Modeling of a Graphene Nanoribbon–based Microfluidic Surface Plasmon Resonance Biosensor

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Abstract
A surface plasmon resonance (SPR) biosensor based on a graphene nanoribbon array in a microfluidic flow cell operating in a flow-over format is studied. The optical response of the biosensor is numerically obtained by using rigorous couple wave analysis (RCWA). The performance of the biosensor is described in terms of the limit of detection, which is calculated as a function of key nanoribbon dimensional parameters, such as strip thickness and width, and fill fraction (nanoribbon width to array period ratio). The analysis shows that there are specific values of the fill fraction that optimize, that is, minimize, the limit of detection for particular nanoribbon dimensions. Fabrication issues are also discussed. This study is expected to assist in the design and implementation of SPR biosensors based on nanopatterned 2D materials.

Keywords Surface plasmon resonance · Affinity biosensor · 2D material · Graphene · Numerical modeling

Introduction
Surface plasmon resonance (SPR) biosensors make use of surface plasmon polariton (SPP) waves to probe interactions between biomolecules and the sensor sensitive surface. A SPP is a perpendicularly confined evanescent electromagnetic wave that propagates at the interface between a metal and a dielectric (i.e., sensing medium) [1–3]. Changes of biomolecule concentration at the metal/dielectric interface produce local changes in the refractive index (RI) in that region. This modifies the propagation constant of the SPP, which can be optically measured by the attenuated total reflection (ATR) method [4].

2D materials, such as graphene, MoS₂, WS₂, MoSe₂, WSe₂, and black phosphorous, have been lately incorporated into SPR sensor configurations in order to enrich the capabilities of these optical sensors [5–9]. These nanomaterials provide new approaches to improve the RI sensitivity of SPR sensors due to their remarkable optical properties. In addition, 2D nanomaterials can provide an extremely high density of active surface sites over a large area, making 2D materials-based SPR devices highly suitable for biochemical sensing. Graphene is particularly interesting as it can also act as a biomolecular recognition element (BRE) in order to enhance the adsorption of biomolecules with carbon-based ring structures [10–12].

Recently, SPR-based RI optical sensors using nanoribbons (thin strips with width less than 100 nm) of graphene and WSe₂ have been proposed and theoretically analyzed [13]. Through numerical simulations employing rigorous couple wave analysis (RCWA), the performance of these nanoribbon-based sensors was compared with that of similar structures containing continuous 2D material films. These authors concluded that the nanoribbon-based sensors should exhibit better sensing performance than that of their counterparts based on continuous 2D films. However, in that work, specific information concerning the width of the nanoribbons and the period of the strip array (assuming a periodic nanoribbon arrangement) was not indicated, and, consequently, no analysis of the effect of these key dimensional nanoribbon parameters on the sensor performance was presented. Additionally, the calculations in [13] were based on a non-realistic value for the refractive index of one of the considered plasmonic materials (aluminum), which diminishes their practical applications.

This paper targets at providing a deeper insight into the performance of a SPR sensor based on a nanoribbon array made of a 2D material–graphene–by using numerical analysis.
modeling. The studied device is a graphene-on-gold configuration operating as an affinity biosensor inside a microfluidic flow cell. The effects of graphene nanoribbon width and thickness and fill fraction (nanoribbon width to array period ratio) on the sensor performance are investigated. Among the multiple characteristics that can be used to describe the performance of a biosensor, probably the most analytically relevant is the smallest concentration of target molecules (analyte) that can be detected. Thus, the limit of detection \( (c_{LOD}) \) is used to characterize the biosensor performance. The presented analysis of \( c_{LOD} \) takes into account not only the influence of the sensor sensitivity and noise, but also the effect of analyte transport to the sensitive regions of the device.

**Device Structure and Modeling**

Figure 1 shows a schematic diagram of the studied device. It consists of a periodic planar array of graphene thin strips (nanoribbons) on an Au layer deposited on a SF10 glass substrate. The width and thickness of the graphene strips are \( w_s \) and \( t_s = L \times t_g \), respectively, where \( t_g \) is the thickness of a monolayer of graphene and \( L \) is the number of monolayers. The period of the array is denoted as \( p \). The thickness of the Au layer is \( t_{Au} = 50 \) nm. A biolayer, made of captured analyte molecules, of thickness \( t_{bio} = 5 \) nm is assumed to be adhered on the sensitive (active) surface of the device, that is, on the top surface of the graphene strips. The refractive indexes of all materials used in the simulations at a wavelength of 633 nm (operation wavelength) are collected in Table 1.

The graphene nanoribbon array is situated on the floor of a microchannel (flow cell) of dimensions \( H_c \) (height) \( \times W_c \) (width) \( \times L_c \) (length). An aqueous solution of the analyte is assumed to flow through the microchannel at a flow rate \( Q \). The analyte molecule diffusivity is considered to be \( D = 6.99 \times 10^{-11} \text{ m}^2/\text{s} \), corresponding to a single stranded DNA molecule [14]. The graphene strips are placed orthogonal to the flow direction, allowing for a 2D simulation domain [15]. The device is optically interrogated by striking a p-polarized 633-nm-wavelength light beam onto the graphene nanoribbon array through the glass substrate at an angle of incidence \( \theta \), as shown in Fig. 1b. The optical reflectance versus the angle of incidence was calculated by using 2D RCWA method (from Rsoft Components Design Suite). The analytical signal of

![Fig. 1](https://example.com/fig1.png)

**Table 1** Main parameters used in the simulations of the graphene nanoribbon–based SPR affinity biosensor

| Parameter                        | Symbol     | Value                  | Ref |
|----------------------------------|------------|------------------------|-----|
| Graphene refractive index        | \( n_{grp} \) | 3.31.149               | 5   |
| Au refractive index              | \( n_{Au} \) | 0.138 +j3.62           | 7   |
| Biolayer refractive index        | \( n_{bio} \) | 1.43                   | 14  |
| SF10 glass refractive index      | \( n_{glass} \) | 1.723                  | 5   |
| Aqueous solution refractive index| \( n_b \)  | 1.33                   | 5   |
| Graphene monolayer thickness     | \( t_g \)  | 0.34 nm                | 5   |
| Au layer thickness               | \( t_{Au} \) | 50 nm                  | 5   |
| Biolayer thickness               | \( t_{bio} \) | 5 nm                   | 14  |
| Analyte diffusion coefficient    | \( D \)    | \( 6.99 \times 10^{-11} \text{ mm}^2/\text{s} \) | 14  |
| Biolayer surface density         | \( \Gamma_0 \) | \( 3.9 \times 10^{-14} \text{ mol/mm}^2 \) | 14  |
| Association rate constant        | \( k_1 \) | 1.8 \times 10^7 (1/M s) | 14  |
| Microchannel height              | \( H_c \)  | 50 \mu m               | 14  |
| Microchannel width               | \( W_c \)  | 3 mm                   | 14  |
| Microchannel length              | \( L_c \)  | 2 mm                   | 14  |
| Flow rate                        | \( Q \)    | 20 \mu L/min           | 14  |
| Intensity noise at the threshold  | \( \sigma_{th} \) | 0.6%                  | 16  |
| Pixels included in the data processing | \( N_s \) | 400                    | 16  |
| Frames averaged for each time record | \( N_t \) | 100                    | 16  |
| Detection time                   | \( T \)    | 600 s                  | 14  |

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the sensor is the value of the angle of incidence at resonance, $\theta_r$. The limit of detection, $c_{LOD}$, is defined as

$$c_{LOD} = \frac{3\sigma}{S_c(c = c_{LOD})}$$  \hspace{1cm} (1)

where $\sigma$ is the noise in the sensor output and $S_c = \frac{d\theta_r}{dc}$ is the sensitivity of the sensor response ($\theta_r$) to the concentration of the analyte ($c$). For affinity biosensors, in which target molecules bind to receptors immobilized on the active surface, $S_c$ can be decomposed into $S_c = S_H(d\Gamma/dc)$, where $S_H = \frac{d\theta_r}{d\Gamma}$ is the sensitivity of the sensor response to changes in the surface density of the captured analyte ($\Gamma$), and $d\Gamma/dc$ indicates the efficiency of analyte transport from the bulk solution to the sensor surface. Thus, $c_{LOD}$ can be written as

$$c_{LOD} = \frac{3\sigma}{S_H \cdot \kappa}$$  \hspace{1cm} (2)

where $\kappa = (d\Gamma/dc)_{c=LOD}$ is the mass transport efficiency. For low analyte concentrations, $\kappa$ can be expressed as [14]:

$$\kappa = \left( \frac{1}{k_1 \Gamma_0 T} + \frac{1}{k_m T} \right)^{-1}$$  \hspace{1cm} (3)

where $k_1$ is the association rate constant, $\Gamma_0$ is the surface density of the biolayer, $T$ is the total detection time, and $k_m$ is the diffusion-limited mass transport coefficient, which is given by [15]:

$$k_m = \frac{k^e_m - k^l_m f}{\frac{\sqrt{2} f - 2f + 1}{k^l_m}}$$  \hspace{1cm} (4)

where $f = w/s$ is the fill fraction, $k^l_m$ and $k^e_m$ denote the diffusion-limited mass transport coefficients corresponding to a continuous layer of graphene and a single graphene strip, respectively, and were calculated as [15]:

$$k^l_m = \frac{D}{H \eta_t} \left( 0.8075(6\eta_t^2 Pe)^{1/3} + 0.7058(6\eta_t^2 Pe)^{-1/6} - 0.1984(6\eta_t^2 Pe)^{-1/3} \right)$$  \hspace{1cm} (5)

$$k^e_m = \frac{Dg}{\pi H \eta_t} \left( 4 - 0.12(6\eta_t^2 Pe)^{3/2} \right)$$

$$\pi H \eta_t \left( 1 - 0.2(6\eta_t^2 Pe)^{1/2} \right)$$  \hspace{1cm} (6)

where $\eta_t = L_t / H_t$, $\eta_s = w_s / H_t$, $g$ is a shape factor ($= 1$ for a strip) [14], and $Pe = Q/W_D$ represents the channel Peclét number.

Surface sensitivity was calculated as $S_f = \Delta \theta_r / \Gamma_0$, where $\Delta \theta_r$ is the difference of the resonance angle with biolayer and the resonance angle without biolayer.

Assuming that the optical response of the biosensor is recorded by using a scientific grade CCD detector, with $N_s = 100$ frames averaged for each time record (which corresponds to a measurement period of approximately 1 s) and $N_s = 400$ pixels included in the data processing, the noise ($\sigma$) can be estimated by means of the following expression [16]:

$$\sigma = K \left( \frac{\sigma_{th}}{\sqrt{N_s}} \right) \left( \frac{w}{d} \right)$$  \hspace{1cm} (7)

where $\sigma_{th} = 0.6\%$ is the total intensity noise at the threshold [16], $w$ and $d$ are the width and contrast of the spectral dip, and $K$ is a factor depending on the relative contributions of the sources of noise. Uncorrelated intensity noise is assumed, which corresponds to $K = 1$ [16]. The term $N_s = N_s N_t$ is the total number of intensity measurements, which includes both spatial $N_s$ and temporal $N_t$, averaging.

Values of the main dimensional and physical parameters considered for the modeling of the graphene nanoribbon-based SPR biosensor in a microfluidic channel are collected in Table 1.

**Results and Discussion**

Figure 2 shows representative spectral reflectances of the studied configuration, calculated for $L = 2$, $w_s = 20$ nm, and $f = 0.7$. Black and red lines correspond to the cases
without (w/o) and with, respectively, an adhered biolayer. The resonance angle, $\theta_r$, is the incident angle at which the reflectance is minimum. It is seen that the presence of the biolayer leads to a shift of $\theta_r$. Such a shift ($\Delta \theta_r$) is used to calculate the surface sensitivity of the sensor, $S_T$, as indicated in “Device Structure and Modeling”. The width (full width at half minimum), $w$, and depth (contrast), $d$, of the SPR dip, used to estimate the noise (Eq. 7), are also indicated.

Figure 3 shows the calculated $c_{LOD}$ as a function of the fill fraction ($f$) for $w_s = 10, 20$ and 30 nm for $L = 1$ (Fig. 3a), $L = 2$ (Fig. 3b), $L = 3$ (Fig. 3c) and $L = 4$ (Fig. 3d). Figure 4 plots $c_{LOD}$ versus $f$ for $L = 1, 2, 3, $ and $4$ for $w_s = 10$ nm (Fig. 4a), $w_s = 20$ nm (Fig. 4b), and $w_s = 30$ nm (Fig. 4c). It is seen in all graphs that $c_{LOD}$ exhibits a minimum for particular values of $f$. Specifically, the lowest $c_{LOD}$, and the corresponding values of $w_s$ and $L$ are collected in Table 2.

In order to gain insight into the reasons causing the appearance of $c_{LOD}$ minima, the parameters that determine $c_{LOD}$ (Eq. 2): surface sensitivity ($S_T$), noise ($\sigma$), and mass transport efficiency ($\kappa$) are plotted as a function of $f$ in Fig. 5 for all considered values of $L$ and $w_s$. It is observed in Fig. 5a that $S_T$ varies approximately proportionally to $f$, which is expected as $f$ represents the device surface fraction covered by the biolayer. On the other hand, Fig. 5c shows that $\kappa$ scales as $\kappa \propto f^{-1}$ (indicative of diffusion-limited conditions [14]) at high fill fraction, and asymptotically approaches a constant value (indicative of reaction limited conditions [14]) at low fill fraction (because $k_m \to k_s ^m$ at low $f$ (Eq. 4)). Therefore, the product $(S_T \cdot \kappa)$ is approximately constant at high $f$ and decreases as $f$ decreases at low $f$.

On another hand, according to Eq. (7), $\sigma$ scales as $w/d$. Figure 5b shows the calculated noise as a function of $f$. At high $f$ values, $\sigma$ increases with $f$ due to the optical losses arising from graphene material absorption, which degrade the resonance dip by making it wider (larger $w$) and shallower (smaller $d$). At low $f$ values, as $f$ decreases, $\sigma$ diminishes monotonically towards a minimum value at $f = 0$ (no graphene material absorption), at which $d$ and $w$ should exhibit their maximum and minimum, respectively, values, corresponding to the resonance dip of the bare Au on glass configuration.

Thus, since at low $f$, $\sigma$ is approximately constant whereas $(S_T \cdot \kappa)$ decreases, and at high $f$, $\sigma$ increases whereas $(S_T \cdot \kappa)$ is approximately constant, maximum values of $c_{LOD}$ are
obtained in these extreme \( f \) intervals, leading to a minimum \( c_{LOD} \) range in between, that is, in the middle-\( f \) region, as shown in Figs. 3 and 4.

Table 2 reveals that there is a tendency for the minimum \( c_{LOD} \) to decrease as \( w_s \) decreases (for a given \( L \)) and \( L \) increases (for a given \( w_s \)). Thin (small \( w_s \)) and thick (large \( L \)) nanoribbons are therefore desirable in order to minimize \( c_{LOD} \). In fact, it is under these conditions (small \( w_s \) and large \( L \)) that the advantageous effect of using nanoribbons, instead of a continuous graphene layer, with a proper fill fraction to reduce \( c_{LOD} \) becomes more remarkable. This is evidenced by Figs. 3 and 4, which show that the \( c_{LOD} \) vs. \( f \) curves exhibit higher contrast between the middle-\( f \) region and the extreme-\( f \) regions as \( w_s \) decreases (for a given \( L \)) and \( L \) increases (for a given \( w_s \)). However, Fig. 3 also shows that, in the middle-\( f \) region, the separation along the \( c_{LOD} \)-axis among the curves for the different values of \( w_s \), for a given \( L \), increases as \( L \) increases. This indicates that the larger the value of \( L \), the smaller the dimensional tolerance of \( w_s \), which is an important issue to be taken into account from a manufacturing point of view.

Although the lowest values of \( c_{LOD} \) are obtained in the middle-\( f \) range of Figs. 3 and 4, that is, approximately in the interval \([0.1, 0.3]\), it is also interesting, for the sake of comprehensiveness, to further analyze the dependence of \( c_{LOD} \) with \( w_s \) and \( L \) for \( f \) values outside this interval, i.e.,

**Fig. 4** Limit of detection (\( c_{LOD} \)) as a function of the fill fraction (\( f \)) for \( L = 1, 2, 3, \) and 4 for a \( w_s = 10 \text{ nm} \), \( b \ w_s = 20 \text{ nm} \), and \( c \ w_s = 30 \text{ nm} \)

**Table 2** Minimum values of \( c_{LOD} \) extracted from Figs. 3 and 4

| \( L \) \( w_s \) (nm) \( f \) | Minimum \( c_{LOD} \) (pM) |
|---|---|
| 1 | 10 | 0.3 | 4.63 |
| 1 | 20 | 0.3 | 4.75 |
| 1 | 30 | 0.3 | 4.83 |
| 2 | 10 | 0.2 | 4.54 |
| 2 | 20 | 0.2 | 4.70 |
| 2 | 30 | 0.2 | 4.82 |
| 3 | 10 | 0.1 | 4.45 |
| 3 | 20 | 0.2 | 4.66 |
| 3 | 30 | 0.2 | 4.79 |
| 4 | 10 | 0.1 | 4.34 |
| 4 | 20 | 0.1 | 4.59 |
| 4 | 30 | 0.2 | 4.79 |
for \( f < 0.1 \) (low fill fraction) and \( f > 0.3 \) (high fill fraction). In this respect, Fig. 3 shows that, at high \( f \), \( c_{LOD} \) values for the different values of \( w_s \) and a given \( L \) converge to the value corresponding to a continuous graphene layer (\( f = 1 \)), which is expected; whereas no convergence is observed at low \( f \) as \( f \) decreases. The latter is because, for a given \( L \), at low \( f \), the variation of \( c_{LOD} \) with \( w_s \) is mainly determined by \( \kappa \) (Fig. 5c) as both \( S_{\Gamma} \) and \( \sigma \) do not vary significantly with \( w_s \) (Fig. 5a, b). On another hand, Fig. 4 shows that, at low \( f \), \( c_{LOD} \) values for the different values of \( L \) and a given \( w_s \) tend to converge as \( f \) decreases, whereas these values diverge in the high-\( f \) regime as \( f \) increases. The low-\( f \) behavior is because, for a given \( w_s \), both \( S_{\Gamma} \) and \( \sigma \) do not vary significantly with \( L \) (Fig. 5a, b) and \( \kappa \) does not depend on \( L \). The high-\( f \) behavior occurs because both \( S_{\Gamma} \) (Fig. 5a) and \( \sigma \) (Fig. 5b) increase with \( L \) for a given \( w_s \). Note that, for high \( f \), the shape of the \( c_{LOD} \) vs. \( f \) graphs plotted in Fig. 4 resemble those of \( \sigma \) vs. \( f \) of Fig. 5b. This indicates that, although \( S_{\Gamma} \) counteracts the increasing effect of \( \sigma \) on \( c_{LOD} \) (Eq. 2), noise contribution is dominant and mainly determines the observed \( L \)-variation of \( c_{LOD} \) at high \( f \) values.

![Fig. 5](image)

Fig. 5 a \( S_{\Gamma} \) and b \( \sigma \) as a function of \( f \) for \( L = 1, 2, 3, \) and 4 and \( w_s = 10, 20, \) and 30 nm. c \( \kappa \) as a function of \( f \) for \( w_s = 10, 20, \) and 30 nm

It should be noted that, according to Eq. (2), \( c_{LOD} \) can be further reduced by increasing the mass transport efficiency \( \kappa \). This can be achieved by decreasing the microchannel dimensions, and/or increasing the flow rate and/or the detection time. For the sake of illustration, Fig. 6 shows the calculated \( c_{LOD} \) as a function of \( f (w_s = 10 \text{ nm}, L = 4) \) for the parameters collected in Table 1 (red circular dots), and for other values of \( H_c (= 10 \mu m) \), \( W_c (= 2 \text{ mm}) \), \( Q (= 30 \mu L/min) \), and \( T (= 20 \text{ min}) \) while keeping the values of the remaining parameters unchanged (black square dots). It is seen that the latter set of \( H_c, W_c, Q \) and \( T \) values lead to a \( c_{LOD} \) reduction for all \( f \); in particular, the minimum \( c_{LOD} \) is drastically decreased from 4.34 pM to 0.65 pM. Note that the optimum fill fraction has also changed (from \( f = 0.1 \) to \( f = 0.2 \)), showing that operational parameters such as flow rate and detection time must be also considered when designing this type of biosensors.

Finally, from a fabrication point of view, it should be mentioned that graphene nanoribbons on flat surfaces can be created by, for example, lithography [17–19], and bottom-up approaches [20–22]. Nanolithography techniques such as scanning probe lithography [23] can also be used to pattern...
nanoribbons on WSe$_2$ [24], MoS$_2$ [25], and black phosphorus [26] thin layers. Note also that the optimum $f$ values for the studied configuration (0.1–0.3) allow the use of array periods that are around one order of magnitude larger than the nanoribbon width, which facilitates the array patterning as undesired proximity effects associated to lithography processes are reduced.

**Conclusions**

The performance of a graphene nanoribbon array–based SPR biosensor operating in a microfluidic flow cell has been analyzed through numerical modeling. In particular, the limit of detection ($c_{LOD}$) of the affinity biosensor has been determined as a function of dimensional parameters of the nanoribbons. The analysis indicates that using nanoribbon arrays with proper fill fractions allow obtaining lower $c_{LOD}$ values than using continuous graphene layers. The advantageous effect of nanoribbons over the continuous layer becomes more evident as the nanoribbon width and thickness decreases and increases, respectively. Thin and thick nanoribbons are desirable to minimize $c_{LOD}$, although this implies more stringent fabrication tolerances. It is also shown that the limit of detection can be further reduced by improving the efficiency of analyte transfer to the sensitive regions, which can be achieved by reducing the microchannel dimensions and/or increasing the detection time and/or the flow rate. This study is also applicable to SPR biosensors based on other 2D materials, and can therefore assist in the design and optimization of these promising type of optical biosensors.

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**Availability of Data and Materials** The datasets generated during and/or analyzed during the current study are available from the corresponding author on reasonable request.

**Declarations**

**Ethics Approval** Not applicable.

**Consent to Participate** Not applicable.

**Consent to Publication** Not applicable.

**Conflicts of Interest** The authors have no conflicts of interest to declare that are relevant to the content of this article.

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