Tuning and Sensitivity Improvement of Bi-metallic Structure-based Surface Plasmon Resonance Biosensor with 2-D ξ -Tin Selenide nanosheets

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Abstract

This manuscript aims to visualize the effect of tin selenide (SnSe) on the sensing application of the SPR biosensors. Tin selenide is the 2-Dimensional material transition metal dichalcogenide family element. By utilizing the extraordinary properties of the SnSe, high sensitivity and high performance of the biosensor is analyzed by utilizing the extraordinary properties of the SnSe. A unique two-dimensional substantial heterostructure, ϵ -tin selenide (SnSe) /graphene layer has been deposited over the metal surface to improve the sensitivity; moreover, the sensitivity is limited to a certain extent. The ϵ – SnSe nanosheet is placed in between two layers of gold (Au) in the Kretschmann arrangement. The proposed configuration has a maximum sensitivity of $214^\circ/\text{RIU}$, which is 93.81% higher than the conventional sensor. The performance parameters like FWHM, detection accuracy and quality factor have been analysed. The ϵ – SnSe material is an air-stable 2-D nanosheet and has application in chemical, medical, and biological sensors.

1. Introduction

In the last few decades, the tremendous development of Surface Plasmon Resonance (SPR) technology has been reported for biomedical applications. Detecting cancer cells, DNA Hybridization, antibody characterization, protein conformation, and the recent detection of the CORONA virus has proved that the technique is more beneficial for real-time applications. In the biosensing field, to detect, analyze and characterize the biomolecule, chemical, environment, and food [1]–[5], these biosensors are highly sensitive and facilitate real-time analysis of the contaminated analyte [6]–[8]. The SPR biosensor works on the phenomenon of the shift of resonance dip. As the refractive index of the sensing layer varies [9], [10], a sharp dip in the resonance curve is obtained due to the adsorption of the incident light in the sensing medium. The Kretschmann configuration is the most acceptable configuration for designing the biosensor [11], [12]. This configuration has a and has a thin layer of metal. The layer of metal is used to absorb the biomolecules or chemical molecules. The metal has poor adsorbability, so few layers of 2-dimensional material is associated with enhancing the biosensors' performance [13]–[17]. When an incident light beam propagates through two media interfaces with the different refractive indexes at a particular angle and suitable dielectric constants (typically a metal with high concentration of free electrons and a dielectric medium), the Surface Plasmon gets excited [18].

Single-layer SnSe also possesses fantastic electrical and optical properties and generates high interest as a 2D material beyond the predecessor members, which displays the same structure as

graphene and phosphorene [19]–[21]. As a classical p-type IV-VI semiconductor, SnSe has a narrow gap [22]. That SnSe has a layered crystal structure similar to those of other IV-VII binary semiconductors, such as SnSe, GeS, GeSe, etc. In experiments, the assumption that unilaminar crystal SnSe would display different thermoelectric properties on different axes was authenticated by Zhao et al. [16], who manufactured hole-doped single-crystal SnSe [18]. A lot of SnSe allotropes have been investigated by density functional theory [16]. Monolayer SnSe not only has outstanding thermoelectric features with

semiconductor properties, but is also an eminent optoelectronic material, but there are few systematic studies on the optic and electric characteristics of SnSe allotropes. A metal like Gold (Au) is not susceptible to oxidation and does not react with most chemicals; and hence it is often used as the metal film in sensors. At the metal-dielectric interface, the intensity of the incident light beam reaches a maximum and decays exponentially into both the medium [23]. Since the metal and wave interaction is lossy, the EM Field of a Surface Plasmon is concentrated in a vast majority in the dielectric medium close to the metal surface. The SPR phenomenon results in a graded reduction in the intensity of the reflected light. Surface Plasmon Resonance (SPR) sensors have generated considerable interest since they allow real-time detection of biomolecular interactions, rapid, level free, and valuable diagnostic tool for diseases having virus size of the order of nanometer or less [24].

In particular, graphene has attractive properties such as tunable electrical & optical characteristics [25], ring-type carbon structure & high surface-to-volume ratio. Thus the adsorbates can easily interact with this structure, increasing the adsorption suitable for biosensors [26], [27].

Enhancing the sensor's sensitivity is an emerging issue and is being studied at different places; different methods and strategies have been proposed. Conventionally Gold (Au) or Silver (Ag) having high free electrons closed to the boundary was chosen, and superimposed over it a dielectric medium of finite thickness helped in generating charge density oscillation at the metal-dielectric interface when monochromatic light was incident over it [28]. SPR technique help in estimating the dimension of DNA [29], RNA or Blood cells which cannot be measured directly due to its sizes less than nanometer; therefore, the indirect measurement technique, which was evolved in 1968 by Kretschmann, opened a new avenue where the refractive index variation interacting with incident light photon and the reflected ray thus quantify the size or the nature of the virus under investigation. The binding of the legend differs; therefore, SPR has successfully detected the diverse effect and resulting refractive index change, for example protein- protein and DNA-DNA etc. [30], [31]. SnSe can enhance the sensitivity of the biochemical sensors. It is an indirect bandgap 2-D semiconductor material, stable in the air.

The manuscript is sectioned as follows, Sect. 2 proposed theoretical model and design consideration for the designed biosensor. Section 3 consists of the results and discussion, and finally, Sect. 4 concludes the proposed work.

2. Theoretical Model And Design Consideration

We consider a modified biosensor configuration, as shown in Fig. 1. Each layer of the materials is assumed to be stacked along the z-axis. BK7 is chosen as a coupling prism in our structure, which is covered with one layer of gold-1, $\epsilon - \text{SnSe}$ layer is close to gold-1 layer, and the other side of $\epsilon - \text{SnSe}$ layer also connects with another gold-2 layer. Graphene layer is used as a biomolecular recognition element, which is close to the gold-2 layer. Besides, the thickness of the two layers of gold is not the same. We set the thickness of two layers of gold as $W_1 = 40 \text{ nm}$ and $W_3 = 20 \text{ nm}$, respectively. The thickness of $\epsilon - \text{SnSe}$ is $W_2 = \text{SnSe} \times 1.5 \text{ nm}$ where SnSe is the layer number of $\epsilon - \text{SnSe}$ sheets.

Graphene is connected with the sensing layer for increasing the ability of macromolecular adsorption of the biosensor, and the thickness of graphene is $W_2 = G \times 0.34nm$, where G is the layer number of graphene sheets. A TM-polarized light is assumed to be incident from one side of the prism and receive reflected light from the other side of the prism. The received data can be used to calculate the reflectivity and sensitivity of the structure.

The refraction index (n_{Prism}) of BK7 layer is expressed as [32]

$$n_{Prism} = \left(\frac{1.03961212\lambda^2}{\lambda^2 - 0.00600069867} + \frac{0.231792344\lambda^2}{\lambda^2 - 0.0200179144} + \frac{1.0104694\lambda^2}{\lambda^2 - 103.560653} + 1 \right)^{\frac{1}{2}}$$

1

where λ is the wavelength of the optical signal.

The RI of the analyte/metal can be computed using the Drude model [33] by the formula:

$$n_{1,3}(\lambda) = \sqrt{1 - \frac{\lambda^2 \lambda_c}{\lambda_p^2 (\lambda_c - i\lambda)}}, \text{ where } \lambda_p \text{ and } \lambda_c \text{ are the wavelengths of silver for plasma and collision}$$

and are taken as $1.6826 \times 10^{-7}m$ and $8.9342 \times 10^{-5}m$, respectively [34].

At $\lambda = 633nm$ the refractive index of $\epsilon - SnSe$ is considered as $4.4 + 3.53i$ [23] in the visible region. In the proposed multi-layered structure, reflectivity is calculated by formula as follows:

$$r_{pm_1d_1m_2d_2} = \frac{r_{pm_1} + r_{m_1d_1m_2d_2} e^{2iK_{m_1}x_{dm_1}}}{1 + r_{pm_1}r_{m_1d_1m_2d_2} e^{2iK_{m_1}x_{dm_1}}}$$

2

where

$$K_{ix} = \sqrt{\left(\frac{2\pi}{\lambda}\right)^2 \epsilon_i - K_z^2} \quad i = prism, metal_1, dielectric_1, metal_2, dielectric_2$$

3

$$r_{prismmetal} = \frac{(\epsilon_{metal}K_{prismx} - \epsilon_{prism}K_{metalex})}{(\epsilon_{metal}K_{prismx} + \epsilon_{prism}K_{metalex})}$$

$$r_{metaldielectric} = \frac{(\epsilon_{dielectric}K_{metalx} - \epsilon_{metal}K_{dielectrix})}{(\epsilon_{dielectric}K_{metalx} + \epsilon_{metal}K_{dielectrix})}$$

The condition for resonance is fulfilled using the Eqs. 2 and 3. The equation computes the sharp downfall in the resonance curve:

$$K_z = K_{sp} = \frac{2\pi}{\lambda} n_0 \sin \theta_{SPR} = \text{real} \left(\frac{2\pi}{\lambda} \sqrt{\frac{\epsilon_m \epsilon_s}{\epsilon_m + \epsilon_s}} \right)$$

4

where n_0 is the RI of the sensing layer, ϵ_s and ϵ_m is the dielectric constant for the sensing layer and analyte/Metal, θ_{SPR} is resonance angle and K_{sp} is propagation constant. The RI of the graphene film is given as $n_5 = 3 + i\frac{C}{3}\lambda$, where $C = 5.44\mu m^{-1}$ [20]. The RI of sensing film is taken as $n_s = 1.373$. The sensitivity and detection accuracy are the performance parameters of the biosensor, and the same is computed for the proposed biosensor. The sensitivity of the biosensor is represented as $S = \delta\theta_{SPR} / \delta n$, where δn is the change in the RI. The detection accuracy (DA) of the biosensor can be expressed as $DA = 1 / FWHM$, where FWHM is the width of the spectra of the SPR curve at that point, the reflectivity is 50% of the maximum value. The high value of the sensitivity and DA is desirable [35]. The quality factor can be expressed as $Q = S \cdot DA = S / FWHM$, directly depends on the FWHM and sensitivity. Table 1 shows the optimized parameters selected for the theoretically examined biosensor.

Table 1
Optimized parameters for the proposed biosensor

Film of the materials	Used material	RI of the material at 633 nm		Thickness (nm)
		Real part (n)	Imaginary part (k)	
Layer I	Prism BK7	1.5151	-	-
Layer II	Au (Metal)	0.18377	3.4313i	$w_1 = 40$
Layer III	ϵ – Tin Selenide	4.4	3.53i	$w_2 = M*1.5$
Layer IV	Au (Metal)	0.18377	3.4313	$w_3 = 20$
Layer V	Graphene	2.4105	-	$w_4 = 0.34*N$

3. Results And Discussions

Emphasis has been made to improve the sensitivity and performance of biosensor low refractive index prism is used. BK7 prism is most suitable because it has a low refractive index. If the prism has a high

refractive index, it provides the sharp dip in the resonance curve compared to the low refractive indices' prism. Low refractive prism provides better values of the FWHM, angle of resonance, the shift in the resonance curve, and sensitivity than the high refractive index's prism. To get the high sensitivity and FWHM, the flat end of the prism is polished with a 40nm thick gold layer. On the top of the Au layer, few layers of the SnSe are deposited to examine the structure's sensitivity. Another Au layer of 20 nm is deposited over the SnSe layer to enhance the sensor's sensitivity. A single metal layer cannot increase sensitivity too much, so using bimetallic layer is used. In the end, the graphene layer is spread over the gold-1 layer.

To extend the discussion, a single layer of the graphene is present, and the SnSe layer is missing in the structure, i.e., $\text{SnSe} = 0$, $G = 1$. Now the impact of the graphene is analyzed, and it is found that offset resonance dip and sensitivity both are improvised and values of $\delta\theta_{SPR} = 0.99^\circ$ and $198^\circ/\text{RIU}$ are claimed respectively (shown in Fig. (c)). The enhanced sensitivity and the increase in resonance curve offset dip in this structure are attributed to the higher adsorption rates due to the large surface area and rich π conjugation structure offered by graphene, making it the appropriate choice for dielectric top layer SPR sensing applications [25], [36].

Another scenario was evaluated for the impact by introducing a single layer of SnSe and a monolayer of graphene in the biosensor ($\text{SnSe} = 1$, $G = 1$). This results in significant shifts and a dip in the resonance curve. The resonance angle was obtained as $\delta\theta_{SPR} = 1.02^\circ$, whereas the sensitivity was $214^\circ/\text{RIU}$ as depicted in Fig. 2 (d). This comparative study reveals that the resonance angle offset increases with the addition of a single layer of graphene, a single layer of SnSe, compared to the conventional sensor. The best results were obtained by adding a single layer of graphene & a single layer of SnSe. Table 2 consists of performance parameters that are directly obtained from the theoretical study of the proposed work. Based on our knowledge, few parameters are better reported than the previously existing literature.

Table 2
Final values of the different parameters for the sensor

Structures (Prism BK7)	Resonance Angle (θ_{SPR})	$\delta\theta$	FWHM	Sensitivity	Quality Factor	DA	$R_{\min}$
1. Bimetallic layer (Au)	76.86	0.97	4.434	194	46.01	0.23	0.22
2. Bimetallic layer + $\epsilon - \text{SnSe}$	77.42	0.98	7.039	196	30.12	0.18	0.30
3. Bimetallic layer + Graphene	77.44	0.99	5.41	206	38.08	0.14	0.43
4. Bimetallic layer + Graphene + $\epsilon - \text{SnSe}$	78.02	1.02	7.947	214	26.93	0.13	0.49

By comparing the relevant data, we can find that the resonance angle offset of the biosensor in our the addition of graphene and the $\epsilon - \text{SnSe}$

layer. The traditional SPR structure contains only a prism and gold layer. Hence, we can conclude that the sensitivity of biosensors in our structure has significantly improved compared with the traditional design by adding graphene and ϵ – SnSe layer.

From the analyses made above, it is clear that the refractive index of the sensing layer can control the sensitivity of the biosensor. To better understand the enhancement of the sensitivity of the biosensor by graphene and ϵ – SnSe layers, we also plot the dependence of the sensitivity of the biosensor on the refractive index of the sensing layer, as shown in Fig. 3. Here we select the sensing layer refractive index range from 1.368 to 1.373. It can be seen that as the refractive index of the sensing layer increases from 1.368 to 1.373, the sensitivity increases significantly.

Table 3
Depicts the analytical values at which the quality factor obtained is maximum

LAYERS	FWHM	DETECTION ACCURACY	SENSITIVITY	QUALITY FACTOR	MINIMUM REFLECTANCE
SnSe = 1 G = 1	7.947	0.1258	214	26.9	0.487
SnSe = 2 G = 1	9.774	0.1023	196	20.1	0.6077
SnSe = 1 G = 2	8.798	0.1137	212	24.1	0.5376

Table 3 comprises the performances of the biosensor for the maximum change in the RI is 0.005, the thickness of the Au layers is chosen 40nm and 20nm, the thickness of the ϵ – SnSe is 1.5nm, and thickness of the graphene 0.34. Table 4 reports the comparative analysis of the earlier published work. Table 4 reflects that all the referenced work is published, and it can consider that all are standard. The proposed work here shows the best performance parameters as well as high sensitivity.

Table 4
Comparative analysis with the earlier reported work

Layers	Wavelength λ	Sensitivity (deg/RIU)	Reference
Chromium, Gold, SnSe nanosheet, sensing layer	633nm	160	[37]
Silver, SnSe, sensing layer	633nm	154	[23]
Gold, SnSe, Gold, graphene, sensing layer	633nm	214	Proposed work

4. Conclusions

The proposed biosensor is numerically simulated and performance parameters is also analyzed for the bimetallic (Au)/ ϵ – SnSe/graphene nanosheet based modified Kretschmann configuration. The study is carried out at 633nm operating wavelength, and highest sensitivity 214 deg/RIU is obtained for the mono layers of the ϵ – SnSe allotrope and graphene. The thickness of the gold layers is 40 and 20 nm respectively is taken in this work. The refractive index of the medium is taken as 1.368. Proposed sensor is useful for the study of the chemical, environment, biomolecule and analytes.

Declarations

Code Availability: Not applicable.

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Availability of data and material: No data available.

Authors' contributions

NS Formulated the problem statement wherein giving the theoretical background for SPR biosensor. He also helped in drafting the manuscript.

BK formulated the problem statement and improvised the quality of the figure and graph.

KPR provided the theoretical background to biosensing and the importance of Optical Biosensing. He also helped in reviewing the manuscript.

AJ provided statistical analysis for the results. He provided the theoretical background to SPR biosensors.

AP provided statistical analysis for the results. He provided the theoretical background to SPR biosensors. He also helped in reviewing and formatting the manuscript.

Ethics approval

Not applicable. The work presented in this manuscript is mathematical modelling only for the proposed biosensor. No experiment was performed on the human body and/or living organism/ animal. So, an ethical approval from an ethical committee is not required.

Consent to participate

All the author willing to participate in the work presented in this manuscript. On the behalf of all author, communicating author is authorized to give their consent.

Consent for publication

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Figures

Figure 1

Heterostructure design of the proposed biosensor

Figure 2

Reflectance vs. incident angle (θ) at the same RI change with ϵ -SnSe and Graphene (a) $\text{SnSe}=0, G=0$, (ii) $\text{SnSe}=0, G=1$, (iii) $\text{SnSe}=1, G=0$, (iv) $\text{SnSe}=1, G=1$

Figure 3

Plot of Sensitivity (Deg/RIU) vs. Sensing layers refractive index

Figure 4

(a), (b) Change in the reflectance w.r.t incident angle (a) variation in the number of tin selenide layers at one graphene layer and (b) variation in the number of graphene layers at one tin selenide.

(a)

(b)

Figure 5

Graphical representation comparing (a) the sensitivity with FWHM and Minimum Reflectance of ϵ -SnSe layers and graphene layer, (b) the sensitivity with Quality Factor and Detection Accuracy of ϵ -SnSe layers and graphene layer