Highly Sensitive Graphene-Based Refractive Index Biosensor Using Gold Metasurface Array

Shobhit K. Patel, Juveriya Parmar, Himitri Trivedi, Rozalina Zakaria, Truong Khang Nguyen, and Vigneswaran Dhasarathan

Abstract—In this letter, a highly sensitive refractive index biosensor applicable in medical devices is proposed. The biosensor design is analyzed for different concentrations of hemoglobin biomolecules. The 70nm shift in the wavelength is observed for two hemoglobin biomolecules with 1.34 and 1.36 refractive index per unit (RIU). The highest sensitivity of 3500 nm/RIU is observed for these hemoglobin biomolecules. The results are analyzed for absorption, electric field and magnetic field. The design is also analyzed for different geometrical parameters like metasurface array thickness and silicon dioxide (SiO2) layer thickness. The increase in metasurface array thickness reduces the absorption of the design. The increase in the thickness of SiO2 layer increases the absorption till 0.6 µm thickness than the response remains the same for other thicknesses. The proposed highly sensitive graphene-based metasurface array biosensor is applicable in medical and biosensing photovoltaic devices.

Index Terms—Graphene, hemoglobin, metasurface array, sensitivity.

I. INTRODUCTION

Optical biosensors are very effective amongst all the biosensors because they detect biomolecules directly and in real-time [1], [2]. Optical biosensors are also widely applicable in medical applications [2], environmental applications [3], industrial applications [4], etc; because of this fast detection in real-time [2]–[4]. Optical biosensors can be used to detect or sense biomolecules by two main detection methods. The first method to detect is based on fluorescence and the second method is label-free detection. Out of these two methods, fluorescence-based detection method uses the tags placed on biomolecules to detect them. The Fluorescence method is an accurate method but has a disadvantage of laborious work due to the labelling of all the samples. Label-free detection method can be done using surface plasmon resonance (SPR), waveguide, photonic crystal fiber, Bragg gratings, etc [5]–[7]. Label-free detection is also low cost method compared to fluorescent method [8].

SPR sensors are applicable in drug discovery in many cancer cases [9]. Drug-serum interactions can also be bounded by using surface plasmon resonance technology detection in biosensors [10]. Localized surface plasmon biosensors are capable of sensing biomolecules and also helpful in the detection of Alzheimer’s diseases [11]. Graphene is sensitive to SPR which is generated through nanoparticles. Metal nanoparticles are illuminated to generate surface plasmons [12]. Graphene is single layer graphite allotrope having high conductivity, good electrical and optical properties [13]. Graphene-based surface plasmon resonance sensors creating interest nowadays because of the excellent optical and electrical properties of graphene. Their excellent properties are mixed with SPR to create sensors applicable in biomedical applications [12]–[14]. SPR photonic crystal fiber is analyzed to give highly sensitive biosensing results [15]. There is another important biosensor based on SPR which uses refractive index difference for sensing of biomolecules [16]. Refractive index biosensors are one of the important label-free detection methods for biosensing. Biosensors utilize the refractive index change to detect the biosensing elements [17], [18].

Sensitivity is very important while designing any optical biosensors [19] and here we propose a highly sensitive refractive index sensor based on graphene metasurface array. The need for this sensitive biosensor is generated because highly sensitive biosensor will be helpful effectively in sensing hemoglobin biomolecules. The refractive index sensor is also analyzed for different concentrations of hemoglobin biomolecules. The proposed biosensor is also analyzed by varying different physical geometrical parameters. The comparison with the previously published similar researches is also presented to show the improvement in sensitivity.

The design and analysis of biosensor is presented in Section-II and results and discussions are presented in Section-III. The conclusions are presented in Section-IV.

II. BIOSENSORS DESIGN AND ANALYSIS

In this section, we present the biosensor design with its different parameter analysis. The biosensor design is presented in Fig. 1 and different parameter analysis is given in equations (1-12). The different parameters analyzed are the sensitivity, graphene conductivity, absorption and reflectance. The gold metasurface array is placed as shown in Fig. 1. The 3×3 array
The parameters used in the above equations are: \( \varepsilon_0 \) - permittivity of free space, \( \omega \) - angular frequency, \( \hbar \) - Planck’s constant, \( e^- \) electron charge value, \( k_B \) - Boltzmann’s constant and \( \sigma_s \) - monolayer conductivity, \( \Gamma \) - scattering rate, temperature - \( T \) (300 K), \( \mu_c \) - chemical potential. Graphene sheet thickness is kept 0.34 nm.

Graphene material’s conductivity mainly depends on its chemical potential. Graphene’s chemical potential is noted by \( \mu_c \) and is represented by \( \mu_C = \hbar v_F / \sqrt{\pi e^2 v_D C / \varepsilon} \), where \( V_{DC} \) - gate voltage and capacitance \( C \) is given by \( C = \varepsilon_0 \varepsilon_R / t \), \( \varepsilon_R \) - dielectric layer static permittivity, t - dielectric layer thickness. This graphene chemical potential can be varied to check its effect on the conductivity of the graphene. The change in conductivity of the graphene sheet impacts the result of the design.

The reflectance of metasurface biosensor can be calculated using the following equations (6-10) [14]. The reflectance is observed for the incident angle (\( \theta_l \)). The wave vector (\( k \)) can be calculated using this angle and frequency \( k = \omega \sin \theta_l / c \). The reflectance is given by:

\[
\begin{align*}
  r(\omega, \theta_l) &= \frac{\omega \cos \theta_l \sum_0 (\omega, \theta_l)}{2i \hbar c k^2 + \omega \cos \theta_l \sum_0 (\omega, \theta_l)}, \\
  \prod_\mu (\omega, \theta_l) &= \text{graphene polarization tensor with } \mu, \nu = 0,1,2 \text{ and } \prod_{tr} = \prod_\mu. \\
  \text{The conductivity is:} \\
  \sigma_{||}(\omega, k) &= -i \frac{\omega}{4\pi \hbar k^2} \sum_0 (\omega, k), \\
  \text{Now the reflectance:} \\
  r(\omega, \theta_l) &= \frac{2\pi \cos \theta_l \sigma_{||}(\omega, k)}{c + 2\pi \cos \theta_l \sigma_{||}(\omega, k)}, \\
  R(\omega, \theta_l) &= |r(\omega, \theta_l)|^2, \\
  \text{Considering the complex part:} \\
  R(\omega, \theta_l) &= \frac{4\pi^2 \cos^2 \theta_l \left[ \text{Re}^2 \sigma_{||}(\omega, k) + \text{Im}^2 \sigma_{||}(\omega, k) \right]}{[c + 2\pi \cos \theta_l \text{Re} \sigma_{||}(\omega, k)]^2 + 4\pi^2 \cos^2 \theta_l \text{Im}^2 \sigma_{||}(\omega, k)}, \\
  \text{Now if } \theta_l = k=0, \text{ The reflectance for the incident wave is given by:} \\
  R(\omega) &= R(\omega, 0) = \frac{4\pi^2 \left[ \text{Re}^2 \sigma(\omega) + \text{Im}^2 \sigma(\omega) \right]}{[c + 2\pi \text{Re} \sigma(\omega)]^2 + 4\pi^2 \text{Im}^2 \sigma(\omega)}, \\
  A(\omega) &= 1 - R(\omega) - T(\omega), \\
  \text{It is observed from the equations (6-12) that reflectance is closely related to conductivity. The conductivity is improved using graphene material. The absorption of the material depends on the transmittance and reflectance as given in equation (12). The absorption results of the proposed biosensor are discussed in detail in section III.}
\end{align*}
\]

### III. Result Analysis

The graphene-based refractive index biosensor using gold metasurface array design presented in the Fig. 1 is simulated using COMSOL Multiphysics for different concentrations of hemoglobin biomolecules. The results are analysed for absorption values, electric field and magnetic field (Fig. 2-5). We have also varied the geometrical physical parameters like...
metasurface thickness ($T_G$) and SiO$_2$ layer thickness ($T_S$) to observe its effect on the response. We have experimentally found out the refractive index for different concentrations of the hemoglobin and the data are compared in Table I. The different concentrations taken are 10, 20, 30 and 40 g/l. The refractive indexes found out are 1.34 RIU, 1.36 RIU, 1.39 RIU and 1.43 RIU respectively for these concentrations [23], [24].

We have analyzed our graphene-based metasurface array design for these hemoglobin biomolecules and the absorption response obtained is presented in Fig. 2. The response is observed between the wavelength range of 0.2 µm and 1.2 µm. The absorption peaks are observed between the wavelength range of 0.7 µm and 0.85 µm. The absorption for this range is enhanced and presented as inset in the figure. From the inset it is clear that $\Delta \lambda = 70$ nm (730 nm to 800nm) wavelength shift is observed between hemoglobin biomolecules with 1.34 RIU and 1.36 RIU with $\Delta n = 0.02$ RIU [23], [24]. The sensitivity of this biosensor is 3500 nm/RIU. The sensitivity is calculated by putting the wavelength and refractive index change value in equation (1).

Optimization of biosensor geometrical parameters:

The physical geometrical parameters SiO$_2$ thickness ($T_S$) and metasurface array thickness ($T_G$) of the graphene-based metasurface array biosensor design is varied to observe its effect on the absorption parameter. The SiO$_2$ thickness is varied from 0.3 µm to 0.8 µm and absorption response is observed between 0.5 µm to 1.2 µm wavelength range (Fig. 3). From the response, it is clear that as the thickness of the SiO$_2$ layer increases the absorption is increased to a certain limit and then it remains almost the same. This is observed because the increase in SiO$_2$ layer absorbs more light in the layer but after a certain limit the phenomenon becomes ineffective and absorption remains the same by increasing from that limit. The same thing is observed in the response and the absorption is maximum at 0.6 µm and after that increase in the SiO$_2$ thickness does not affect the absorption much and it remains the same as shown in Fig. 3.

The absorption response for different values of metasurface array thickness ranging from 0.05 µm to 0.3 µm. The high values of absorption are observed between 0.55 µm to 1.05 µm wavelength range and the highest absorption is observed for 0.6 µm thickness.

**Table I**

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<th>Hemoglobin</th>
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<td></td>
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**Fig. 2.** Absorption response of graphene-based metasurface array biosensor design for wavelength ranging from 0.2 µm and 1.2 µm and hemoglobin biomolecules for different concentrations with refractive index 1.34 RIU, 1.36 RIU, 1.39 RIU and 1.43 RIU. The peaks in the waveform are observed between 0.7 µm and 0.85 µm wavelength. The inset figure is showing the wavelength peak difference of 70 nm between n = 1.34 RIU and n = 1.36 RIU.

**Fig. 3.** Absorption response for different values of SiO$_2$ thickness ranging from 0.3 µm to 0.8 µm. The high values of absorption are observed between 0.6 µm to 1 µm wavelength range and the highest absorption is observed for 0.6 µm thickness.

**Table II**

<table>
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The electric field intensity and magnetic field density are presented for the biosensor design in Fig. 5. The results are...
The graphene-based highly sensitive biosensor using meta-
surface array is presented. The metasurface array in the form
of C-shaped split-ring resonators and thin wires are used to
improve the sensitivity in this design. The designed biosensor
is having the highest sensitivity of 3500 nm/RIU. The broad
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that the designed biosensor is also having the best results for
0.6 µm SiO2 thickness and 0.1 µm metasurface array thickness.
The proposed biosensor is also compared with previously
published similar sensors [23]–[26] and the sensitivity is found
highest in the proposed sensor. The advantage of this biosensor
is its sensitivity. The disadvantage of this biosensor is that
its only applicable in sensing hemoglobin biomolecules. The
proposed biosensor has the capability to become a building
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AQ:4 = Please confirm the postal code for Ton Duc Thang University.
AQ:5 = Please provide an updated Table 2, which reflects the changes made to the numbering of references.
AQ:6 = Please provide the publisher name and publisher location for Ref. [9].
AQ:7 = Please note that Refs. [14] and [23] were identical in your originally submitted manuscript. Hence, we have deleted Ref. [23] and renumbered the subsequent references. This will also be reflected in the citations present in the body text.
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I. INTRODUCTION

Optical biosensors are very effective amongst all the biosensors because they detect biomolecules directly and in real-time [1], [2]. Optical biosensors are also widely applicable in medical applications [2], environment applications [3], industrial applications [4], etc; because of this fast detection in real-time [2]–[4]. Optical biosensors can be used to detect or sense biomolecules by two main detection methods. The first method to detect is based on fluorescence and the second method is label-free detection. Out of these two methods, fluorescence-based detection method uses the tags placed on biomolecules to detect them. The Fluorescence method is an accurate method but has a disadvantage of laborious work due to the labelling of all the samples. Label-free detection method can be done using surface plasmon resonance (SPR), waveguide, photonic crystal fiber, Bragg gratings, etc [5]–[7]. Label-free detection is also a low cost method compared to fluorescent method [8].

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The design and analysis of biosensor is presented in Section-II and results and discussions are presented in Section-III. The conclusions are presented in Section-IV.

II. BIOSENSORS DESIGN AND ANALYSIS

In this section, we present the biosensor design with its different parameter analysis. The biosensor design is presented in Fig. 1 and different parameter analysis is given in equations (1-12). The different parameters analyzed are the sensitivity, graphene conductivity, absorption and reflectance. The gold metasurface array is placed as shown in Fig. 1. The 3 × 3 array
of metasurface array elements in the form of C-shaped splitting resonators and thin wires are placed over the graphene layer which is based on SiO$_2$ substrate. The light falls on the metasurface array layer which concentrates it on the graphene layer and substrate layer. Graphene’s high conductivity helps in increased absorption in SiO$_2$ layer. The total length of the structure is kept at 2 µm. The thickness of SiO$_2$ layer (T$_S$) is 0.6 µm, thickness of gold metasurface elements (T$_G$) is 0.1 µm and thickness of graphene layer is 0.34 nm. The width of C-shape element (C) is 0.55 µm. The biomolecule layer is placed above the metasurface elements and the thickness of biomolecule layer is kept at 1.4 µm.

SPR biosensors having angle of incidence fixed, the sensitivity is defined by the ratio of shift in wavelength obtained by varying the refractive index of biomolecules [20]:

$$S = \frac{\Delta \lambda}{\Delta n},$$

where S is the sensitivity of biosensor, $\Delta \lambda$ is the wavelength difference for two different materials used for biosensing and $\Delta n$ is the refractive index difference between these two biomolecules.

Graphene is very important in designing biosensor because of its excellent optical and electrical properties. The conductivity of graphene monolayer is given by Kubo formula in equations (2-5) [21], [22]:

$$\varepsilon (\omega) = 1 + \frac{\sigma_0}{\varepsilon_0 \omega \Delta},$$

$$\sigma_{\text{intra}} = -\frac{j e^2 k_B T}{4 \pi \hbar} \left( \frac{\mu c}{k_B T} + 2 \ln \left(\frac{\mu c}{\gamma k_B T} + 1\right) \right),$$

$$\sigma_{\text{inter}} = -\frac{j e^2}{4 \pi \hbar} \ln \left(\frac{2 |\mu c| - (\omega - j 2 \Gamma) \hbar}{2 |\mu c| + (\omega - j 2 \Gamma) \hbar}\right),$$

$$\sigma_s = \sigma_{\text{inter}} + \sigma_{\text{intra}},$$

The parameters used in the above equations are: $\varepsilon_0$ - permittivity of free space, $\omega$ - angular frequency, $\hbar$- Planck’s constant, $e$- electron charge value, $k_B$- Boltzmann’s constant and $\sigma_s$- monolayer conductivity, $\Gamma$ - scattering rate, temperature - $T$ (300 K), $\mu_c$ - chemical potential. Graphene sheet thickness is kept 0.34 nm.

Graphene material’s conductivity mainly depends on its chemical potential. Graphene’s chemical potential is noted by $\mu$ and is represented by $\mu = \mu_c + e V_{DC} / \pi e^2 / \varepsilon_0$, where $V_{DC}$- gate voltage and capacitance $C$ is given by $C = \varepsilon_0 \varepsilon_{0D} / (\pi L_d^2)$, $\varepsilon_{0D}$-dielectric layer static permittivity, t-dielectric layer thickness. This graphene chemical potential can be varied to check its effect on the conductivity of the graphene. The change in conductivity of the graphene sheet impacts the result of the design.

The reflectance of metasurface biosensor can be calculated using the following equations (6-10) [14]. The reflectance is observed for the incident angle $(\theta_i)$. The wave vector $(k)$ can be calculated using this angle and frequency $k = \omega \sin \theta_i / c$.

$$r(\omega, \theta_i) = \frac{\omega \cos \theta_i \prod_{\text{00}} (\omega, \theta_i)}{2 i h c k^2 + \omega \cos \theta_i \prod_{\text{00}} (\omega, \theta_i)},$$

$$\prod_{\mu, \omega} (\omega, \theta_i)$$ is graphene polarization tensor with $\mu$, $\nu = 0, 1, 2$ and $\prod_{r, \omega} = \prod_{\nu, \mu} .$$

The conductivity is:

$$\sigma_{\|}(\omega, k) = \frac{\omega}{4 \pi h k^2} \prod_{\text{00}} (\omega, k),$$

Now the reflectance:

$$r (\omega, \theta_i) = \frac{2 \pi \cos \theta_i \sigma_{\|}(\omega, k)}{c + 2 \pi \cos \theta_i \sigma_{\|}(\omega, k)},$$

$$R (\omega, \theta_i) = \left| r (\omega, \theta_i) \right|^2,$$

Considering the complex part:

$$R (\omega, \theta_i) = \frac{4 \pi^2 \cos^2 \theta_i [\Re^2 \sigma_{\|}(\omega, k) + \Im^2 \sigma_{\|}(\omega, k)]}{[c + 2 \pi \cos \theta_i \Re \sigma_{\|}(\omega, k)]^2 + 4 \pi^2 \cos^2 \theta_i \Im^2 \sigma_{\|}(\omega, k)},$$

Now if $\theta_i = k = 0$, The reflectance for the incident wave is given by:

$$R (\omega) = R (\omega, 0) = \frac{4 \pi^2 \left[\Re^2 \sigma (\omega) + \Im^2 \sigma (\omega)\right]}{[c + 2 \pi \Re \sigma (\omega)]^2 + 4 \pi^2 \Im^2 \sigma (\omega)},$$

$$A (\omega) = 1 - R (\omega) - T (\omega),$$

It is observed from the equations (6-12) that reflectance is closely related to conductivity. The conductivity is improved using graphene material. The absorption of the material depends on the transmittance and reflectance as given in equation (12). The absorption results of the proposed biosensor are discussed in detail in section III.

III. RESULT ANALYSIS

The graphene-based refractive index biosensor using gold metasurface array design presented in the Fig. 1 is simulated using COMSOL Multiphysics for different concentrations of hemoglobin biomolecules. The results are analysed for absorption values, electric field and magnetic field (Fig. 2-5). We have also varied the geometrical physical parameters like...
TABLE I

COMPARATIVE TABLE OF HEMOGLOBIN RIU FOR DIFFERENT CONCENTRATIONS

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Fig. 3. Absorption response for different values of SiO$_2$ thickness ranging from 0.3 $\mu$m to 0.8 $\mu$m. The high values of absorption are observed between 0.6 $\mu$m to 1 $\mu$m wavelength range and the highest absorption is observed for 0.6 $\mu$m thickness.

TABLE II

THE COMPARISON OF THE SENSITIVITY OF THE PROPOSED DESIGN COMPARED TO THE PREVIOUSLY PUBLISHED DESIGNS [25]–[28]

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The absorption response for different values of metasurface array thickness ($T_G$) is observed for 0.5 $\mu$m to 1.2 $\mu$m wavelength range. The thickness is varied from 0.05 $\mu$m and 0.3 $\mu$m range. The absorption is observed in the high range for 0.55 $\mu$m and 1.05 $\mu$m frequency range for all the values of thickness. The higher absorption is observed for the low values of thickness. The highest absorption is observed for 0.1 $\mu$m metasurface array thickness. This is observed because if we increase the thickness than there is a chance of more absorption in the metasurface rather than in the substrate. The concentration of light to the substrate is also decreased.

The electric field intensity and magnetic field density are presented for the biosensor design in Fig. 5. The results are
The graphene-based highly sensitive biosensor using metasurface array is presented. The metasurface array in the form of C-shaped split-ring resonators and thin wires are used to improve the sensitivity in this design. The designed biosensor is having the highest sensitivity of 3500 nm/RIU. The broad absorption spectrum in the range of 0.5 µm to 1.1 µm wavelength is observed with more than 80% absorption. The designed biosensor is analyzed for different geometrical physical parameters of the design. From the results, it is evident that the designed biosensor is also having the best results for 0.6 µm SiO₂ thickness and 0.1 µm metasurface array thickness. The proposed biosensor is also compared with previously published similar sensors [23]–[26] and the sensitivity is found highest in the proposed sensor. The advantage of this biosensor is its sensitivity. The disadvantage of this biosensor is that its only applicable in sensing hemoglobin biomolecules. The proposed biosensor has the capability to become a building block for future medical and biosensing photovoltaic devices.

IV. CONCLUSION

The graphene-based highly sensitive biosensor using metasurface array is presented. The metasurface array in the form of C-shaped split-ring resonators and thin wires are used to improve the sensitivity in this design. The designed biosensor is having the highest sensitivity of 3500 nm/RIU. The broad absorption spectrum in the range of 0.5 µm to 1.1 µm wavelength is observed with more than 80% absorption. The designed biosensor is analyzed for different geometrical physical parameters of the design. From the results, it is evident that the designed biosensor is also having the best results for 0.6 µm SiO₂ thickness and 0.1 µm metasurface array thickness. The proposed biosensor is also compared with previously published similar sensors [23]–[26] and the sensitivity is found highest in the proposed sensor. The advantage of this biosensor is its sensitivity. The disadvantage of this biosensor is that its only applicable in sensing hemoglobin biomolecules. The proposed biosensor has the capability to become a building block for future medical and biosensing photovoltaic devices.

REFERENCES