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Investigation of Controlled Zinc Oxide/Copper Nanoparticles for Optoelectronic Application

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ABSTRACT

We report surface plasmon resonance in zinc oxide/coper core-shell study between 300 and 800 nm. Two main areas are discussed. First, the optical behaviour of single particles such as silver, copper and zinc oxide are observed. It is shown in particular that the zinc oxide is highly transparent with average transmittance exceeding 83% in the wavelength region lying between 300 and 800 nm. The simulation results also show that the size of the zinc oxide is not very sensitive on the transparency of the material. Secondly, the plasmonic oscillations of the ZnO/Cu configuration are evaluated according to the size of the deposited copper layer. Surface plasmon resonance tends to evolve in the short wavelength direction and around 390 nm which is the characteristic peak of copper nanoparticle. These results suggest that ZnO/Cu nanoparticles are a good candidates for optoelectronic applications.

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Introduction

Research on the metallic nanostructures has been quite intensive over the years thanks to their ability to generate surface plasmon resonances, which are collective oscillations of conduction electrons of the metals at the interface with a dielectric media. Plasmon resonances provide the capability of the localization and manipulation of light at nanoscale [1]. This field of science constitutes a fundamental study of nanotechnology. Metallic nanoparticles find applications in optical, thermal, magnetic, sensor devices and catalysis due to their small dimensions and special properties such as high surface area to volume ratio and high heat transfer.

Nanotechnology is the most promising technology dealing with understanding and control of matter at nanoscale (i.e. dimensions between approximately 1 nm and 100 nm) and shape effect. The distinctive optical properties of the noble metallic nanoparticles (gold, silver and copper) have made them an irresistible prospect in electronics, optical devices, sensors, and catalyses [2]. Metallic nanoparticles with the unique optical and electrical properties have been widely investigated during the past decades. Among these, silver nanoparticles are the most intensively studied because of their unique properties and applications. However, several publications show that, size and morphology are the most essential parameters for the applications of nanoparticles. For example, significant use of nanoparticles is in drug release and drug targeting. It has been observed that particles size influences the drug release. Nanoparticles are more powerful since they give huge surface area to drug collaboration because of their small size. Form these two parameters we can add surrounding medium [3].

Copper nanoparticles received much attention due to its high electrical conductivity, high melting point (1085°C), low electrochemical migration behavior, excellent solderability and low material cost. Due to their unique catalytic and physicochemical properties, copper nanoparticles were employed in process like metal ion reduction, electrical and thermal conductivity [4]. Copper based nanoparticles find many applications such as, use in preparation of disinfectant for wastewater [5], high antibacterial activity against B. Subtilis [6], appropriate catalytic potential [7], dye reduction properties [8]. Furthermore, copper nanoparticles (Cu-NPs) coated drugs are extensively used to destabilize cancerous and tumorous cells [9].Recently, (Cu-NPs) have obtained much attention because of their applications in wound dressings and biocidal properties [10], potential modern utilizations, for example, gas sensors, catalytic process [11], high temperature superconductors and solar cells [12].

Zinc oxide, with its unique physical and chemical properties, such as high chemical stability, high electrochemical coupling coefficient, broad rand rang of radiation absorption and high photostability, is a multifunctional material [13-14]. In materials science, zinc oxide is classified as a semiconductor in group II-VI, whose covalence is on the boundary between ionic and covalent semiconductors. A broad energy band (3.37 eV), high bond energy (60 meV) [15] and high thermal and mechanical stability at room temperature make it attractive for potential use in electronics, optoelectronics and laser technology [16]. Zinc oxide, which exhibit a wide variety of nanostructures (spherical, nanorod, nanowire), possesses unique semiconducting, optical and piezoelectric properties [17]. Therefore, ZnO-based nanomaterials have been studied for a wide variety of applications such as nano-electronic/nano-optical devices, energy storage, cosmetic products, nanosensors, and biological labels, electrical and optical devices [18-19].

In recent years several authors have instigated the optical properties for metal or oxide metallic nanoparticles using experimental method, in order to understand the

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behaviour of the optical properties of nanoparticles when changing some of the parameters of size, shape, and environment. That's how, reflectance spectra for silver and copper studied by K. Shanks and al [20] demonstrated the environmental durability of silver and copper reflectors which have a very high solar reflectance. Thus, the reflectance of silver and copper deposited one glace is roughly 97% and between 40 and 97 % in 300 nm and 1700 nm, respectively. However, several results have shown that zinc oxide is very transparent. For example, experimental studies carried out by Ngom et al [21], showed a transparency rate of 87% for ZnO and tungsten doped. Another study of Qiong Zhou et al [22], shows that the zinc oxide are capable of absorbing a small amount of visible region (300-800nm) (86% transmittance).

In this manuscript, a simulation based on quasi-static, we have been developed to describe the optical properties of silver and copper nanospheres and in the following word we study zinc oxide nanoshells by using Mie theory to model the absorption spectra. We theoretically show the possibility of combining metallic nanoparticles with oxides for optical characterization. We also present the theoretical results and compare them with experimental results and discuss the limitations introduced by size and composition effects. The thickness and the composition were determined by Mie theory. We describe a multilayer Mie-like model that was developed to study the effects of spherically symmetric refractive index distributions on the scattering patterns of light scattered from a single cell. We also study the effect of composition.

Experimental

The basic multilayer sphere model for electromagnetic scattering was described in the literature long ago [23]. Enhancements to the model and to the computational algorithms are described in this article [24]. The basic model is depicted in Fig. 1. In figure 1, α_i is the Mie size parameter of the spherical shell which depends on radius R_i measured from the cell center and λ_i which is the wavelength of the radiation in the external medium. m_i is the refractive index of the respective layer. The simulations done in this study are based on the previous code developed in our articles [25] for calculating optical properties (absorbance, transmittance and reflectance) and for calculated the Mie effective absorption section in [26]. The simulation was done using the optical values (n, k) suggested by E. Palik [27], E. Palik [28-29] for silver, copper and zinc oxide, respectively, for complex refractive index of material and refraction index for medium.

By always placing in the quasi-static approximation and starting from Laplace's equations, we obtain the expression of the α-polarisability of the nanoshell configurations and it is given by the following expression [25] :

$$\alpha = 4\pi R_2^3 \varepsilon_0 \left\{ 1 - \frac{3\left[(m_2 - m_1) R_1^3 + (2m_2 + m_1) R_2^3\right] m_o}{2(m_2 - m_1)(m_o - m_2) R_1^3 + (2m_2 + m_1)(2m_o + m_2) R_2^3} \right\} \quad (1)$$

With m_o is the dielectric constant of surrounding medium, m_o=1,333, water refraction index. The dielectric function of the core is noted $m_1 = n_1^2 - k_1^2 + i2n_1k_1$ and the dielectric function of the shell is noted $m_2 = n_2^2 - k_2^2 + i2n_2k_2$ [30]

In the case of a multilayer sphere (particularly for core-shell system), the expression of the effective absorption cross section as a function of the dielectric functions of the three media or simply of the polarisability α is given by [31]:

$$\sigma_{abs} = \frac{2\pi}{\lambda \varepsilon_0} \operatorname{Im}(\alpha) \quad (2)$$

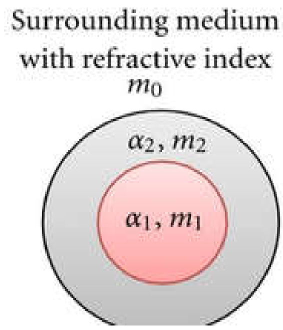

Figure 1: Core/shell configuration

Results and Discussion

Transmittance, reflectance and absorption of copper and silver nanoparticles

Figure 2 shows the results obtained for optical studies of both silver metal (dotted line curve) and copper metal (solid line curve) respectively as function of wavelength. These two materials were considered in a spherical shape, of radius equal to 40 nm, particles are deposited on a glass substrate and the surrounding medium is considered to be air. The results come from the model detailed in our article [25] where the absorption, reflectance and transmittance functions were detailed. First of all, it is essential to clearly state that these three optical properties describe how this varies with the size of the particles as demonstrated in our previous articles [32].The assumed wavelengths cover the visible range of 300-800 nm, although, in principle, the range can be extended to the ultraviolet and near-infrared wavelengths as well.

The study shows a well differentiated evolution for each optical parameter.

It can be seen that silver metal reaches its maximum transmission at 320 nm at a rate of 34.28% which is much higher than that of copper in the spectral range 300 to 500 nm, whereas from 500 nm onwards copper increases with a higher transmission rate than silver by about 14.05% at 580 nm (Fig. 2.B). Thus it is assumed that between 300-500nm silver particles transmit more than copper particles and between 500-700nm the reverse occurs.

Figure 2.B shows a high abrupt behaviour in between 300-400 nm for Ag-NPs. The same behaviour of the material is experimentally observed by the study carried out Wan Maisarah Hukhtar et al [33]. This very light signal between (35.3% transmittance) is probably due to excitation of the surface plasmon polaritons.

For reflectance (Fig 2.A), the evolution curve shows a simulated profile of the two particles in the spectral range 300-800 nm. The simulations clearly show that the silver particle has a higher reflection rate than copper throughout the entire study range, except in the 300-320 nm range where copper reflects more.

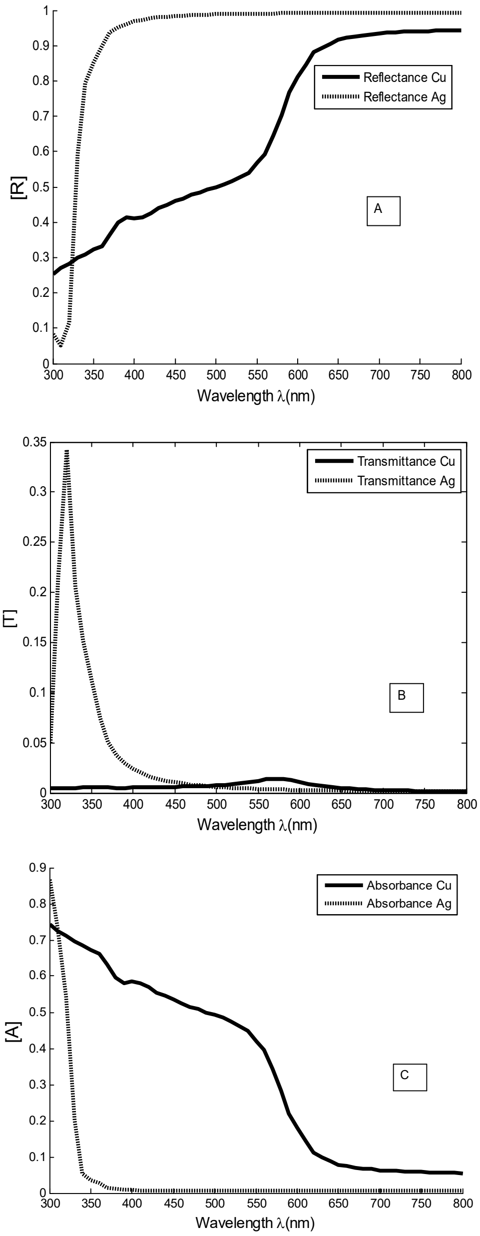

Figure 2: Optical properties of copper (solid curve) and silver (dotted line curve) film in air of (A) Reflectance, (B) Transmittance and (C) Absorbance

Following remarks can be made these curves. For most optical applications, high reflectance in the visible range is very important. All the deposited film in this work show high reflectance up to 97.3%. The same behaviour is observed for copper with a maximum reflection of 93.69% at 710 nm.

For the absorption phenomenon (Fig 2.C), it is clear that copper metal absorbs more than silver. It is observed that silver only absorbs between 300 and 390 nm, with a maximum of 86.64% at 300 nm. Beyond 390 nm, the behaviour of silver becomes linear with 0% absorption. For copper, the evolution is decreasing from 74.26% (300 nm) to 5.4% (800 nm).Finally, the simulations carried out show that silver particles reflects more than copper particles but less absorbs.

Transmittance, reflectance and absorption of zinc oxide

Either the same device as above, in which we consider spherical ZnO nanoparticles with a radius of 40 nm deposited on a glass substrate. The whole system is studied in air. Table 1 shows the maximum absorbance, transmittance and reflection values.

Table 1: Optical characteristic of ZnO

Material: ZnO	Transmittance	Reflectance	Absorbance
%Max	≈83%	27.48%	75.69%
Wavelength (nm)	[500-800]	380	300

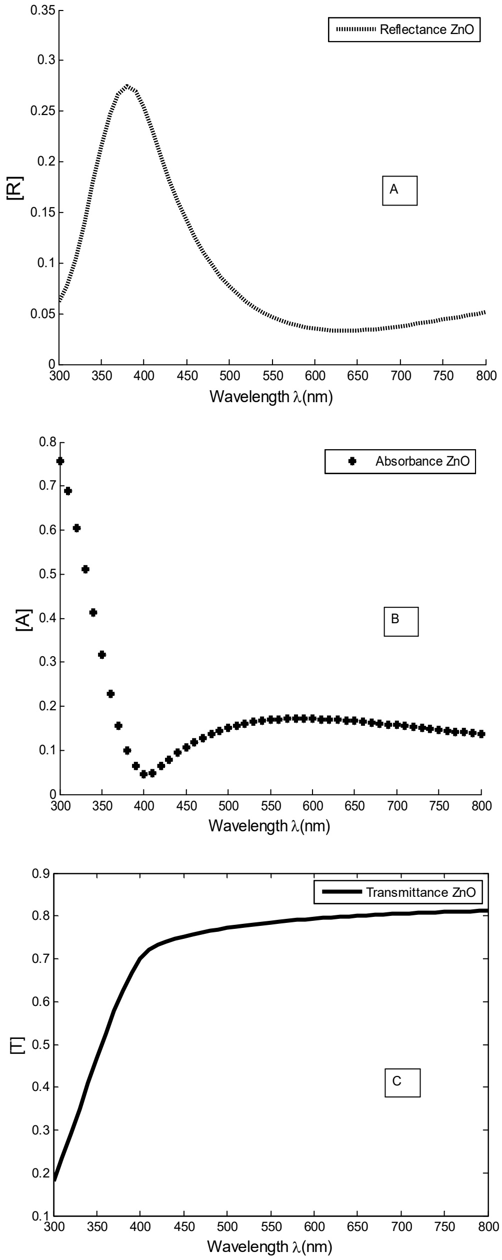

Figure 3: Optical properties of ZnO nanoparticles in air (A) Reflectance, (B) Absorbance (C) Transmittance

Table 1, like Fig. 3, clearly shows that ZnO is a highly light-transmissive material. However, depending on the behaviour indicated by the simulation results (Fig. 3), it is first of all necessary to clearly distinguish two important levels:

-From 300 to 400nm, ZnO can be considered an absorbent with about 75.69% at 300nm, Fig. 3.B.

-From 400 to 800 nm, ZnO has a high transmittance with a minimum of 69.92% at 400, Fig. 3.C.

From the theoretical simulation results, the transmission rate of zinc oxide was at 83% while the experimental value of the transmission obtained at 87%. This result was obtained by Ngom et al [21]. The shift in transmission rate from the theoretical value was to the formation of tungsten-doped ZnO. Zinc oxide is a semiconductor with a larg direct band gap of 3.37 eV, and the results showed that ZnO nanoparticles are transparent for the visible radiation (i.e. between 400 nm and 800 nm).

Optical properties of core/shell nanostructure : Copper coating on zinc oxide nanosphere

Figure 4(a), shows the dependence of the transmittance of the ZnO nanoparticles as a function of their sizes. Simulations were performed for different sizes (radius, r=10, 20, 25, 35, 45, 50, 100 nm) of ZnO nanoparticles deposited on glass substrate. The surrounding mediun is considered to be air. Results of the numerical simulation indicate that all spectral havesimilar evolution. It can be seen that the transmittance for ZnO is not dependenttoo much on the particle size. Above 420 nm, there is an overlapping of the curves and a slight downward shift between 400 and 420 nm as the ZnO size increases.

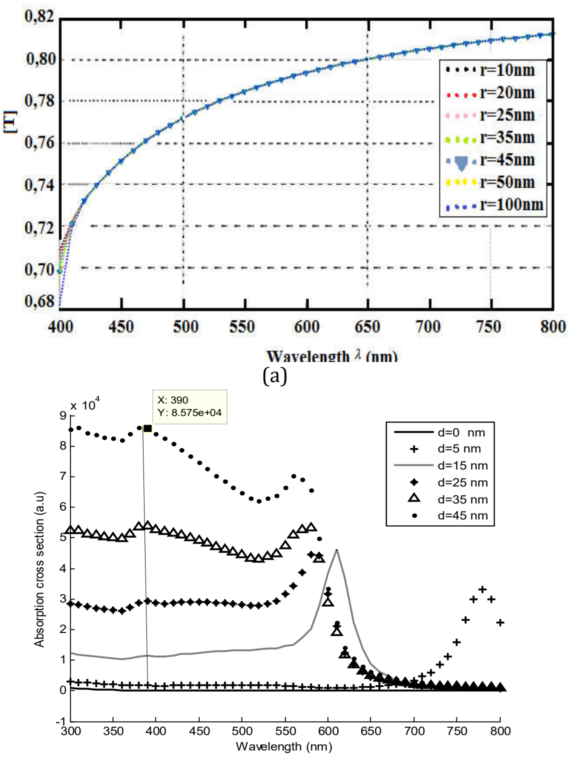

Figure 4: (a) Size effect on transmittance for Zinc oxide, (b) Absorption cross-section for the Zinc Oxide/Cu

Core/shell ZnO/Cu before and after Cu coating were generally spherical, because the Mie theory used in this work can only be applied to spherical structures. Evolution of the calculated absorption section spectrum of copper nanoparticles in zinc oxide at various copper size by equation (1) and (2).We have characterized the optical properties of these aqueous dispersions for ZnO/Cu core/shell nanoparticles by using matlab programation.The absorption peaks showed the characteristic signals for metal nanoparticles and these peaks are called as SPR peaks.We show the absorption spectra of aqueous dispersions of ZnO/Cu nanoparticles with different shell thicknesses (5, 15, 25, 35 and 45 nm). We have plotted the absorption spectrum for ZnO/Cu with zinc oxide cores were 30 nm radius and the nanoparticle was considered to be immersed in water (refractive index=1.33). It was assumed that the nanoparticle was illuminated by a plan wave source in order to excite all resonance modes of the particles within the visible and one part of infrared(IR) region spectrum.

Before coating, the zinc oxide nanoparticles presented any plasmon peak. After coating, Fig. 4(b) spectra show two peaks resulted from the ZnO/Cu nanoparticles, a characteristic surface plasmon peak were observed at red et near infrared region, respectively. Therefore, by increasing the size of the Cu-NPs, we can tune the surface plasmon resonance (SPR) into the near infra-refrared region from the visible range. Due to the surface plasmon band of ZnO/Cu is more sensitive to the shell size. Therefore the size of the copper is an important parameter for anothers application.Referencing to [34-35], this result show that, for potential application in SPR based biosensors, we'll have to use a size less than or equal to 15 nm.

Thus, numerically we have the results below : Firstly, surface plasmon resonance is located at 780 nm, 610 nm, 585 nm, 580 nm and 560 nm for the shell thichness equal to 5 nm, 15 nm, 25 nm, 35 nm and 45 nm, respectively. Secondly, in these results we could observe a particular pattern of peaks corresponding to the oscillation modes of colloidal copper nanoparticles based on the Mie theory. Theless intense peakis observed in the green spectral regionwas at 390 nm. The position of this peak was not sensitive to the change in coating thickness, while its intensity increased accordinly as thicker copper shells were formed.This last observation is conresponding for copper nanoparticle surface plasmon resonance peak and this result has reparted byJ. S. Duque and al [36]. It is seen from Fig. 4(b) that intensity of the red shift of surface plasmon resonance increase with increasing copper nanoparticles sizes.

This semiconductor (ZnO) possesses some complementary aspects and natural advantages like good transparency, which according to our study does not depend too much on its size, especially in the visible region from 420 to 800 nm. In the ZnO/Cu particles, the particles seem to be less and less resonant with increase of copper size, and therefore it is easy to say something about the average particle size from the ratio of the core radius (ZnO) to the shell thickness (Cu) for the expected applications. Quantitatively, surface plasmon resonance of ZnO/Cu nanoshell particle is localized after 390 nm.

Conclusions

In summary, a theoretical model to describe the effective absorption cross section by zinc oxide/copper nanoshell using Mie theory were studied and simulated. We presented in this work a complete study of the optical properties such as absorbance, transmittance, and reflectance of the silver, copper and zinc oxide nanoparticles. The copper deposed on ZnO has changed dramatically in their optical responses as compared to the bulk zinc oxide. The optical response of ZnO/Cu depends on the copper size. The tuning of SPR resonance towards the near IR-visible or visible region can make the zinc oxide core/shell nanoparticles as an ideal substitute for copper. Increasing size of copper nanoparticles deposited resonance band is shifted towards the shorter wavelengths in the vicinity of the characteristic band of the copper nanoparticle which is equal to 390 nm. The detailed study here has shown that zinc oxide is a very transparent material (more than 81%), copper is considered absorbent between 300 and 600 nm and reflective between 600 and 800 nm and finally silver metal is a very reflective material in the region studied between 300 and 800 nm.

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