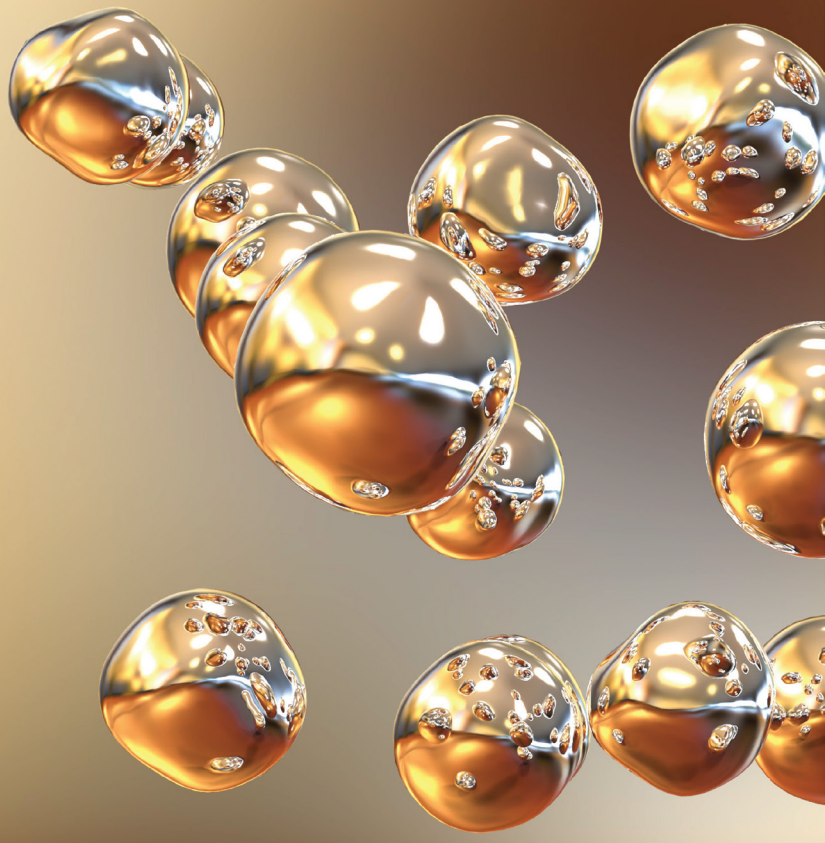
THE GREAT WORLD OF NANOTECHNOLOGY



Marcos Augusto de Lima Nobre

(Organizador)

VOL II

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PREFACE

The insertion of new and enhanced materials based on materials belonging to the Nano scale in the day-by-day has growth up in a silent way. In part, a number of works in the nanotechnology stemming of theoretical research using Density Functional Theory (DFT) and sophisticated simulation methods; another part is associated to the protected technologies associated to the military and patented nanomaterial and its process. In this sense, open access to recent aspects on the nanostructures application and properties can be reached in this book. Here, an interesting set of chapters gives opportunity of access texts that reach process and processing of nanostructures, applications of nanotechnology, advanced techniques to theoretical development. A broad set of nanostructures are here covered such as, nanocrystal, superficial nanograins, inner microstructures with nanograins, nanoaggregates, nanoshells, nanotubes, nanoflowers, nanoroad, nanosheets, Also, reveals new investigations areas as grainboundary of nanograins in ceramics and metals. A great number of software has been used as a tool of development of Science and Technologies for nanotechnology COMSOL Multiphysics 5.2. Phenomena and properties has been investigated by recent or classical techniques of materials characterization as Localized Surface Plasmon Resonance (LSPR), X-ray photoelectron spectroscopy (XPS), Field Emission Gun Scanning Electron Microscopy (FEG-SEM) with Energy Dispersive Spectroscopy (EDS), Raman Scattering Spectroscopy (RSS), X ray diffraction (XRD), ^{57}Fe Mössbauer spectroscopy, UV-vis spectroscopy, dynamic light scattering (DLS), Atomic Force Microscopy (AFM), and Field Emission Gun Scanning Electron Microscopy (FEG-SEM). In this sense, collections of spectra from Mössbauer spectroscopy, UV-vis spectroscopy and Infrared spectroscopy can be found. As a matter of fact, some chapter's item can be seemed as specific protocols for synthesis, preparations and measurements in the nanotechnology.

I hope you enjoy your reading.

Prof. Dr. Marcos Augusto Lima Nobre

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PLASMONIC RESPONSE OF GOLD- SILICA AND SILVER- SILICA METAL CORE NANOSHELLS BY OPTIMIZING THE FIGURE OF MERIT

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ABSTRACT: Plasmonic nanoaggregates have great potential for sensing applications, due to their spectral sensitivity response to the surrounding environment of the nanostructure and efficient extinction efficiency. A novel refractive index (RI) based SPR nano scale sensor which composed of metallic (Au/Ag) core and silica shell has been analysed theoretically. To simulate the

proposed geometry in radio frequency module, 3-D finite element method (FEM), an efficient tool to investigate the electromagnetic properties and electric field distribution surrounding the nanoparticle surface, based on commercially available software COMSOL Multiphysics were utilized. In FEM simulation, to determine the scattered field distribution and extinction cross section the nanostructure surface was divided into extremely small mesh elements which lead to finer size. The background field amplitude was fixed 1 Vm^{-1} and to restrain the simulation from any reflection, a perfectly matched layer was created around the nanostructure. We found dielectric metal nanoshells to be more absorbing with gold core compared to silver core, leading to high figure of merit (FoM) for Ag nanoshell 3.0, having the same silica shell thickness 5 nm. This result points out that our proposed multilayer geometry can be extended to other systems, enabling the plasmonic sensors have a high FoM over wide spectral ranges with phenomenal accuracy).
KEYWORDS: RI. FEM. COMSOL. FoM.

1 INTRODUCTION

The plasmonic metallic nanostructures have been extensively investigated substantial subject which stimulated the interest of the researchers due to their fascinating optical characteristics, conspicuous features linked with geometric tunability (M. Alejandro-Arellano et al., 2000), as well as advancements in particle synthesis which makes them valuable in the fields of controlled drug delivery, imaging, nonlinear optics and various biosensing modalities (Yang, X et al., 2015). An interesting phenomenon provokes when light interacts with noble metals nanostructures (Au/Ag) known as Localized Surface Plasmon Resonance (LSPR), i.e., the collective oscillations of the conduction electrons (Non- Propagating) prompted by electromagnetic radiation having a pertinent wavelength. In nanoscale regime, the resonances for Au/Ag exhibits within the visible region i.e.,400-700 nm (A. Maier, 2007).

It has been studied that Refractive index (RI) based sensitivity and figure of merit, (defined as the ratio of sensitivity to the linewidth) (Jéssica et al 2021), depends on the geometry of the Au nanoparticles (Klantsataya. E et al., 2015; Z Yang et al., 2008). For single particle LSPR sensor, the fundamental principle utilizes the fact that LSPR spectrum position varies as a function of dielectric host medium (A.D. McFarland et al., 2003; Klantsataya et al., 2015) revealed the fact narrow full width at half maximum (FWHM) will result in the enhancement of LSPR sensing behavior.

In this study, we evaluate the Au/Ag nanoshell based on gold/silver core encapsulated by silica shell having nano scale varying thickness likewise their tune ability linked with surface plasmon resonance were also theoretically investigated. The simulations were carried out by Finite Element Method (FEM) based on partial differential equations commercially available software COMSOL Multiphysics 5.2, which is an

efficient tool to explore the electromagnetic properties as well as electric field distribution on the surface of nanostructures having various shapes to predict the unprecedented dependence of the plasmonic properties of core-shell nano scale structure (Du. C, M et al., 2014). The background field amplitude was fixed 1 Vm-1 and to restrain the simulation from any reflection, a perfectly matched layer was created around the nanostructure.

2 LSPR MATERIAL DEPENDENCE

The fundamental optical characteristics of metal nanoparticles may be predicted calculating their size-dependent dielectric function. The dielectric function value of a metal is influenced by its structure. As the metallic nanoparticle becomes smaller, electron scattering on the NP surface becomes more pronounce, therefore affecting the dielectric function value of the material. The complex dielectric function $\epsilon(\omega)$ can be described by Drude model, which also account for the conduction electrons scattered by the nanoparticle surface Therefore, dielectric function can be written as[1]:

$$\epsilon(\omega) = \epsilon_{inter}(\omega) + \frac{\omega_p^2}{\omega(\omega+i\gamma)} \quad (1)$$

Where ϵinter(ω) depicts interband transitions, ωp represents plasmon frequency and γ is a phenomenological scattering parameter. The plasma frequency can be expressed for bulk metal by:

$$\omega_p = \left(\frac{Ne^2}{\epsilon_0 m_e}\right)^{1/2} \quad (2)$$

Where me is the effective mass of an electron, N is the number density of free electrons, e is the charge of an electron and ϵ_0 dielectric function of free space (vacuum). For nanoparticles, the scattering parameter has inherent contributions of the intrinsic properties of the material as well as from interface scattering and therefore, it can be described as $\gamma = \gamma_{bulk} + \gamma_{scat}$.

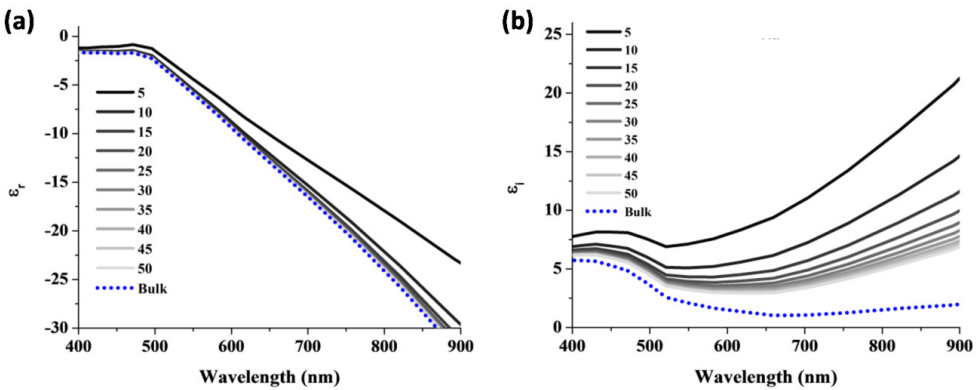
Interface scattering becomes significant when the effective electron path length L_{eff} is larger than to the nanoparticle itself (L.J. Mendoza et al 2014). The effective path length for convex shapes particles, for instance sphere, rods, cubes etc., is expressed as $L_{eff} = 4V/S$ where V is the volume and S is the surface area (Ross et al. 2015). Thus, the scattering parameter is given by $\gamma_{scat} = AV_f/$Leff where A is scattering efficiency (L.J. Mendoza et al 2014). For gold, γ_{bulk} = 1.07x10^{14} s^{-1}, V_f = 1.40x10^6 ms^{-1}, while for silver γ_{bulk} = 3.22±1.22x10^{13} s^{-1}, and V_f= 1.39x10^6 ms^{-1} **(**P.B Johnson 1973**)**. Table 1 depicts the radii of spheres and effective electron path lengths with respect to their sizes.

Table 1: Nanospheres radii and their corresponding effective electron path lengths (L_{eff}).

Sphere Radius (nm)	L_{eff}
5.0	6.67 nm
10.0	13.34 nm
15.0	20.00 nm
20.0	26.67 nm
25.0	33.34 nm
30.0	40.00 nm
35.0	46.67 nm
40.0	53.34 nm
45.0	60.00 nm
50.0	66.67 nm

The figure 1 depicts the dielectric functions of Au materials, considering size correction, showing that decreasing the nanoparticle radius (effective path length) both real (ϵ_r) and imaginary parts (ϵ_i) of metallic dielectric changes. The real part of dielectric function depicts the pattern of electron polarization in the medium as a consequence of incident field and determines LSPR peak position.

Figure 1. Complex dielectric function based on surface scattering of real (a) and imaginary part (b) of gold materials with different radii (5, 10, 15, 20, 25, 30, 35, 40, 45, 50 nm).



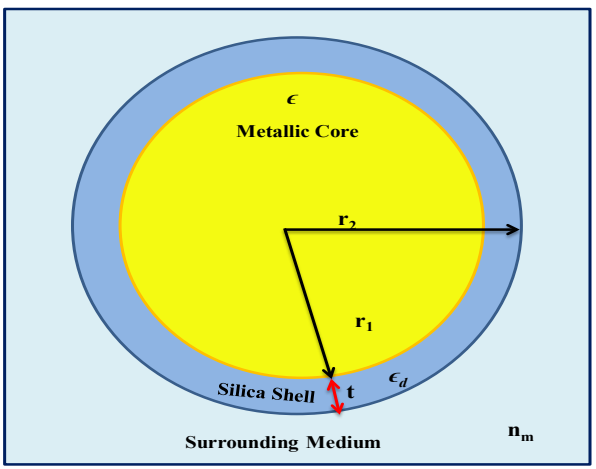
However, imaginary part ϵ_i describes energy dissipation or loss in the materials. Decreasing of the nanoparticle size leads to a lower polarizability of the metal, while increases the magnitude of ϵ_i Figure.1 (b). Therefore, higher loss is expected for metallic particle with 5nm radius than to 50 nm radius nanostructures due to surface scattering. One can notice that on decreasing effective path length (particle radius), both real and imaginary parts of $\epsilon(\omega)$ are affected. Real part of $\epsilon(\omega)$ Figure. 1(a) becomes less negative, showing that metals are less polarizable, while increase in magnitude of imaginary part

Figure.1 (b) of $\epsilon(\omega)$ causing increase in overall losses in the NP. In particular, these effects yield overall poor plasmonic metal.

3 MODELLING AND CALCULATIONS

Figure 2 illustrates the schematic geometry of Au/Ag silica coated nanoshell being investigated in this work. The inner most region is attributed as core having radius and dielectric function r_1 and ϵ respectively. The second portion represents the shell with radius r_2 and dielectric function ϵ_d, where shell thickness is given by $t = r_2 - r_1$.

Figure. 2. Schematic model of the gold/silver silica coated nanoshell.



The interaction between core-shell based metallic nanostructures and electromagnetic radiation in this study was ascribed by Mie theory which elucidated well the solution of Maxwell's equation with appropriate boundary conditions. The general solution for the potion in each region (i= 1, 2, 3) is expressed as in Eq. 3, (Erickson, T. A. and J. W. Tunnel, 2009):

$$V_i = \left[L_i r + \left({}^{M_i}/_{r^2}\right)\right] \cos(\theta) \qquad (3)$$

Where radius r, angle θ and coefficients L_i, M_i can be calculated by satisfying the boundary conditions (Averitt, R. D et al., 1999). When the dielectric function is geometrically tunable, the effect of electron scattering can be interpreted by practicing the modified bulk collision frequency as expressed in Eq. 4:

$$\Gamma = \Gamma_{bulk} + \frac{Av_f}{m_f} \qquad (4)$$

Where Γ_{bulk}, m_f and v_f are collision bulk frequency, electron mean free path and Fermi velocity respectively. In case of single refractive scattering and for simple Drude model A = 1(Averitt, R. D et al., 1999). For nanoshells, the bulk dielectric function can be modified to analyse the free electrons scattering, which is given by Eq. 5:

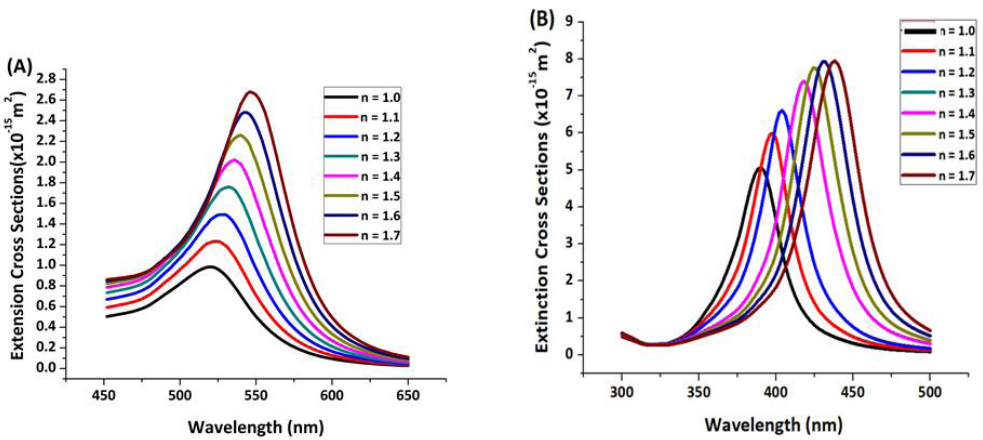
$$\epsilon(m_f, \omega) = \left(1 - \frac{\omega_{bp}^2}{\omega^2 + i\omega\Gamma}\right) + \epsilon(\omega)_{int} \qquad (5)$$

Here, the second term represents the Interband transitions, ω_{bp} bulk plasma frequency and ϵ (m_f, ω) is dielectric function (size dependent).

4 RESULTS AND DISCUSSION

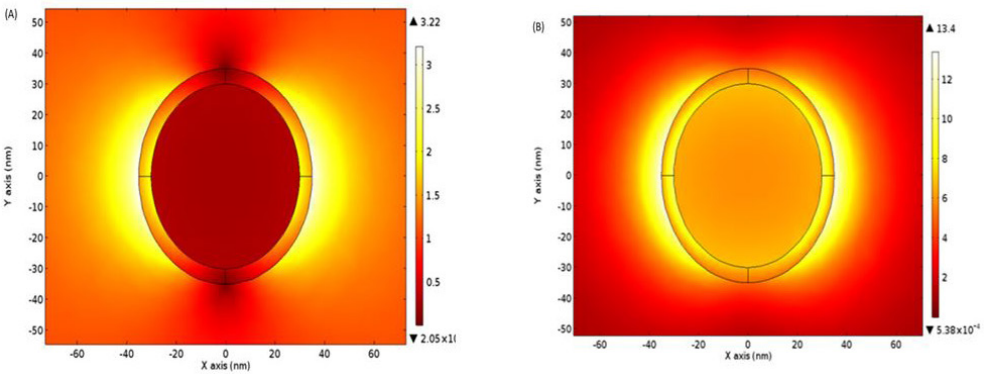
In this probed model, our focus is on the optimization of the figure of merit by varying the refractive index of the surrounding medium. Despite their simplest core- shell based structure, these metallic nanostructures can still display refractive index tunability based on their LSPR effect. Fig. 3 (A & B) depicts the response of the λ_{LSPR} and extinction spectra of Au/Ag@SiO_2 nanoshells to the refractive index of the neighbouring medium respectively. For both nanoparticles, by varying the RI the extinction varied significantly in the visible regime and exhibits red shifts which obviously results increase in the intensity of the extinction peak. In case of Au@SiO_2 shell, the extinction spectra continued to red shift in the visible region and the peak wavelength approaches 540 nm. On contrary, the extinction peak of Ag@SiO_2 shell initially blue shifted near UV regime and eventually the peak wavelength was recognizable around 430 nm in the visible region.

Figure. 3. The LSPR extinction for gold (A) and silver (B) nanoshells



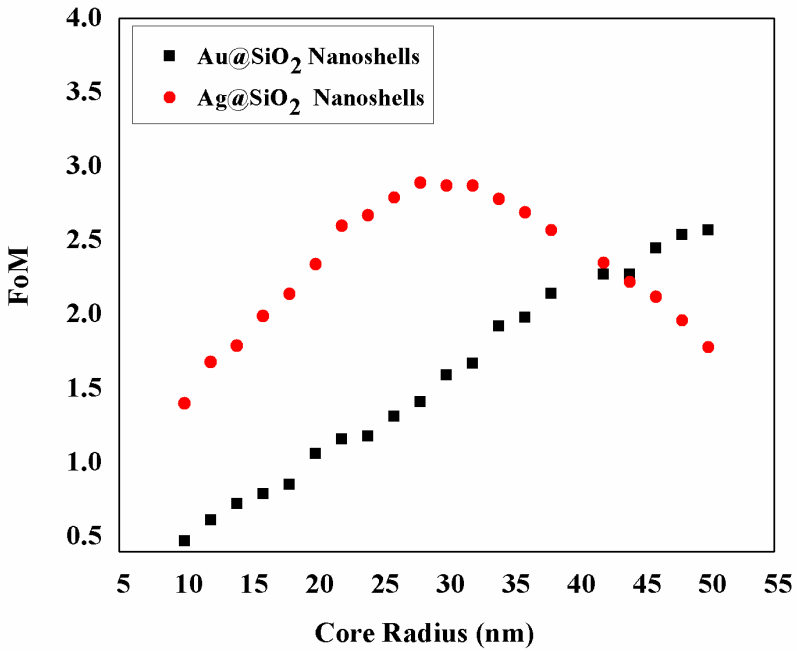
The E-field profile around the nanoshells for the dipole mode (gold and silver) can be observed in Figures 4 A) and 4 (B) respectively. The changes in normalized electric field of nanoshells were compared at the LSPR wavelengths.

Figure. 4: Electric field profile for gold (A) and silver nanoshells (B).



For linear polarization, the electric field distribution is expressed by the two lobes which ensure the influence of plasmon dipole. LSPR peak position can be determined not only by the real part of nanostructure dielectric function, but also by the RI of the surrounding medium. In refractive index based LSPR sensors, bulk sensitivity reflects how LSPR peak position varies by changes in the RI of the local medium.

Figure. 5. Figure of merit for gold and silver nano shells



The effects of NP core radius on the FoM determining nanoparticle sensing efficiency with varying radii from 10 - 50 nm were theoretically analysed as depicted in figure 5. As the Ag nanosphere radius increases, the FWHM increase due to radiation damping factor (Hu, H., Novo et al., 2008). Therefore, FoM of Ag nanoshells decrease as the particle size grows, as shown in Figure 5. Albeit, the ε_i value of Ag dielectric function is less than that of Au across visible region, therefore less damping occurs, resulting narrow FWHM (Figure 2(A) and Figure 2(B) and high values of FoM for silver particles. The calculated value of FoM (3.0) of silver nanoshells (r = 30 nm) is higher than the reported values of more complex shapes, such as single Au nanorod (1.3) (Mayer, K.M et al., 2008), Au nanostar (1.9) (Nehl, C.L et al., 2006), Au pyramid (2.2) (Nehl, C.L et al., 2006) Ag nanocube (1.6) (Sherry, L.J et al., 2005).

5 CONCLUSION

The theoretical results indicated a nonlinear behavior of the bulk and refractive indexed based sensitivity as function of the varying shell thickness. Moreover, LSPR peak shift is also determined by changing surrounding medium and the shell thickness. Our results possess high figure of merit for silver nanoshell (3.0) as compared to gold nanoshell (2.50) having silica shell thickness 5 nm for each nanostructure. The proposed approached can be extended to engineer the efficiently use of different nanostructures on molecular biosensing.

6 ACKNOWLEDGEMENT

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amorphous composite based on carbon/nanoparticles and Semiconductor Ferroelectrics. Member of the Program of Post-Graduation in Chemistry at UNESP - Campus of São José do Rio Preto, IBILCE UNESP – SP, Brazil.

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