanoparticles have a diversity of interesting properties between which they emphasize the electrical ones, optical, catalytic and the applications in biomedicine like antibacterial and antiviral, same that depend on their morphology and size

Noble metal nanoparticles (i.e. gold, silver and copper) have unique optical properties that arise due to the collective oscillation of conduction band electrons along the particle upon excitation by an electromagnetic field, which is known as the plasmon resonance. The interaction between light and electrons of the metal particle is illustrated in Fig. 1.

These electrodynamics properties render nanomaterial as bright contrast agents for imaging and highly effective tools for therapeutic applications. Noble metal particles, as gold nanostructures have been shown to be relatively biologically compatible, showing low or negligible cellular toxicity (Pourbaix et al., 1984; Lewinski et al., 2008).

Gold $[\text{Xe}]4f^{14}5d^{10}6s^1$ and silver $[\text{Kr}]4d^{10}5s^1$ nanoparticles are both excellent plasmonic materials, they each have a fixed plasmon resonance for a given geometry. Gold nanosphere band is localized toward 516 nm (Feis et al., 2014) and for silver nanosphere nanoparticle this band is observed at ≈ 400 nm (Chen et al., 2005; Ashkarran et al., 2013). Gold-silver alloys, on the other hand, hold great promise because their plasmonic resonance wavelength can be tuned by changing the alloy composition. Compared with single particles, core/shell particles have many practical applications, especially in the biomedical and electronic fields. In the biomedical field, core/shell nanoparticles are mainly used for controlled drug delivery (Wu et al., 2010), for bioimaging (Daneshvar et al., 2008; Law et al., 2009), for cell labeling, as biosensors (Nie et al., 2014; Qiu et al., 2010) applications. These particles are called nanoshells particles, which constitute a special class of nano-composite materials. They consist of concentric particles, in which particles of one material are coated with a thin layer of another material (Xia et al., 2000; Caruso et al., 2001).

Gold and silver nanoparticles are used in different fields including drug delivery, sensing and detection. The wide applicability is due to their extremely chemical and physical, high surface area, tunable optical, stability, properties small size and non-cytotoxicity. Targeted gold nanoparticles delivery interacts with the cancerous cell. Silver nanoparticles have proven worthy in inhibiting the microbial proliferation and microbial infection. In short, the wide applicability of the silver and gold nanoparticles is due to the novel properties of the nanoparticle, which help with applications excellent catalytic, good biocompatibility, large surface area and conductivity.

While much work has been done on spherical core-shell systems. Recently, we (Sambou et al., 2016) demonstrate that nanoshell $\text{Au}@\text{SiO}_2$ nanoparticles shown a maxima peak at 530 nm using local refractive index as water ($n=1.33$). (Zhang et al., 2013) have examined $\text{Au}@\text{Ag}$ core-shell nanoparticles by calculating the absorption and scattering efficiency, it is well known through that the plasmon resonance is very sensitive to change in thickness of

the silver nanoparticle. The maximum resonance band shift to the weak wavelengths with increasing thickness of the silver.

To determine the plasmon resonance, for nanoparticles with a spherical symmetry, Mie scattering theory provides a rigorous solution that describes well the optical spectra of spheres nanoparticles of any size. Mie's theory predicts that below a certain size, less than one-tenth of the optical wavelength, the position and width of this band should remain constant, independently of size (Horvath et al., 2009; Alvarez et al., 1997).

The paper is structured as follows. In section 2, we present theoretical aspect, as analytical expressions of Reflectance, Transmittance and equation corresponding of the absorption cross section of nanoshell structure allowing measuring the optical phase spectrum. In section 3, we report optical properties results of Au-Ag alloys and (Au-Ag)@SiO₂ core-shell nanoshell nanoparticles, respectively. Nanoshell particles are optimized with different ratios of Au and Ag nanoparticles and these alloys are coated with various shell thickness of SiO₂ nanoparticle, which demonstrates potential applications in visible region, as optical field's photonic and plasmonic waveguides. According to Mie theory, we calculate the optical phenomena, such as the Absorbance, Reflectance, Transmittance and Absorption cross section.

Method and Materials:-

Reflectance (R) and transmittance (T) equation for single nanosphere particles can be written as (Heavens et al., 1955):

$$R = \left\{ A_1 \exp\left(\frac{4\pi h k_2}{\lambda}\right) + A_2 \exp\left(-\frac{4\pi h k_2}{\lambda}\right) + 2\sqrt{A_1 A_2} \cos\left(\phi_2 - \phi_1 - \frac{4\pi h n_2}{\lambda}\right) \right\} \times \left\{ \frac{\exp\left(\frac{4\pi h k_2}{\lambda}\right) + A_1 A_2 \exp\left(-\frac{4\pi h k_2}{\lambda}\right)}{2\sqrt{A_1 A_2} \cos\left(\phi_2 + \phi_1 - \frac{4\pi h n_2}{\lambda}\right)} \right\}^{-1} \quad (1)$$

$$T = \frac{n_3}{n_1} \{ B_1 B_2 \} \left\{ \exp\left(\frac{4\pi h k_2}{\lambda}\right) + A_1 A_2 \exp\left(-\frac{4\pi h k_2}{\lambda}\right) + 2\sqrt{A_1 A_2} \cos\left(\phi_2 + \phi_1 - \frac{4\pi h n_2}{\lambda}\right) \right\}^{-1} \quad (2)$$

Where,

$$A_{1,2} = \frac{(n_{1,2} - n_{2,3})^2 + (k_{1,2} - k_{2,3})^2}{(n_{1,2} + n_{2,3})^2 + (k_{1,2} + k_{2,3})^2} \quad B_{1,2} = 4 \frac{(n_{1,2})^2 + (k_{1,2})^2}{(n_{1,2} + n_{2,3})^2 + (k_{1,2} + k_{2,3})^2} \quad (3) \quad (4)$$

$$\phi_{1,2} = \begin{cases} \arctan \left[2 \frac{n_{1,2} k_{2,3} - n_{2,3} k_{1,2}}{(n_{1,2})^2 - (n_{2,3})^2 + (k_{1,2})^2 - (k_{2,3})^2} \right] + \pi, \\ \text{if } (n_{1,2} k_{2,3} - n_{2,3} k_{1,2}) \geq 0 \text{ and } [(n_{1,2})^2 - (n_{2,3})^2 + (k_{1,2})^2 - (k_{2,3})^2] < 0; \\ \arctan \left[2 \frac{n_{1,2} k_{2,3} - n_{2,3} k_{1,2}}{(n_{1,2})^2 - (n_{2,3})^2 + (k_{1,2})^2 - (k_{2,3})^2} \right] - \pi, \\ \text{if } (n_{1,2} k_{2,3} - n_{2,3} k_{1,2}) < 0 \text{ and } [(n_{1,2})^2 - (n_{2,3})^2 + (k_{1,2})^2 - (k_{2,3})^2] < 0; \\ \arctan \left[2 \frac{n_{1,2} k_{2,3} - n_{2,3} k_{1,2}}{(n_{1,2})^2 - (n_{2,3})^2 + (k_{1,2})^2 - (k_{2,3})^2} \right] + 0, \\ \text{if } [(n_{1,2})^2 - (n_{2,3})^2 + (k_{1,2})^2 - (k_{2,3})^2] \geq 0. \end{cases} \quad (5)$$

(n₁, k₁), (n₂, k₂) and (n₃, k₃) are the refractive indexes of the substrate, film and surrounding medium, respectively, with n_i and k_i (i=1, 2, 3) represent real and imaginary part of refractive index for each material considered. The

absorbance (A), is given by $A = 1 - R - T$. In equation (1) and (2), h is equal to $2r$, where r is the radius of the nanosphere particle distributed on the surface of the substrate (i.e. glass).

According to the Mie theory, the absorption cross section σ_{abs} of nanoshells is given by (Erickson et al.):

$$\sigma_{abs} = \frac{2\pi}{\lambda \epsilon_0} \text{Im}(\alpha) \quad (6)$$

Where $\epsilon_0 = 8.85 \times 10^{-12}$ F/m, is the permittivity of vacuum and α the polarizability defined by equation (7):

$$\alpha = 4\pi R_2^3 \epsilon_0 \left\{ 1 - \frac{3[(\epsilon_b - \epsilon_a)R_1^3 + (2\epsilon_b + \epsilon_a)R_2^3] \epsilon_m}{[2(\epsilon_b - \epsilon_a)(\epsilon_m - \epsilon_b)R_1^3 + (2\epsilon_b + \epsilon_a)(2\epsilon_m + \epsilon_b)R_2^3]} \right\} \quad (7)$$

With ϵ_m is the dielectric constant of surrounding medium ($\epsilon_m = n^2$) where n is refractive index. The dielectric function of the core is noted $\epsilon_a = \epsilon_r + i\epsilon_i$ and the dielectric function of the shell is noted $\epsilon_b = \epsilon_r + i\epsilon_i$. The complex dielectric function and the complex index of refraction are defined as:

$$\epsilon_a = \epsilon_b = (n + ik)^2 \Rightarrow \begin{cases} \epsilon_r = n^2 - k^2 \\ \epsilon_i = 2 \times nk \end{cases} \quad (8)$$

The optical parameters n and k which designate the refractive index where n is the real part and k is imaginary part.

Results and Discussion:-

Transmittance, reflectance and absorption of gold and silver nanoparticles:-

Fig. 2 shows comparisons spectra of transmittance, reflectance and absorption for the gold (solid curve) and the silver nanoparticles (dotted line curve) respectively as a function of wavelength. These spectra were obtained using the nanoparticles considered to be spheres of radius equal at 30 nm and particles are deposit on the surface which is the glass for the surrounding medium considered is air. The dielectric constants used for simulations were taken from (Rioux et al., 2014) for gold and silver nanoparticles and for air theses constants provide of M. Green (Green et al., 2008).

Optical spectra were taken in the photon wavelength range between 400 and 800 nm. Clearly, we observe except reflectance, gold nanoparticles are presented the optical results better than silver nanoparticles. One rate of reflectance superior at 95% is observed for silver nanoparticle from visible to near infrared region whereas for gold nanoparticle, we observe two regions: the first one for the wavelength values 400 nm $< \lambda <$ 480 nm, the rate reflectance is about 29.68% and the second region, corresponds to high wavelength value (500 nm $< \lambda <$ 800nm), reflectance rate increase strongly up to 97%. The optical absorption peak of gold nanoparticle has been found at between 400 nm to 465 nm, the absorption is maxima. After a visible region, the curve falls of 65% about 2.13% in the near infrared region (\approx 650 to 800 nm).

In short, we can assert that in all band spectra (400 to 800 nm), gold nanoparticles are much absorbed than silver nanoparticles and the last quote is reflected than the first. For most optical applications, high reflectance in the spectrum band (of visible about near infrared range) is very important. In this work, Silver nanoparticles show high reflection up to 97%. For transmittance characteristic, we can also see that the intensity of the peaks decrease with the wavelength for silver nanoparticles whereas this figure show two characteristic for gold nanoparticles; strong at 500 nm (\approx 11%) and become weak in near infrared.

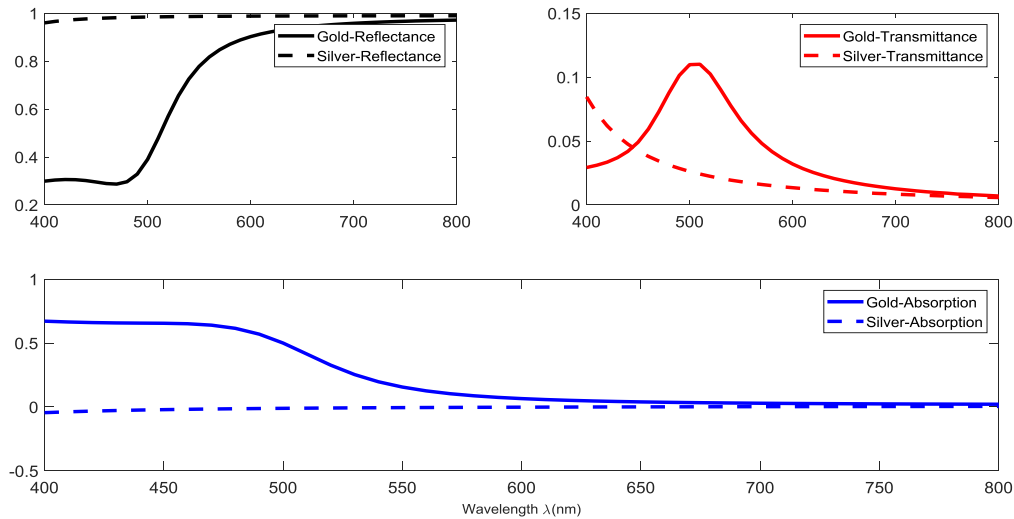


Fig. 2:-The optical properties of gold (solid curve) and silver (dotted line curve) film in air

Optical Absorption of Au-Ag alloy:-

As may be seen from the graph, the behaviour of the optical absorption for alloy formation is entirely different from that for alloy composition. The same protocol is described in the previous paragraph. The size of Au-Ag alloys nanoparticles plotted in Fig. 3 was estimated to be 30 nm and immersed in water.

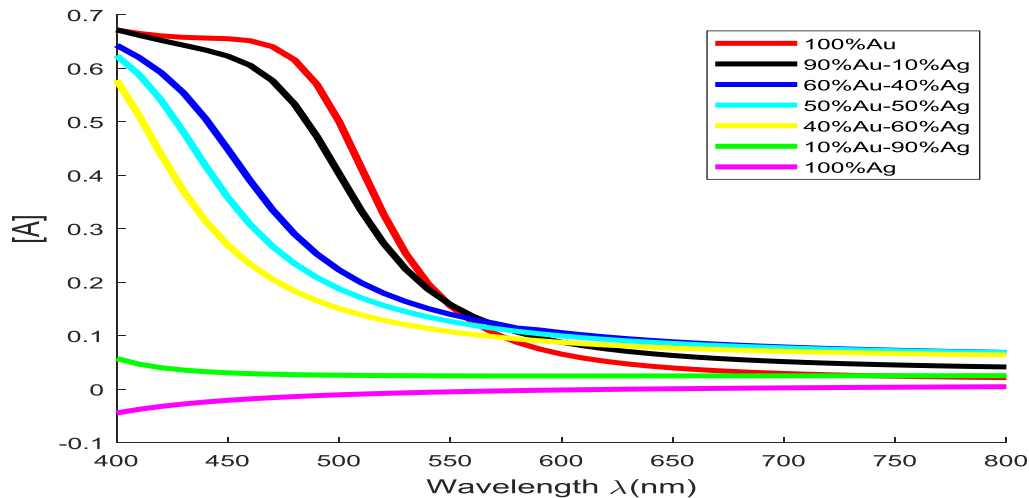


Fig. 3:-The optical absorption of gold-silver alloy

Theory studies were performed with different percentages of gold and silver nanoparticles. In this figure, we have inserted the results of measurements for seven alloy structures as illustrated. For a given materials composition, the absorption rate decreases. Then we observe two different parts.

In the first part, before 560 nm, we have illustrated in Fig. 3 that the increasing of Ag percentage in the alloy composition decreases the nanoparticles' absorption in the visible region. However, in the second region (>560 nm) where the absorption is weak, we see two approaches: the first one for the gold percentage superior to silver percentage, the absorption rate increases, and the second corresponds to silver percentage superior to gold percentage, the optical absorption rate decreases.

Optical absorption cross section of gold-silver alloy:-

Fig. 4 shows the optical absorption cross section of simple gold and silver nanoparticles and also presented the optical absorption cross section of gold-silver alloys nanoparticles. These materials radius is 30 nm and study in water ($n=1.333$). Optical properties were measured in the wavelengths region of 300-800 nm.

In Fig. 4, all absorption spectra band are located in the green spectral region, caused by the oscillations of the nanoparticle electrons on surface. For gold nanoparticles (red curve), the spectrum exhibits a maximum of absorbance at 517 nm with less intense peaks and silver nanoparticles are localized at 400 nm. According to the work by (Frederix et al., 2003) and (Solomon et al., 2007), our results are in good agreement with the literature, which has been confirmed our simulation model of studies of the optical response. The result indicates the much weaker band of gold nanoparticle than silver nanoparticle band as illustrated in Fig. 4.

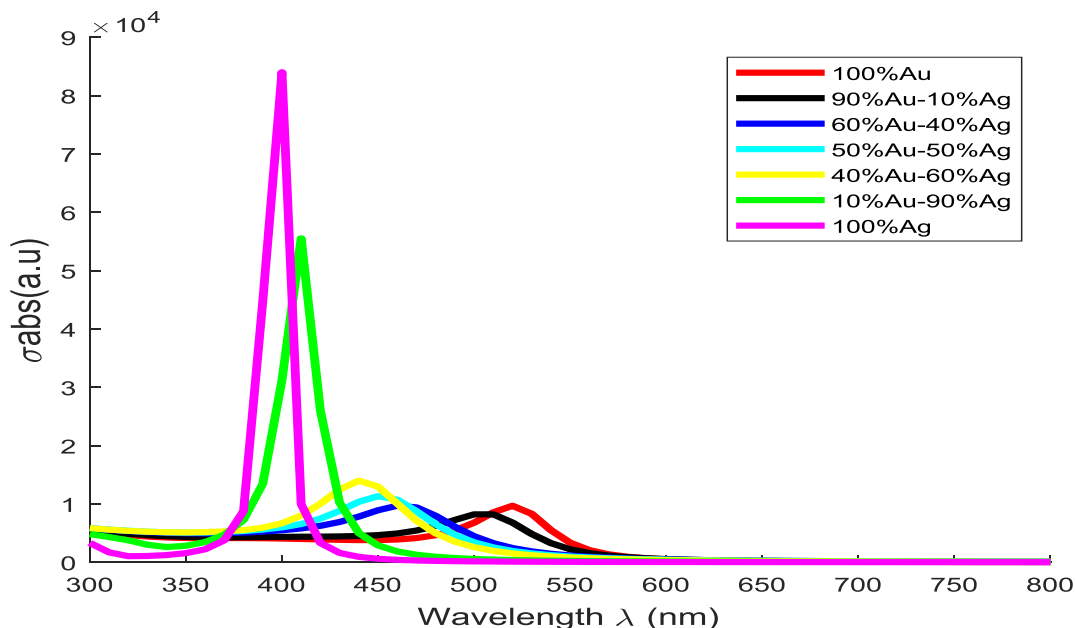


Fig. 4:- Spectral distribution of the absorption cross section based on alloy composition

To investigate the effect of the rate of silver nanoparticles coated gold particles on the optical properties and the plasmon resonance surface characteristics, the plasmon shift measured at different percent of gold and silver particles. Firstly, we can clearly observe that with increasing silver percentage, the characteristic resonance plasmon absorption peak is blue shift. The alloy absorption magnitude increase when the silver percentage growth in the system Au-Ag. The resonance band of the Au-Ag nanoparticles is located at 517 nm, 505 nm, 465 nm, 450 nm, 440 nm, 410 nm and 400 nm respectively for 100% Au, 90% Au-10% Ag, 60% Au-40% Ag, 50% Au-50% Ag, 40% Au-60% Ag, 10% Au-90% Ag and 100% Ag.

In consequence, the gold / silver rate ratio tends to decrease during alloy formation leading to the blue shift observed of the plasmon response. Moreover, such results are in accordance with the works of (Link et al. 1999) that showed a strong blue shift of the optical properties (plasmon absorption) during the formation of alloy.

Characterizations of (gold-silver)/silica nanoshell:-

For silica coating, nanoshells with Au-Ag alloy was selected since these nanoshells absorb light in the visible spectrum. Properties of shell materials (metal or semiconductor) having thickness in nanometer, become important when they are coated on dielectric cores to achieve higher surface area. Sometimes they are referred as core shell or core@shell particles also. Using Equation 6, we obtained Fig. 5 which shows the theoretical results of the effect on the silica shell size absorption cross section spectrum deposited on Au-Ag alloys (radius, $R_1 = 30$ nm).

The thickness of silica layer plotted in Fig. 5 was estimated to be 0, 10, 20, 30, 40 and 50 nm. The alloys core whit those nanoshells show optical absorption in the visible range of the electromagnetic spectrum. Numerical simulation results show the evolution of peak position and full width at half maximum (FWHM) as a function of silica shell

size. The Plasmon resonance of gold-silver/silica core/shell generated on performed nanoparticles undergoes toward a red shift as the shell thickness increase.

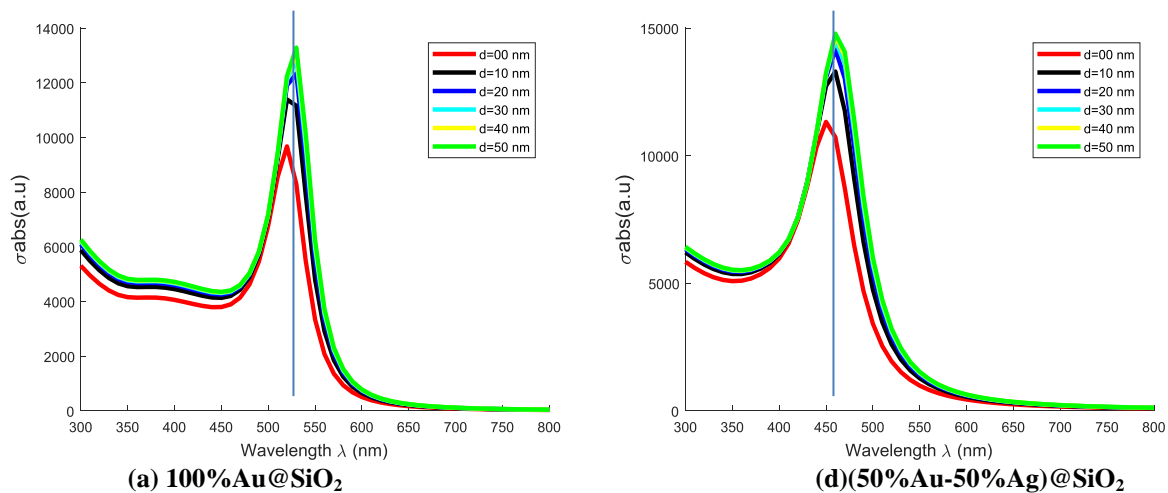
Position of absorption band shows small variations toward the longer wavelength and increase in intensity with silica shell size. A Visible spectrum of (Au-Ag)@SiO₂ show the absorption bands with maxima at 400-530 nm corresponding to so-called plasmon resonance. Resonance activity of (Au-Ag)@SiO₂ depends on the size of the silica shell. We consider figure 5.d, which shows the effect of varying the thickness of a shell of SiO₂ placed on (Au-Ag) nanosphere of 30 nm radius. The plasmon resonance of a SiO₂ shell on an Au-Ag core becomes evident toward red region when the SiO₂ is present at a sufficiently large thickness. In this case an interesting mixed system results, with a tunable plasmon response available from the Au-Ag core and a switchable one from the SiO₂ shell as show in Fig.5. Numerical simulation results which estimate maximum absorption of system are reported in table 1.

Table 1:-Maximum absorption of the (Au-Ag)@SiO₂ core@shell nanosphere immersed in water (n=1.333)

Au-Ag alloy	100%Au	90%Au-10%Ag	60%Au-40%Ag	50%Au-50%Ag	40%Au-60%Ag	10%Au-90%Ag
RPS_{MAX}(nm)	517	505	465	450	440	410
(Au-Ag)@SiO ₂	100%Au@SiO ₂	(90%Au-10%Ag)@SiO ₂	(60%Au-40%Ag)@SiO ₂	(50%Au-50%Ag)@SiO ₂	(40%Au-60%Ag)@SiO ₂	(10%Au-90%Ag)@SiO ₂
RPS_{MAX}(nm)	530	510	470	460	450	420
ΔRPS_{MAX}(nm)	13	5	5	10	10	10

This table, present a decreasing evolution of the resonance plasmon surface. The resonance band decrease toward weak of wavelength with increasing both of thickness of the silica film and rate of silver nanoparticles, whereas, the resonance band has a tendency of increasing in intensity. Optimization of the conditions of optical proprieties revealed that the ratio between the alloy composition and thickness of silica is a key parameter that controls the homogeneity of the materials.

In short, we can be noticed in the spectra that silver present in the system, resonance band is relatively weak. Clearly, the resonance band increase with increasing silver percentage and the band plasmon of the (Au-Ag)@SiO₂ nanospheres are blue shifted relative to resonance band of Ag nanoparticle that is one movement toward 400 nm.



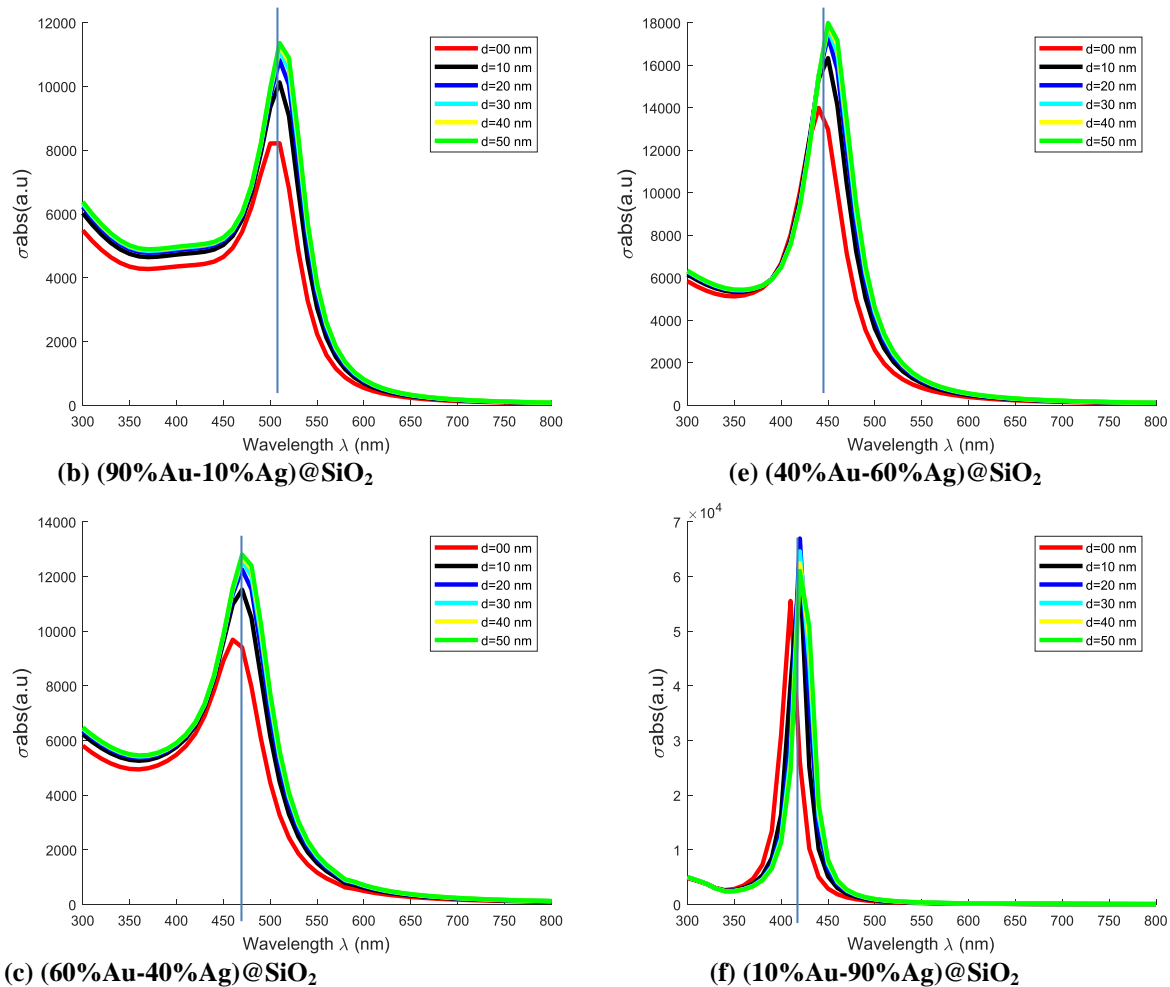


Fig. 5:- Optical absorption cross section of (Au-Ag)/SiO₂ nanoshells in water ($n=1.333$) function of shell thickness

Concerning the optical properties of the (Au-Ag)@SiO₂, the silica shell does not modify significantly the plasmon response of Au-Ag alloy. Silica coated Au-Ag revealed only a slight blue red shift (about ≥ 5 nm) of the λ_{\max} in comparison to the non-coated Au-Ag (see table 1). Plasmon resonances of silver and gold nanoparticles are localized at 400 nm and at 517 nm, respectively. In the case of Au-Ag alloy nanoparticle, the surface resonance plasmon band is moved towards silver nanoparticles characteristic. Finally, since the plasmon response of nanoshell is also probably influenced by the formation of Au-Ag alloy.

Conclusion:-

In summary, in our study we were interested at the effects of the percent of gold and silver in film and at the evolution of the plasmon resonance of the gold-silver alloy and (gold-silver)@silica thin film. Before that we have measured gold and silver nanoparticles optical properties (i.e. absorbance, reflectance and transmittance). Our model based on the Mie theory distribution allowed us to fit the resonances peaks. We showed that the optical absorption of gold-silver alloy decreases when the silver rate increases. This study shows also that absorbance rate of silver much negligible in comparison with the gold nanoparticle particularly in the visible range. The optical properties of Au-Ag alloys nanoparticles are highly dependent on metal composition. Au-Ag alloys spectra have a high resonance at blue spectrum (peaks near 400 nm), caused by the oscillations of the electrons on nanoparticle surface. Finally, since the formation response of nanoshell is also probably influenced by the formation of Au-Ag alloy, however Au-Ag coated with SiO₂, plasmon resonance band involves a weak absorption towards higher wavelengths. In conclusion, we keep that whatsoever the size and structure composition of the (Au-Ag)@SiO₂ nanoshell are absorbed only in visible rang which makes them good candidates for optical windows.

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