Reflectivity Optimization of the SPR Graphene Sensor

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Received: 10 October 2017; accepted: 14 February 2018

Abstract. In this study, the optimization of the surface plasmon resonance (SPR) sensor based on graphene–silver substrate was investigated. We simulated the reflection spectrum that depends on the metal thickness and the number of graphene layers. The addition of a certain number of graphene layers based on the knowledge that silver oxidation decreases the sensitivity of the sensor improved the sensitivity S_{RI} , while the detection accuracy decreased. To optimize the sensor performance, the investigation focused on monolayer graphene. Furthermore, genetic algorithms were used to optimize the SPR biosensor reflection by adjusting the coefficients of the system, which are the incidence angle and metal layer thickness.

Keywords: surface plasmon resonance (SPR), biosensor, graphene, genetic algorithm

1. Introduction

Many recent works have undertaken the manufacture and development of biosensors using various sensing methods. Surface plasmon resonance (SPR) is one among them. This method investigates a wide range of different putative interactions with a variety of biomolecules in real time without labelling [1, 2].

Surface plasmon is a charge-density oscillation propagating along the interface between metal and dielectric medium. The evanescent wave of the surface plasmon is affected and determined by the properties of the metal and dielectric, and it can be excited by a light. The Kretschmann configuration is one of the techniques that are used in this excitation [3]. It is based on the observation of the reflected light spectrum obtained either by “angular interrogation” or “wavelength interrogation”. Generally, in SPR sensors the angular approach is the preferred due to its low angular resolution [4–6]. It is based on the measurement of changes in the resonant angle extracted from the curve when changing the angle of incidence and keeping the wavelength constant. In addition to the resonance angle, this spectrum is characterized by two other factors are half width at half maximum (FWHM) and reflection minimum $R_{\min}$ [5].

The noble metals [7, 8] as Ag and Au are more commonly used to support the propagation of surface plasmon polariton waves (SPP) at visible light wavelengths [4, 9, 10]. How-

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ever, silver substrates seems to be the most appealing, because it provides in the metal-sensing layer interface a high value of electric field enhancement with low imaginary part of refractive index [11]. When compared with gold, the plasmon coupling in the silver has a sharper angular resonance peak [12], but it presents a drawback that it can be easily oxidized and the poor chemical stability thus affecting the performance of SPR-sensors [11, 12]. Many of methods were designed in an attempt to overcome this problem, for example the use of bimetallic silver–gold layers [13, 14], a thin oxide films TiO_2 or SnO_2 [15–17], In addition to thin films of carbon such as the amorphous carbon [18], amorphous carbonated silicon ($\text{a-Si}_{0.63}\text{C}_{0.37}$) [19–21] and the newer one is, graphene [13].

Graphene is a carbon nanomaterial with two-dimensional (2D) sheet structure composed of sp^2 carbon atoms arranged in a honeycomb lattice [21]. It has been proposed as an addition of layers that can prevent the oxidation of silver metal and at the same time have a high capacity of efficient adsorption of biomolecules [11, 22], due to their high surface area and rich π conjugation structure. This π orbitals form a dense cloud that blocks the gap within its atomic rings. Thus preventing the oxygen from interacting with them [13, 21–23]. Further, the graphene layers possesses very fascinating properties include the optical conductivity in the visible-infrared range, the lowest resistivity, high electron mobility, and relatively low cost [1, 24, 25].

Based on these properties, Wu et al. [26] have investigated graphene-on-gold SPR biosensor and demonstrated that the biosensor with L graphene layers is more sensitive than the conventional gold thin film SPR biosensor. The improved sensitivity is due to the increased adsorption of biomolecules on graphene. Maharana et al. [27] incorporated graphene as dielectric over layer on SPR active metal Ag for enhancing electric field at the sensing layer interface. The proposed sensor shows enhanced performance as compared to widely reported Au on Ag configuration. Choi et al. [28] have demonstrated that sensitivity of graphene-on-silver SPR biosensor can be greater than the sensitivity of the conventional gold-film-based biosensor. as well as the graphene layers prevent silver oxidation. Islam et al. [4] have studied the enhancement of the sensitivity and adsorption efficiency of a localized surface plasmon resonance (LSPR) biosensor that includes a layer of graphene sheet on top of the gold layer. The LSPR graphene biosensor has better sensitivity with lower operating wavelength and larger number of graphene layers have also been studied. In other works [29], Djeflal et al. proposed a new graphene-based sensor called Dielectric Modulated Graphene Field Effect Transistor (DMG-FET), for high-performance biomolecule sensing applications.

In this paper, we shall present an ultra-stable high performance SPR sensor based on a graphene on Ag configuration, which depends on the thickness of the silver layer and the number of graphene layers.

In the light of new optimization techniques, we used a genetic algorithm (GA), which is a heuristic method. It was found to be a powerful tool to solve and optimize problems based on the survival of the fittest. The theory of the evolution of species in their natural environment is an artificial transposition of basic concepts of genetics and survival of laws set forth by Charles Darwin: the most adapted individuals survive and reproduce. These same mechanisms will be used in the implementation of the GA [30, 31], which will be validated by a problem which searches for the optimum reflectivity of graphene SPR biosensor. The optimization procedure starts with the acquisition of different data related to the parameters in the problem, which are mainly the interval of incidence angle and the metal thickness layer. A good agreement is observed between the theoretical generalized multi-layer model and the optimization result. The obtained results allow us to conclude that the reflection fitness function is minimal whatever the number of iterations.

The SPR sensor with the Kretschmann configuration consists of a glass prism, a silver film which can experimentally deposited onto the prism by hot evaporation with depositing rate of $0.5 \text{ \AA/s}$ at a vacuum of $1 \cdot 10^{-6} \text{ Torr}$ [32], and a sample in sequence [33]. Kretschmann configuration is a simple model for excitation SPR, that understood with observed the SPR curve (the change reflected intensity R versus function angle incidence). When the light wave traverses the prism it generates an evanescent wave that propagates and penetrates in the metal surface with propagation constant k_x as in Eq. (1).

$$k_x = (2\pi/\lambda_0)n_1 \sin\theta \quad (1)$$

where θ is the angle of incidence of light with the normal to the interface, and n_1 is the prism refractive index.

For a specific angle of incidence, k_x can be adjusted to match the SPP and its interaction with the light wave influences the reflectivity R [26]. In the simulation of the SPR biosensor with a graphene-on-silver substrate (shown in Fig. 1a), we used the same previous model to which a new element was added: the graphene layer is sandwiched between the silver and a sensing film layer, to absorb the biomolecules (e.g. ssDNA) present in the water. Practically, The transfer of graphene onto silver SPR interfaces can be achieved in several ways as dry transfer technique. The transfer of large-area graphene grown by chemical vapour deposition (CVD) onto metal substrates is an attractive technique that enables us the control of transferred graphene layers number [13].

The composition of the sensor is as follows:

- A tunable laser (632.8 nm),
- A prism made of silica glass BK7 ($n_1 = 1.515$, depth $d = \infty$),
- A silver metal layer whose dielectric function follows the Lorentz–Drude model.

The relationship is given by

$$\epsilon_m(\omega) = 1 - \left(\frac{f_0 \omega_p^2}{\omega(\omega + i\Gamma_0)} \right) + \sum_{j=1}^k \frac{f_j \omega_p^2}{\omega_j^2 - \omega^2 + i\omega_j} \quad (2)$$

where $\omega = (2\pi c/\lambda)$ is the light frequency, ω_p the plasma frequency, k the number of oscillators with frequency ω_j , strength f_j , and lifetime $(\Gamma_j)^{-1}$; while $f_0 \omega_p^2$ is the plasma frequency associated with intra-band transitions with oscillator strength f_0 and damping constant Γ_0 [33].

- A graphene layer of refractive index $n_{gr} = 2.7 + i1.4$ [27] and thickness $d = L \times 0.34 \text{ nm}$ [28], where L is the number of graphene layers.
- A bio-sensing layer $n_s = 1.33$.

Fig. 1. (a) Configuration of the graphene-on-silver SPR biosensor [1]. (b) Schematic diagram of the multi-layer model [33]

We will use a generalized N -layer model [1, 33, 34], shown in *Fig. 1b*. To generate a SPR curve and obtain the reflectivity of the incident p-polarized light [27], all the layers are stacked along the z direction. The arbitrary medium layer is defined by the thickness (d_k), dielectric constant (ϵ_k), permeability (μ_k), and refractive index (n_k). The tangential fields at the first $z = z_1 = 0$ and at the last boundary $z = z_{N-1}$ [1] are related by

$$\begin{bmatrix} U_1 \\ V_1 \end{bmatrix} = M \begin{bmatrix} U_{N-1} \\ V_{N-1} \end{bmatrix}, \quad (3)$$

U_1, U_{N-1} are the components of electric field at the boundary of the first and N th layers of structure, respectively, while V_1, V_{N-1} are the components of magnetic field and M is the characteristic matrix of this structure, which is given by

$$M = \prod_{K=2}^{N-1} M_K \quad (4)$$

with

$$M_k = \begin{bmatrix} \cos \beta_k & -i \sin \beta_k / q_k \\ -i q_k \sin \beta_k & \cos \beta_k \end{bmatrix}, \quad (5)$$

where

$$q_k = [\epsilon_k + n_1^2 \sin^2 \theta]^{1/2} \epsilon_k^{-1} \quad (6)$$

and

$$\beta_k = (2\pi d_k / \lambda) [\epsilon_k + n_1^2 \sin^2 \theta]^{1/2}.$$

The amplitude reflection coefficient (r_p) for p-polarized light is:

$$r_p = \frac{(M_{11} + M_{12}q_N)q_1 - (M_{21} + M_{22}q_N)q_N}{(M_{11} + M_{12}q_N)q_1 + (M_{21} + M_{22}q_N)q_N}. \quad (7)$$

The reflection coefficient of the N -layer model for p-polarized light is:

$$R_p = |r_p|^2. \quad (8)$$

For an enhanced performance of biosensors, the sensitivity is an important issue for its application. It depends on a number of factors such as the operating wavelength and material characteristics, including the refractive index of dielectric layer, refractive index of prism, metal film, and film thickness, etc. They are selected to optimize the resonance condition [2]. The sensitivity of the sensor is defined as [1]:

$$S = (\Delta\theta_{\text{res}} / \Delta n) \quad (9)$$

where the change in the resonance angle is $\Delta\theta_{\text{res}}$, corresponding to a change Δn in the refractive index of sensing layer. The resonance angle (θ_{res}) corresponding to the minimum reflection coefficient (R_{min}) is determined from the $R - \theta$ curve.

Another important requirement for a high-performance SPR sensor is its high detection accuracy (which is inversely proportional to the FWHM). It can be noted that the precision of the measured SPR angle depends strongly on the width of the SPR curve [11, 35].

2. Design parameters and results

2.1. Effect of film thickness on the SPR-sensor

Without graphene layers: The metal thickness is a very important parameter in surface plasmon resonance. It defines the maximum dynamic and shape of the plasmon [7, 33].

Figure 2 shows the calculated results as a series of SPR curves for silver thickness increasing in steps of 10 nm changing from 10 nm to 100 nm. The resonance angle (θ_{res}) increases in the range of 53–67.8°.

The thickness of the silver layer is optimized to such an extent that the resonance dip in the SPR curve becomes the sharpest with a reflection minimum close to zero. So the best value is 51.26 nm, which can be precisely extracted from the curve of reflectivity as a function of the silver layer thickness (inset of Fig. 2).

Fig. 2. Silver thickness layer effect on the reflection spectrum

It is not necessary a very high thickness, because as shown in Fig. 2, the maximum is around 100 nm. Above this thickness, the evanescent wave due to the total reflection on the prism cannot cross the metal to generate plasmon at the metal/dielectric interface. Thus, the minimum of reflectance moves away from zero and the SPR reflection spectra start to broaden. In the case of a thin metal (10–30 nm), it tends to limit the total reflection on the prism because there is not enough metal to absorb the incident wave and oscillate plasmons. For these small thicknesses, it is seen that the reflectivity on one hand does not drop to a minimum of zero. On the other hand, the resonance is large, which reduces the sensor sensitivity. Furthermore, it was found that the variation in metal thickness decreases the amplitude of the resonance peak, which means a reduction in the transfer of incident energy to surface plasmons. The optimization of the thickness of the metal is important to work in the best coupling conditions and have the best sensitivity of the SPR sensor [7, 33].

With graphene layers: In order to study the effect of the film thickness on the SPR graphene sensor, we should take into account the number of graphene layers. Figure 3a illustrates that when the number of graphene layers L increases, it leads to a shift of the resonance peak, a

Fig. 3. (a) SPR reflectance and (b) change in the SPR angle as the number of graphene layers increases (0–5 layers), with sensing layer refractive index of 1.330

variation of its amplitude, and a change and modification of the width of its peaks which become wider respectively. For five graphene layers, the plasmonic angles shift from 67.81° to 69.02° . Therefore, it was shown in *Fig. 3b* that the change of the position of the minimum surface plasmon varies linearly with the number of additional graphene layers.

Additionally, one can note that the reduction in the amplitude of the resonance dip is a direct consequence of the change in the propagation constant of surface plasmons as a function of the number of graphene layers. The latter becomes different of the wave vector tangential component of incident light and hence the resonance condition is not fulfilled (i.e., SPs become damped) [10, 13]. To minimize this effect, we optimize the silver layer thickness that decreases when the number of graphene layers increases (see *Table 1*).

The silver thickness for which the reflectivity is zero represents the optimum thickness. *Figure 4a* illustrates the variation of reflectivity as a function of silver layer thickness for various numbers of graphene layers.

Table 1. Optimized values of silver thickness in terms of the number of graphene layers for corresponding changes in the resonance angle for zero reflectivity in the SPR. The refractive index of the sensing film was 1.33

L graphene layers number	D_{Th} silver thickness (10^{-9} nm) theory	θ_{Th} theoretical resonance angle (degree)	$R_{min(Th)}$ minimum reflection coefficient (a.u.)
0	51.26	67.817	6.4485 E–004
1	47.22	68.058	6.3873 E–004
2	44.23	68.298	4.2867 E–004
3	41.84	68.538	2.0990 E–004
4	39.85	68.778	5.8913 E–005
5	38.14	69.099	5.5688 E–007

Fig. 4. (a) Variation of reflectivity with respect to the thickness of the silver layer. (b) Angular reflectivity spectra corresponding to optimum values of the thickness of silver for graphene layers (1–5 layers)

Figure 4b shows the angular spectra of reflectivity corresponding to these optimum values. It is noted that the optimization of the silver thickness provides a shallower dip in the reflectivity curve. The results are summarized in Table 1.

2.2. Graphene layers effect on detection accuracy and SPR-sensor sensitivity enhancement

It is important to make a good choice for the maximum number of graphene layers, which must be fixed for the high performance of the biosensor. This fixed number must be taken in order to give a high sensitivity enhancement and a high detection accuracy.

In Fig. 5a, the sensitivity enhancement as a function of the number of graphene layers L after the absorption of biomolecules is plotted, assuming the same refractive index change $\Delta n = 0.005$. It is found that adding more graphene layers allows an increase in sensitivity. The

Fig. 5. (a) Variation in sensibility enhancement and (b) detection accuracy versus the number of graphene layers (1–5 layers)

Fig. 6. SPR reflection spectra for monolayer graphene layers as a function of the incident angle with various values for sensing medium refractive index

Fig. 7. Variation of detection accuracy with sensing layer refractive index for monolayer graphene at two wavelengths (632.8 nm in our study and 633 nm in Ref. [27]) for comparison

sensitivity enhancement value can be 1.4% for the monolayer graphene biosensor, 3.1% ($L = 2$), 4.5% ($L = 3$), 6.1% ($L = 4$), and 9.3% ($L = 5$). Graphene is a good absorbent of biomolecules, since the increase of graphene layers means the increase in the absorption of biomolecules, thus an increase in sensitivity [10]. *Figure 5b* shows the effect of the number of graphene layers on detection accuracy. We can see that the increase in the number (or thickness) of graphene layers causes a reduction in detection accuracy from 0.818 to 0.221 degree⁻¹. The reason of this decrease is the breadth in the curve, as a result of the damped extension of SP field into the graphene layer, due to the amortization in the absorption of surface plasmon at the silver/graphene interface, which is also due to the optical properties of graphene (non-zero imaginary refractive index part) [10, 13, 26]. Finally, it is clear that the use of graphene as a protective layer in the Kretschmann configuration based on silver becomes important if the number of layers decreases. The following work will address the case of a monolayer graphene layers on a thin silver film.

For the monolayer graphene SPR sensor, we have theoretically demonstrated that the refractive index of the sensing film has an important influence on the reflection spectra and detection accuracy, because any change in the properties of the dielectric medium are determined by the surface plasmon evanescent waves [5]. *Figure 6* shows that when the sample refractive index n_s changes by 0.04 RIU (from 1.33 to 1.37), the width of resonance values increase from 1.7881 to 2.3642. This increase in the width of the curve leads to difficulties in a θ_{SPR} measurement [10, 26], and therefore to a decrease in the accuracy detection of the refractive index measurement, as depicted in *Fig. 7*. The results are compared with those obtained by Maharana et al. (2013) [27] for an operating wavelength of 633 nm. As it can be seen, they are in good agreement with the same change of sensing layer refractive index.

3. Genetic optimization

A developed analytical expressions can be exploited to formulate the objective functions to optimize hence the device performance using the most known metaheuristic techniques known as genetic algorithm approach [36]. The GAs are searching processes based on the laws of natural selection and genetics, from Darwinian's theory of survival of the fittest. They use probabilistic transition rules instead of deterministic rules. The GAs are iterative stochastic algorithms that work upon groups of codified points over an initial population. The population of potential solutions is represented by individuals or chromosomes (parents) that

evolve through successive iterations called generations. The fittest individual will survive a generation and generate offspring, which is evaluated by the quality of its fitness function and with a similar fashion to their parents. This is the last individual, which might be stronger. In order to allow a population of solutions to converge to optimal solutions, we follow iteration process steps based on the genetic triangle: reproduction, evaluation, and selection [37–40]. In this work, we will be searching for the best value of resonance angle and thickness film, which will give the optimum reflection fitness function. The various implementation shell steps of this methodology are summarized below.

Initialization: The first phase of the AG consists to choose randomly the initial population that contains a group of individuals whom are the different operators of an AG that would be applied.

Selection: Selection is supposed to be able to compare each chromosome in the population. Good chromosomes and forms a mating pool it is likely to be selected to reproduce, the most useful method is the roulette wheel scheme (RWS).

Crossover: Crossover operator is applied to the mating pool with the hope that it creates a better offspring and that is through exchanging information among strings of the mating pool with a crossover probability P_c . A string point crossover operator is used here which is performed randomly by choosing a crossing site along the string and by exchanging all bits on either side of the chosen site.

Mutation: This operator randomly flips some of the bits in a chromosome, i.e. 1 is mutated into 0 and vice versa. Mutation is also used to maintain diversity in the population.

Fitness evaluation: The evaluation phase permits to determine whether the optimum is obtained or re-executed another cycle of the AG. Each individual is codified then evaluated using the objective functions [30, 37–40].

In this work, we focus on optimizing the physical and geometrical design parameters of our proposed device (i.e. the value of resonance angle and thickness film) with the aim of reaching their greatest performance (i.e. the value of the reflection fitness function). The GA-based approach can be appropriate for investigating and optimizing the proposed SPR graphene sensor design through optimized the minimizing the reflection R . Typically, based on matrix for an N -layer system, the objective function can be expressed by the following equation:

$$\text{Fitness}(X) = R,$$

where X denotes the design physical and geometrical vector presented by: $X = (\theta, D)$. Evidently, a global optimization problem based on mono-objective procedure is mainly illustrated by a candidate solution in well defined search space and a set of constraints that should be respected [36]. These constraints can be given in our case by the following conditions:

- $x \in [x_{i,\min}, x_{i,\max}]$, $x_i \in X$ (each design variable should be confined within a given range). In this framework, the intervals of incidence angle θ_{inc} is $[10 \dots 90]$ (degree) and thickness metal layer D is $[30 \dots 100]$ (nm), respectively,
- L is the number of graphene layers and is equal to one,

Fig. 8. Best fitness plot for the minimization of the reflection function. The plots show the results from *Table 2*

Table 2. Summary of results of the GA application

Population size	20
Number of generations	51
Method used in selection	Roulette wheel scheme (RWS)
Crossover probability P_c	0.8
Mutation probability p_m	0.01
Optimum minimum reflection coefficient $R_{\min}$ (a.u.)	4.3591 E-10
Resonance angle optimize $d\theta_{\text{Ag}}$ (SPR) (degree)	67.927
Silver thickness optimized D_{Ag} ($\cdot 10^{-9}$ nm)	48.404
Execution time (s)	≈ 0.732634

for which a quick stabilization of the objective function can be achieved where the estimated error is extremely small. After performing the optimization procedure, we can notice the outstanding capability of the proposed GA-based optimization for reaching the global optimum which is depicted in *Fig. 8*. In this context, this figure shows the evolution of the fitness function, $Fitness(X)$, against the generation number, where it is obvious that a quick stabilization of the objective function nearly after 51 iterations is achieved.

The convergence of the GA for optimum reflectivity is represented by the best parameters found in *Table 2*. The choice of a population with a small size leads to a fast resolution, even with a large number of evaluations of the fitness function, and it is effective in terms of time. Therefore, no change in the metal thickness or resonance angle after these values of size was noticeable. The obtained results considering these parameters are summarized in *Table 2*.

4. Conclusion

We have presented in this paper, using numerical simulations, the influence of two parameters on the material characteristics of the SPR-sensor based on graphene and silver substrates. These parameters are the thickness of silver layer and the number of graphene layers on the shape of reflectivity curve. For the conventional SPR biosensor, it is found that a deviation of the metal thickness compared with the optimum value $D_{\text{opt}} = 51.26$ nm causes a shift in the resonance peak, a change of its width, and a variation of its amplitude. After the addition of graphene layers, which are used as a layer to protect the silver metal from oxidation, the reflectivity does not drop to a minimum of zero because the surface plasmons become damped. This leads to the optimization of the silver layer thickness, which decreases when the number of graphene layers increases to enable the realization of the resonant condition and thereby increase the efficiency of surface plasmons excitation. On the other hand, we have presented a discussion concerning the enhanced performance of SPR graphene sensor, which is assessed in terms of sensitivity and detection accuracy. We have found that the sensitivity enhancement value can be increased from 1.4% for the monolayer graphene to 9.3% for five layers, but the detection accuracy decreased from 0.78 to 0.46 degree⁻¹. Afterwards, we focused on the case of a monolayer graphene on a thin silver film used in the implementation of the GA. Its application has allowed us to obtain the searched results related to the problem of determination of the optimum reflected by optimal reflection, as well as seeking the best parameters allowing us to determine them according to the incidence angle and metal thickness.

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