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Enhancement of localized surface plasmon resonances of silver nanoparticles array upon the presence of graphene coatings: LSPR biosensor

Mohamed El Barghouti^{1,*}, Abdellatif Akjouj² and Abdellah Mir¹

¹Laboratory of Advanced Materials Studies and Applications (LEM2A), Physics Department, Faculty of Science, Moulay Ismail University, B.P. 11201 Zitoune, Meknes, Morocco

²Institute of Electronics, Microelectronics and Nanotechnology, UMR CNRS 8520, Lille University, FST, Department of Physics, 59655 Villeneuve d'Ascq, France

E-mail: **Corresponding author.* Mohamed.elbarghouti@gmail.com

Abstract. We study, in this article, the resonance of localized surface plasmon (LSPR) on silver nanoparticles (AgNPs) deposited on a glass substrate and coated with a graphene. The resonance peaks in the absorption spectra of the nanostructure SiO_x/AgNPs/Graphene, show significantly different profiles such as the thickness of the graphene, and the refractive index of the surrounding environment which is, gradually, varied. Numerical simulation reveals that movements in plasmon resonance peaks result from coupling between the AgNPs networks and the covering graphene layer. In addition, the LSPR modes shift to red from 412 nm to 548 nm when the thickness of the graphene layer deposited on silver nanoparticles changes from 0.34 nm to 9 nm. Light coupled with LSPR modes propagating along a AgNPs-graphene interface is calculated and compared to a conventional AgNPs-based LSPR biosensor. The result of the comparison depicts an improvement in the sensitivity of the biosensor from 26 to 106 nm/RIU.

1. Introduction

Optical properties of metal nanoparticles (Ag or Au) and their addressability via spectroscopic techniques have put them in the forefront of fundamental and applied research fields, with applications ranging from nanoparticles array wave guiding to biosensor designs [1-3]. Localized surface plasmonic resonance biosensors (LSPR) are the most advantageous biosensors thanks to their ability to provide biomolecular detection with higher sensitivity, mainly because they eliminate the time-consuming marking process that reduces molecular binding disturbance [4,5]. Understanding their properties is significantly more complex when they are deposited on a substrate, or when they interact [6,7]. It is quite clear from already available theoretical and experimental studies (on metallic nanoparticles) that several parameters do govern their behavior [8,9].

Spectacular progress on the control of optical properties of graphene material sheets stimulated the search for new types of two-dimensional materials. Graphene considered as a very promising new kind of material with possible applications in several domains (including bio-plasmonics). It's well established that AgNPs strongly absorb within narrow frequency bands in the visible range as their localized surface plasmon resonances get excited. Besides, the effect of the dielectric environment has been widely studied in the Refs [10-12]. A part of



this work consists of investigating the influence of graphene layers (whose refractive index is n_g [13,14]) on AgNPs optical properties. Graphene has aroused considerable interest in recent years because of its distinctive electrical and optical properties [15,16], a highly sensitive LSPR biosensor using the graphene layer to improve the sensitivity caused by the light absorbed by the graphene [10,17].

This is performed in the case when the graphene layers are either deposited on the nanoparticles array. Specifically, we will examine the effect of varying progressively the thickness of graphene layers from 0.34 nm to 9 nm. Different dielectric layers of refractive index n_2 are deposited on top of AgNPs/graphene, which are in turn sitting directly or indirectly on a substrate made of SiO_x (n_1). We study numerically how graphene layers are deposited; those are $\text{SiO}_x/\text{AgNPs}/\text{graphene}$. The corresponding geometry is well illustrated in Fig. 1. The incident light (along the y-axis) wave whose electric field is polarized along x-axis is also highlighted. Throughout the paper, the geometrical parameters of the structure will be fixed such as the particles length is $l = 25\text{nm}$, $a = 70\text{nm}$ is the lattice parameter (along x-axis), and $h = 20\text{nm}$ is the nanoparticles height.

Since graphene has important properties, to absorb the biomolecular and, thus, allows a significant change in the refractive index of the graphene-AgNPs interface to protect the structure, which avoids the oxidation of nanoparticles Ag [2]. The numerical results show that, compared to graphene in an LSPR substrate based biosensor/AgNPs, graphene in an LSPR substrate based biosensor/AgNPs/graphene offers a 304.60% higher sensitivity for nine graphene layers.

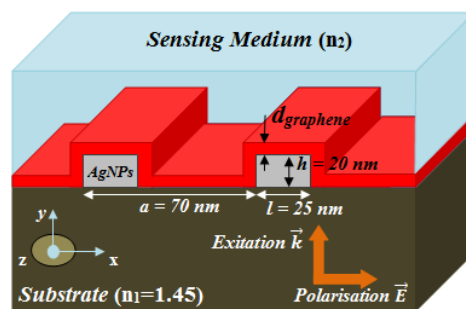


Figure 1. Schematic representation of AgNPs when covered with few layers of graphene and another dielectric lying on top (detection medium of refraction index n_2), and deposited on a SiO_x substrate ($\text{SiO}_x/\text{AgNPs}/\text{graphene}/\text{dielectric}$).

2. Analysis and modelling part

2.1. Simulation model

The optical properties of silver nanoparticles (AgNPs) are determined numerically within a wavelength domain ranging from UV to near IR, specifically, [300 nm-800 nm]. Simulations are performed using Finite Element Method (FEM) [18]. Actually, the simulation method has already been well documented in Refs [19,20]; it focuses on solving Maxwell's equations in the frequency domain by discretizing space in small finite elements (having various possible shapes such as triangular, tetrahedral (3D) etc.). Our calculation is carried out in a two-dimensional (2D) domain with Ox and Oy axes oriented as shown in Fig. 1.

The configuration drawn in Fig. 1 that consists of silver nanoparticles (having length, l , height, h and interparticle distance, $a-l$) deposited on substrate (of refractive index $n_1=1.45$), and coated with a graphene layer of the refractive index n_g . This last interface is furthermore covered by a non-absorbing medium of refractive index n_2 such as air and water. For the

calculations, the following geometrical parameters were chosen: $l=25$ nm, $h=20$ nm, $a=70$ nm. This corresponds to an average metal coating of 37% [21].

An incident plane wave is generated within SiO_x substrate of refractive index $n_1 = 1.45$, and is launched along the Oy axis (direction of propagation). The plane wave is TM polarized and its associated electric field vibrates along the Ox axis. The calculation is meant to simulate an infinite array of nanoparticles along Ox axis, and the structure is supposed to be infinite along the direction of Oz. The space discretization is realized by means of free triangular mesh and such that the maximum size of elements is adjusted depending on the material considered. In all dielectrics (layer on top of AgNPs and SiO_x substrate), the elements size is no more than $(\frac{\lambda_0}{2n})/5$, where n is the index of SiO_x or water/air. In Fig. 1, is sketched the simulation models in the considered configuration, that is, when nanoparticles are covered with thin layers of graphene. The frequency-dependent complex permittivity of silver (Ag) metal is described by the Lorentz-Drude model [22-24]:

$$\varepsilon(\omega) = \varepsilon_{r,\infty} + \sum_{m=0}^M \frac{f_m \omega_p^2}{\omega_m^2 - \omega^2 + j\omega\Gamma_m} \quad (1)$$

where $\varepsilon_{r,\infty}$ is the relative permittivity at infinite frequency, ω_p the plasma frequency, and ω_m , f_m and Γ_m are the resonance frequency, the strength (weight) and the damping rate of the m^{th} oscillator, respectively. The Lorentz-Drude model uses M damped harmonic oscillators to describe the small resonances observed in the metal's frequency response. The value of the dielectric constant at infinite frequency ($\varepsilon_{r,\infty}=1$) and all the other parameters of L-D model are taken from reference Ref. [23].

The graphene deposited between the AgNPs and the detection medium to increase the macromolecular absorption capacity of the biosensor (fixation of the biological element). The thickness of the graphene is varied as $d_{\text{graphene}} = L \times 0.34$ nm, where L is the number of graphene layers and its complex refractive index (n_g) in the visible range is an absorbent and dispersing material, the refractive index of which is given by the following Eq. (2) [13]:

$$n_g = 3.0 + i\frac{C_1}{3}\lambda \quad (2)$$

The constant $C_1=5.446 \mu\text{m}^{-1}$ [16], and λ is the wavelength of the incident light in μm . The refractive index of the detection medium is chosen as $n_2 = 1.00$ or 1.333 .

2.2. Principle of LSPR sensing

When the dielectric properties of the local environment are modified by the adsorption of analytes to be analysed, there will be a peak wavelength of the LSPR observed shift to red in the absorption spectrum (Fig. 3). Since the refractive index (RI) is related to the dielectric properties, LSPR biosensors can detect the RI changes in the local environment caused by the adsorption of substances to be analysed. The correlation between the peak wavelength displacement ($\Delta\lambda_{\text{LSPR}}$) and the RI change (Δn_2) is illustrated as follow:

$$\Delta\lambda_{\text{LSPR}} = S\Delta n_2 \quad (3)$$

where S is the refractive index sensitivity of the interface, Δn_2 is the variation of local refractive index of the detection medium [25]. The refractive index sensitivity in nm/RIU (change of nanometer per refractive index unit) is thus defined as,

$$S = \frac{\Delta\lambda_{\text{LSPR}}}{\Delta n_2} \quad (4)$$

3. Results and Discussion

In this work, we have studied the detection potential of a 2D structured network of Ag nanoparticles. Through a detailed study using the FEM method, we showed the plasmonic response of the structured presented in Fig. 1. The theoretical performance comparison between structured biosensors and conventional biosensors using a purely propagatifs plasmon, suggests to the importance of nanostructure in enhancing the sensitivity not only in terms to the response of a biosensor without a graphene but also in relation to the impact of nano-structuring of the surface which generates only a loss of performance. We have found that the prevalence of LSPR modes can lead to an increase in terms of sensitivity to a deposit of a graphene layer, while being sensitive to variations of the d_{graphene} with the incidence wave normal to the substrate and resulting in a gain of 304.60%.

In order to determine the effect of graphene layer thicknesses on LSPR of the hybrid nanostructure, the absorption spectrums of the glass/AgNPs/graphene with different graphene layers thicknesses are plotted in Fig. 2. The refractive index (n_2) of the surrounding environment is set to $n_2 = 1.333$ and the graphene layer thickness varies from 0.34 to 9 nm. Each nanostructure presents the plasmonic resonance mode in the absorption spectrum.

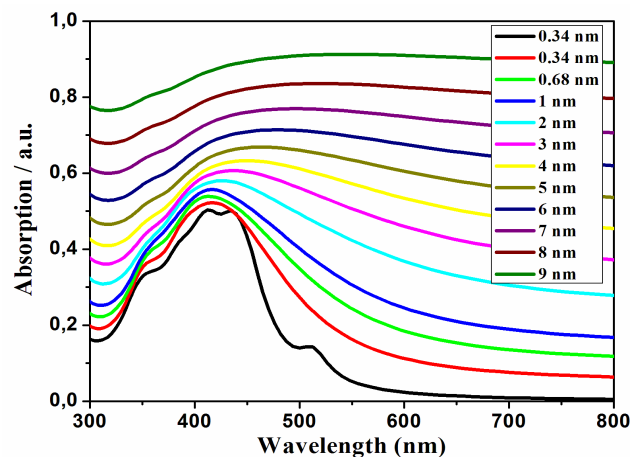


Figure 2. Absorption spectrum of the nanostructure depicted in Fig. 1. The 12 spectrums are given for different values of graphene thickness varying from 0.34 nm to 9 nm.

Numerical results reveals that movements in plasmon resonance peaks result from coupling between the AgNPs networks and the covering graphene layer. In addition, the LSPR modes shift to red from 412 nm to 548 nm. Giving a very significant increase in the resonance wavelength of 136 nm, when the thickness of the graphene layers deposited on Ag nanoparticles varies from 0 to 9 nm.

Specifically, the LSPR in the glass/AgNPs/graphene nanostructure is the convolution of resonance mode; the red-shift is likely due to different dielectric environments probed on the opposite faces of the nanohybrids when the d_{graphene} is varied from 0.34 to 9 nm. It is further believed that the red-shift comes from charge transfer between the graphene layers and Ag nanoparticles array. Therefore, the plasmon resonance on the glass/AgNPs/graphene nanostructures can be tuned by changing the thickness of the graphene layers [10,26].

As the refractive index (RI) of the sample growing, the wavelength also increases. We investigate the sensitivity of the Ag nanoparticles LSPR biosensor in the bio-substances RI range of 1.00-1.333, and the corresponding linear fitting curve of the resonance wavelength with respect to the refractive index is shown in Fig.3a.

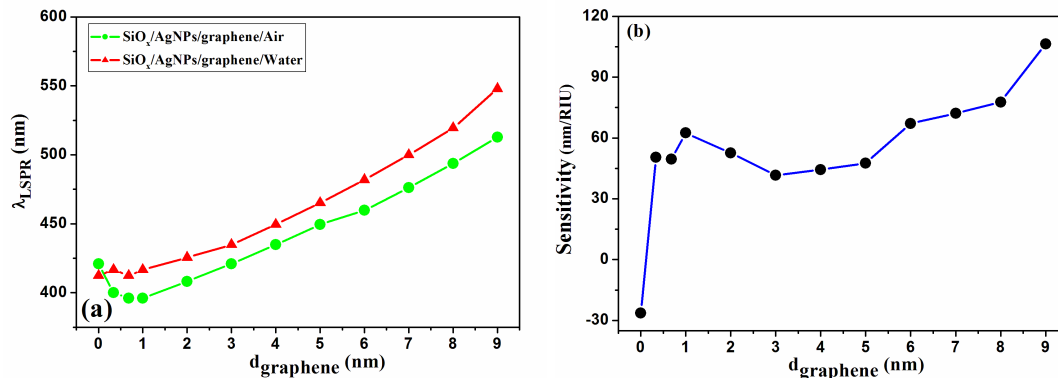


Figure 3. (a) Evolution of the wavelength of LSPR versus the thickness of graphene (b) Variation of the sensitivity versus $d_{graphene}$.

Sensitivity investigation was conducted for graphene layer and biomolecules instead of conventional relatively large number of biomolecules thickness approach without graphene. The sensitivity improved by 304.60% (4.04 times) when the pure AgNPs conventional biosensor was covered with graphene layer of the $d_{graphene} = 9$ nm.

In this situation, the shift in resonance wavelength (λ_{LSPR}) for the background nanostructure, and increases in a same method for both graphene layer enrichment. This phenomenon can also be seen in Fig. 3a, the shift in resonance wavelength to red, depending on the thickness of dielectric graphene layer ($d_{graphene}$).

Fig. 3b shows that calibration plot of the sensitivity changes with the thickness of graphene. Several numerical simulations were carried out to find how sensitivity is related on the thickness of graphene filed of the nanoparticles. This hybrid nanostructure offers the thinnest design and the better sensing performance than both metal and based approaches, in particular the sensitivity improvement of this design that of pure Ag nanoparticles biosensor. Fig. 3b shows the biosensing performance of the proposed glass/AgNPs/graphene hybrid systems with various graphene layer thicknesses. As indicated, the proposed nanostructure can operate in the visible range, and the sensitivity of detection increases with the thickness of the graphene layer. It is observed, the sensitivity changes when going from $d_{graphene} = 0$ nm to $d_{graphene} = 9$ nm.

Adding that in the case of depositing thinner thickness of graphene, there is a strong shift towards the red resonance. Furthermore, it was further explained that the sensitivity of nanosensors based on this phenomenon is related to the shifting of the resonance wavelength of the plasmon response of the nanoparticles with $d_{graphene}$, that is to say in the regions of d where the variation of the $\Delta\lambda_{LSPR}$ is large, the sensitivity is optimal. This indicates a greater effectiveness of bio-detection with these structures.

In a second phase, several numerical simulations were carried out to determine how the sensitivity changes according to the thickness of graphene deposited on the nanoparticles. The results obtained appear in Fig. 3. First, an increase in sensitivity is detected for graphene thicknesses between 0 and 1 nm for nanostructure. In fact, (Fig.3b) $S = 26.30$ nm/RIU for $d_{graphene}=0$ nm and $S = 62.50$ nm/RIU for $d_{graphene}=1$ nm, corresponding to an increase of about 137.60%. Then the sensitivity decreases from $d_{graphene}=1$ nm, to reach 41.61 nm/RIU when $d_{graphene}=3$ nm.

On the other hand, for ($d_{graphene}$), either when the nanoparticles are covered by different graphene slips $3nm \leq d_{graphene} \leq 9nm$ or when the AgNPs are covered with 27 layers of graphene ($d_{graphene} = 9$ nm), the resonance wavelength is $\lambda_{LSPR} = 512.82$ nm for $n_2=1.00$ and $\lambda_{LSPR} = 547.94$ nm for $n_2=1.333$, which corresponds to a sensitivity of $S = 106.44$ nm/RIU.

First of all, there has been an increase in sensitivity (glass/AgNPs/graphene) for thicknesses of graphene between 3 and 9 nm. Indeed, $S = 41.61$ nm/RIU for 3 nm and $S = 106.44$ nm/RIU to 9 nm.

Fig. 4 shows the electric field strength of the nano-system at different thicknesses from the graphene layers to the surface plasmon resonance.

The excited electrical field distributions on plasmon resonance, at a normal incidence structures, are represented by cards of electric field in Fig. 4, which indicates that this mode is a dipolar localized surface plasmon, whose hot spots (areas of high field strengths) are pushed to the top and lower corners of the AgNPs of the structure lacking the graphene layer (see Fig. 4a), and to lower corners of higher AgNPs graphene layer.

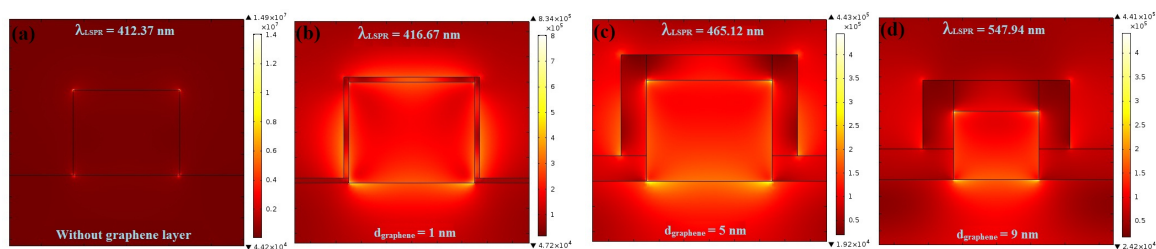


Figure 4. Maps of electric field in glass/AgNPs/graphene structure for $d_{\text{graphene}} = 0\text{nm}$ under monochromatic incident at 412.37nm (a), for $d_{\text{graphene}} = 1\text{nm}$ at 416.67nm (b), for $d_{\text{graphene}} = 5\text{nm}$ at 465.12nm (c) and for $d_{\text{graphene}} = 9\text{nm}$ at 547.94nm (d).

AgNPs/graphene based sensor exhibits efficient charge transfer due to low optical absorption efficiency (about 2.3%) of graphene [27]. Generous electron transfer from graphene to Ag nanoparticle will occur in case of optical excitation as the work function of these materials are related as graphene (4.5eV) < Ag (4.74eV) [28,29]. This process will lead to a larger electric field enhancement at the sensing inter-face thereby resulting in a higher sensitivity to the target biomolecules [30].

For the nanostructure $\text{SiO}_x/\text{AgNPs}/\text{graphene}$ in [Fig. 4b, c and d] the fields are, basically, limited to the space between the graphene layer and the dielectric of detection which are lower than the AgNPs. It can be seen by comparing Fig. 4a that when the nanoparticles without graphene layer have hot spots at each of their corners.

Moreover, adding 1 nm of graphene has resulted in an increase in the amplitude of the electric field in the interparticle space followed by a decrease in the peak value intensity. This observed decrease in field strength is due to the coupling between the plasmon mode and the Fabry-Perot cavity modes connected to the presence 5 and 9 nm of the graphene layer and sensing medium presented above the Ag nanoparticles [2,30].

The absorption spectra of the glass/AgNPs/graphene nanostructure for different refractive indexes of the surrounding medium are shown in Fig. 5a. A series of refractive indexes $n_2 = 1; 1.333; 1.45; 1.6; 2; 2.5$ and 3 are used in the calculation.

The red-shift in the plasmonic mode from 449.44 to 597 nm bears a positive relationship with the refractive index between 1 and 3. This variation is due to the change in the refractive index of the surrounding medium around the glass/AgNPs/graphene nanostructure, resulting in changes in the resonance condition and location of the resonance modes [5,10]. The geometric parameters of the Ag nanoparticles are: $l = 25$ nm, $h = 20$ nm, $a = 70$ nm and $d_{\text{graphene}} = 5$ nm.

This phenomenon can also be seen in Fig. 5b, the change in resonance wavelength shifts to the refractive index of the sensing medium (n_2). Fig. 5b shows that calibration curve is linear, and graphene/AgNPs hybrid nanostructure provides the thinnest design and the better sensing

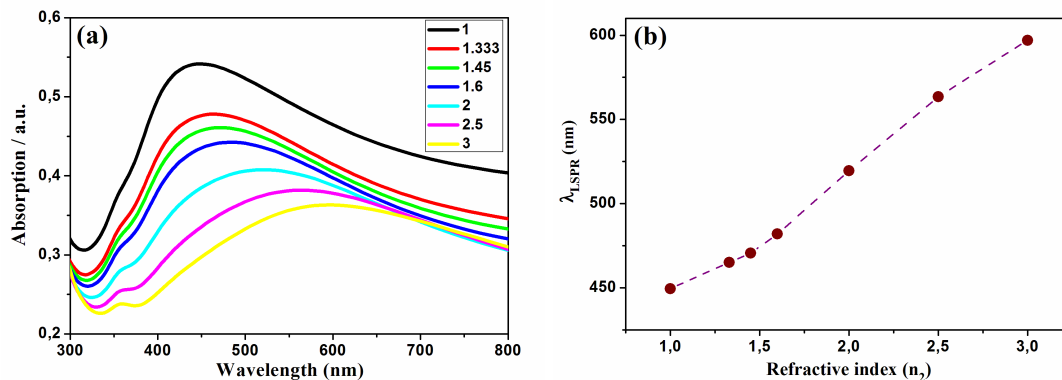


Figure 5. (a) Absorption spectrum corresponding to the structure glass/AgNPs/graphene, for defeated refractive index of the detection medium. (b) Evolution of the resonant wavelength of the LSPR versus refractive index of the detection medium (n_2).

performance than both metal and graphene based approaches, in particular the sensitivity enhancement of this design is 4.04 times larger than that of pure metal nanoparticles biosensor.

4. Conclusions

This paper presents the designing of graphene coated two types of biosensor. The wavelength interrogation mode for setting up localised surface plasmon resonance condition is used to analyze the proposed LSPR biosensors. The spectral study to detect for the biological elements using a compatible sensitive medium. Silver nanostructures (AgNPs) on the glass and covered with a thin dielectric layer of graphene, indicates that this study allows a better insight of systems based on resonant metal nanoparticles coupled with graphene layers. To confirm the model, we compared the resulting spectrum obtained through numerical simulations of LSPR to those released without a layer of graphene. From a practical standpoint, it opens the door to nanotechnology in a controlled and predictable manner. The spectral properties of systems based on Ag-graphene nanoparticles to enhance their particular applicability for sensing applications. This study opens the way to highly sensitive sensors and improved biosensors.

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