Zinc Sulfide, Silicon Dioxide, and Black Phosphorus Based Ultra-Sensitive Surface Plasmon Biosensor

Bhishma Karki (✉ magnum.photon@gmail.com)

Tribhuvan University - Tri-Chandra Multiple Campus <https://orcid.org/0000-0003-4351-8099>

Youssef Trabelsi

College of Arts and Sciences in Muhail Asir, Physics Department, King Khalid University, Abha Saudi Arabia

Arun Uniyal

DIT University

Amrindra Pal

DIT University

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Zinc Sulfide, Silicon dioxide, and Black phosphorus based ultra-sensitive Surface Plasmon Biosensor

Bhishma Karki^{*1,2}, Youssef Trabelsi^{3,4}, Arun Uniyal⁵, Amrindra Pal⁵

¹Department of Physics, Tri-Chandra Multiple Campus, Tribhuvan University, Kathmandu-44600, Nepal

²National Research Council Nepal, New Baneshwor-10, Kathmandu-44600, Nepal

³College of Arts and Sciences in Muhail Asir, Physics Department, King Khalid University, Abha, Saudi Arabia.

⁴University of Tunis El Manar, Photovoltaic and Semiconductor Materials, ENIT, Belvedere 1002, Tunis, Tunisia.

⁵Department of ECE, DIT University, Dehradun-248009, India

*Corresponding Author: Email address: magnum.photon@gmail.com, Mob: +977-9851014005

Abstract: - The optical biosensor is the emerging research area in the field of bio-photonics. The black phosphorus zinc sulfide-based hybrid configuration is suitable for implementing and analyzing ultrasensitive biosensors. Ag/Zinc sulfide/silicon dioxide/black phosphorus-based biosensor has been implemented in the proposed work using the modified Kretschmann configuration. The sensitivity improvement of the designed SPR sensor is analyzed in the different arrangements of the layers. The thickness of the layers of all the materials has been optimized. The thickness of the Ag metal layer is optimized and taken as 45 nm. The sensitivity and quality factor measured here is as high as 664.6°/RIU and 200 at 1.37 refractive index—the P-polarized light source of 633nm wavelength. The proposed biosensor confirms tremendous growth in terms of sensitivity, detection accuracy, and quality factor compared with the traditional SPR sensors. Zinc sulfide has multiple applications in the sensing fields, like sensors based on UV rays, lasers, and gas.

Keywords: Surface plasmon resonance; Zinc Sulfide; SiO₂; Sensor

1. Introduction:

The surface plasmon resonance is a condition that exists when a P polarized wave is an incident on the interface of a metal and dielectric material through a coupling prism. Some part of incident light penetrates into the metal layer, generally known as an evanescent wave, causing excitation of the free electrons, which finally produces surface plasmon polaritons (SPPs) [1]. The matching of the wave vector and evanescent wave vector intensity two gives rise to resonance. The whole mechanism is based on the attenuation total reflection (ATR). The electrons on the metal surface absorb the incident wave, decreasing the reflected light [2]. The refractive index (RI) of metal generally varied in accordance with the incident angle known as surface plasmon resonance (SPR) angle [3], [4]. Surface plasmon resonance is gaining more popularity due to unique features like greater sensitivity, real-time response during monitoring, high detection accuracy, fast response,

etc. [5]. These sensors have a wide area of applications in the development of vaccines, drugs, doping, food safety testing, various diseases detection, gene treatments, and some other areas [6], [7].

The conventional sensors are implemented using the Otto or Kretschmann configuration, with coupling metal and sensing layers. The metals such as silver (Ag) [8], gold (Au) [9], aluminum (Al), and copper (Cu) [10] are primarily used in SPR design. The sensitivity obtained using the conventional configuration of the biosensor (i.e., prism, metal, and sensing layer) is generally low. So, to improve the sensitivity to an extent, modification in the conventional configuration is required. Mishra et al. [10] proposed an SPR sensor for deoxyribose nucleic acid (DNA) hybridization using Ag and graphene layer with silver nitride (Si_3N_4) achieved a maximum sensitivity of $212.2^\circ/\text{RIU}$ with figure of merit (FOM) and detection accuracy (DA) as 138.24 RIU^{-1} and 9.124, respectively. Singh and Raghuvanshi [11] theoretically investigate a gas detection SPR sensor design using Au layer (bi-metallic) and two BP layers, found the sensitivity as $245.5^\circ/\text{RIU}$ at the operating wavelength 633 nm. Pal and Jha [11] proposed SPR sensor design with the help of two silver metal layers and barium titanate (BaTiO_3) layer and achieved $280^\circ/\text{RIU}$ sensitivity. Maharana et al. [12] theoretically analyzed an SPR sensor design using Otto configuration with graphene film as the basic recognition element (BRE). The sensitivity of the proposed work was enhanced by 10.71% at 700 nm and by 5% at 1000 nm. Verma et al. [13] attained high sensitivity of SPR biosensor using an air gap as high as 2.35 times of basic graphene-based sensor in the absence of airgap. Chen et al. [14] designed an SPR based bio gas-sensor with MgF_2 as a coupling prism. The rhodium (Rh) and a silver (Ag) metal used as bimetallic layers found sensitivity as high as $220^\circ/\text{RIU}$. Bijalwan et al. give an SPR sensor design with graphene (2D-material) nanolayers and metal layers of Au, Al sensitivity of $160^\circ/\text{RIU}$ is calculated and with other combination of WSe_2 with Au and Al (bi-metal films) sensitivity of $163^\circ/\text{RIU}$ was obtained [15]. Karki et. al demonstrated a SPR bio-sensor with dual metal layers of Nickel and Ag between them a franckeite layer is inserted. The highest sensitivity of $352^\circ/\text{RIU}$ is achieved [16]. These researches clearly signify that after addition of 2D materials in the conventional SPR sensor structure the sensitivity tremendously increases.

In the designing of SPR based bio-sensors, commonly used metals are gold (Au), silver (Ag), and copper (Cu). Moreover, the merits of Ag, such as high oxidation susceptibility, its more excellent

SPR ratio, the narrower curve of SPR, make its widespread use in sensor designing due to its upper edge to Au properties. Although the disadvantage that performance parameters like sensitivity, the figure of merit (FoM), detection accuracy (DA) change is limited using Ag alone in the SPR designs can be improved by using 2D material. The commonly used 2D materials in sensing applications are graphene, black phosphorus, and transition metal dichalcogenides (TMDCs). Since 2014, with honeycomb lattice structure BP gaining popularity in SPR sensor designing. The optoelectronic and mechanical features such as high surface to volume ratio (SVR) [18], higher biomolecular adsorption energy than graphene, and molybdenum disulfide (MoS₂) [19], high mobility, low extinction constant [20], direct bandgap. Although BP oxidizes quickly, adding other material resulting in low oxidation as molecular binding is effective inside the sensing layer.

The other material which we are using in our work is zinc sulfide (ZnS). It is a compound semiconductor with exclusive optical properties like a broad energy band gap of 3.72 eV and 3.77 eV and excellent chemical, thermal stability. Its popular applications are in the sensing field, including lasers, gas, light sensors [21]. With the entry of ZnS in photonics, optoelectronics significantly grew over the years and gained popularity [22]. Another optimized layer in which we include our proposed design is silicon dioxide (SiO₂). Its inclusion in our design is due to its unique properties like higher surface area (700 and 500 m^2/g), good chemical and thermal stability, larger bio compatibility, and non-toxic makes it useful for adsorption, in bio sensing catalysis [17], [23], [24].

Section 2 explains the constructional part of our design and various mathematical expressions used to model the SPR sensor. Section 3 gives the results and discussions for the proposed sensor. Section 4 consists of the conclusion of the proposed work.

2. Designing and modeling of sensor

A. Prism and metal layer refractive layer indices

The proposed design based on modified Kretschmann configuration consists of a coupling prism (BK₇ type) with layers of Ag metal, ZnS, BP, and sensing medium (analyte) to produce required surface plasmons (SPs), as shown in Figure 1. These SPs get generated when a P-polarized wave (TM mode) is incident on the coupling prism, generating an evanescent wave along the interface section of prism and metal and penetrates the structure. At resonance angle (θ_{res}) the SPs generates

at the next interface of metal (Ag) and ZnS. It is used as a fundamental recognition element with some exclusive optical properties of black phosphorus (2D material). The refractive index of the sensing medium changes as BP adsorbs biomolecules on its surface.

The refractive index μ_{BK7} expressed by the numerical formula [25]:

$$\mu_{BK7}^2(\lambda) = \frac{1.03961212\lambda^2}{\lambda^2 - 0.00600069867} + \frac{0.231792344\lambda^2}{\lambda^2 - 0.0200179144} + \frac{1.01046945\lambda^2}{\lambda^2 - 103.560653} + 1 \quad (1.1)$$

Here λ is the wavelength of incident light. The He-Ne source is used as a source, and the wavelength is 633 nm.

The metallic layer's refractive index calculation is given by Drude-Lorentz model [26]:

$$\mu_m = 1 - \frac{\lambda^2 \lambda_c}{\lambda_p^2 (\lambda_c + i\lambda)} \quad (1.2)$$

Figure1. Constructional view of ZnS and SiO₂ based SPR sensor

Where, λ_c and λ_p is the collision and plasma wavelengths, having values $1.7614 \times 10^{-5} \text{m}$ and $1.4541 \times 10^{-7} \text{m}$ respectively for Ag (metal) layer. The other layers which we are using in our proposed SPR sensor design, ZnS, have a refractive index of $2.35521 + i * 0.00523$ [27], SiO₂ has 1.4570, and of BP layer is $3.531 - i * 0.04087$ [11]. The refractive index for sensing layer is computed numerically as:

$$\mu_s = 1.33 + \delta\mu \quad (1.3)$$

Here $\delta\mu$ = change in sensing media refractive index.

B. Modeling theory

The performance analysis of the SPR biosensor is done by employing a reliable Transfer matrix method (TMM) and Fresnel equations. The TMM used here does not include any approximation. The multilayers used here have a thickness and dielectric dimension of d_r and ϵ_k , respectively.

The different thicknesses of layers mounted on prism are, for Ag, ZnS, SiO₂, and BP layer as 45 nm, 0.8 nm, 50 nm, and 0.34 nm, respectively. The matrix formation for tangent component for starting and ending boundary given by [28]:

$$\begin{pmatrix} P \\ Q \end{pmatrix} = T \begin{pmatrix} P_{n-1} \\ Q_{n-1} \end{pmatrix} \quad (1.4)$$

Here, at the first boundary, P and Q are electric, and magnetic field components are P_{n-1} and Q_{n-1} are electric and magnetic field components at the last boundary. T indicates characteristics transfer matrix, its value given by [29]:

$$T = \prod_{k=2}^{n-1} T_r, T_r = \begin{bmatrix} T_{11} & T_{12} \\ T_{21} & T_{22} \end{bmatrix} = \begin{bmatrix} \cos\beta_r & -i\sin\beta_r/q_r \\ -iq_r\sin\beta_r & \cos\beta_r \end{bmatrix} \quad (1.5)$$

Here, T_r = r th layer matrix

Here β_r, q_r are the optical admittance and phase factor respectively given by,

$$\beta_r = d_r Y_0 \sqrt{(\epsilon_r - n^2 \sin^2 \theta)} \quad (1.6)$$

$$\text{and phase factor, } q_r = \sqrt{(\epsilon_r - n^2 \sin^2 \theta)} / \epsilon_r \quad (1.7)$$

here, θ = incidence angle, and Y_0 = free space wave number.

The mathematical expression for the reflection coefficient r_p is given by [30]:

$$r_p = \frac{|(C_{11} + C_{12}q_n)q_p - (C_{21} + C_{22}q_n)|^2}{|(C_{11} + C_{12}q_n)q_p + (C_{21} + C_{22}q_n)|^2} = R_p^{1/2} \quad (1.8)$$

Here, R_p = reflectivity of input polarized radiation.

The tabulation chart characterizing different SPR parameters is summarized in table 1. The characteristic parameters that define the SPR biosensor reliability are Sensitivity (S), which defines

the ratio of SPR angle change with the RI change. The second is the Figure of merit (FOM) which gives information about the quality of the bio-sensor. The third one is detection accuracy, which gives information about the sensor's accuracy, and lastly, the Full width half maximum (FWHM) talks about resonance curve width at 50% reflectivity.

Table 1: SPR characterization defined by parameters

Parameters	Formulas	Unit
Sensitivity(S)	$S = \delta\theta_s / \delta\theta$	Degree(RIU) ⁻¹
Figure of Merit (F.O.M)	$F = S * DA$	RIU ⁻¹
Detection Accuracy (D.A.)	$DA = (FWHM)^{-1}$,	(Degree) ⁻¹
Full-width half maximum (F.W.H.M)	$FWHM = \theta_b - \theta_a$,	Degree

3. Results:

The prism coupler having greater RI gives sharp resonance curves in contrast to lower values coupler prism. But the BK7 prism with lower RI for SPR sensor gives better sensitivity [31]. The coming figure 4 gives an idea about the outcomes of inserting Zinc sulfide and Black phosphorus films over the conventional biosensor with RI at 1.33 and 1.335. A silicon dioxide (SiO₂) layer is also taken with a constant thickness of 50 nm after optimization in the proposed work. Figure 2(a) shows the plot for reflectivity with different incidence angles from (55 to 90°) for conventional SPR sensor ($Z = 0, B = 0$) with basic Kretschmann configuration in figure giving sensitivity, $S = 103.6^\circ/\text{RIU}$, and change in incidence angle, $\Delta\theta = 0.518$. The sensitivity and reflectivity are the basic sensors analysis parameters which can be varied by placing layers (2 D materials) between prism and sensing layer. So, after adding the ZnS layer ($Z = 1, B = 0$) over the Ag metal layer, and SiO₂ layer also added in the sensor configuration, get sensitivity as $S = 102.6^\circ/\text{RIU}$ and $\Delta\theta = 0.513$, reducing reflectivity (shown in Fig. 2 (b)). The third case, with a single BP layer ($Z = 0, B = 1$) along with SiO₂ layer and no ZnS layer between metal and sensing layer, is shown with figure 2(c), the parameter, sensitivity, comes out to be $S = 103^\circ/\text{RIU}$ with $\Delta\theta = 0.515$.

At last, figure 2(d) gives the impact of single ZnS and single BP layer ($Z = 1, B = 0$) on the conventional sensor, increasing sensitivity values up to $107^\circ/\text{RIU}$ and lowering the reflectivity value of reflectivity. This happens due to the absorption of the incident light at the BP and sensing layer because surface plasmons are generated. With the continuation to this, the next set of figures

shown by figure 3 gives an idea about the layer's combinations (ZnS and BP) impact on the performance of SPR sensor at variable RI varying from (1.33 to 1.36) with the change of 0.005.

(a): Z, B = 0 at RI = 1.33,1.335

(b): Z = 1, B = 0 at RI = 1.33,1.335

(c): Z = 0, B = 1 at RI = 1.33,1.335

(d): Z, B = 1 at RI = 1.33,1.335

Figure 2(a to d): Representing impact of incidence angle on reflectivity at RI=1.33 and 1.335

Figure 3: Sensitivity variation for refractive index (sensing layer)

Figure 3 shows sensitivity alteration with sensing media RI (varies from 1.33-1.36). For different zinc sulfide (Z) and black phosphorus (B) layers, the minimum sensitivity and maximum sensitivity are given in table 2.

Table 2: Maximum and minimum sensitivity for different cases of zinc sulfide and black phosphorus

Z/B layer case	Minimum Sensitivity (deg/RIU)	Maximum Sensitivity (in deg/RIU)
Z = 0, B = 0	103.4	118
Z = 0, B = 1	102.6	121.2
Z = 1, B = 0	103	126
Z = 1, B = 1	107	129.2

The analysis made here clearly concludes that with modifications on the conventional sensor ($Z = B = 0$), the sensitivity increases by inserting zinc sulfide and black phosphorus layers. The sensitivity value is maximum for the case of $Z = 1$ and $B = 1$, i.e., $S = 129.2^\circ/\text{RIU}$ at $\text{RI} = 1.36$.

The sensitivity and reflectivity values are being calculated with $Z = 0, B = 0$ as $118^\circ/\text{RIU}$ (maximum), and 0.05676 (minimum) for $\text{RI} = 1.36$, shown in Fig. 4(a). Further, by adding a single layer of black phosphorus ($\text{BP} = 1$) without any zinc sulfide ($Z = 0$) layer [figure 4(b)], the sensitivity and reflectivity come out to be $121.2^\circ/\text{RIU}$ (maximum) and 0.05558 (minimum) respectively at $\text{RI} = 1.36$. The following case is shown by figure 4(c), giving maximum sensitivity as $126^\circ/\text{RIU}$ and minimum reflectivity as 0.04745 at $\text{RI} = 1.36$ for single zinc sulfide and no

black phosphorus layer (i.e., $Z = 1, B = 0$). Figure 3(d) gives the best values at $RI = 1.36$, maximum sensitivity as $129.2^\circ/RIU$, and minimum reflectivity as 0.04501 for one zinc sulfide and one black phosphorus layer (i.e., $Z = B = 1$).

Figure 5(a): Detection accuracy and sensitivity plot with combinations of layers (for Z and B)
 (b): FWHM and QF plot with combinations of layers (for Z and B)

Figure 5(a) shows the variation in sensitivity (S) and detection accuracy (DA) for combination layers for different zinc sulfide (Z) and black phosphorus (B). The interchange of both parameters is visible from the multilayer plot in figure 5(a). The sensitivity and DA of the sensor should be high for the better performance of the sensor. The DA is low here, and it needs to be improved. The other two performance parameters, Full width half maximum (FWHM) and quality factor (QF) alteration with the same combination of layers (Z/B), as shown in figure 5(b). The parameter FWHM should be low so that the resonance angle can be computed precisely. The high value of the quality factor is desirable.

The subsequent results for which figure 6 is drawn show the sensitivity range for different refractive indices varying from 1.33 to 1.36. The sensitivity varies from 277.8°/RIU to 664.6°RIU. For RI 1.36, the SPR sensor shows maximum sensitivity.

Figure 6: Sensitivity versus RI (sensing layer) plot

Figure 7 shows a plot for reflectivity alteration with incidence angle variation in zinc sulfide and black phosphorus layers, respectively. The optimum thickness value taken for our SPR design is 45nm for the silver (Ag) layer; for Silicon dioxide (SiO_2), it is 50 nm. In figure 7 (a) gives the analysis for performing parameters after changing the zinc sulfide layer ($Z = \text{variable}$) with constant black phosphorus layer set as $B = 1$. The sensitivity comes to about $664.6^\circ/\text{RIU}$.

The significance of the statements also boosts up by seeing the increase in reflectivity and SPR curve broadness. With the following figure 7(b), using single-layer zinc sulfide and variable black phosphorus layer (i.e., $Z = 1, B = \text{variable}$), we got the maximum sensitivity for our proposed SPR sensor design as high as $266^\circ/\text{RIU}$. The resonance curve's shift and reflectivity's decrement are seen as we increase the black phosphorus layer.

The distribution of the electric field enhancement with respect to the distance, which is orthogonal

(a) SPR curves for the case of $B = 1$, Z = varies

(b) SPR curves for the case of $Z = 1$, B = varies

Figure 7 (a & b): Graphs showing the impact of layer variation on sensor's performance to the prism interface for different numbers of black phosphorus layers, is shown in Fig. 8.

Fig. 8: The field component of P-polarized optical signal at resonance condition with ($N_s = 1.33$):

(a) x-components of an electric field; (b) y-components of electric field

Fig. 8 (a), (b) shows the X-component of the electric field and Y-component of the electric field at the $RI = 1.33$ of the sensing layer. The electric field distribution inside the sensor is shown in figure 9. The electric field intensity is high at the metal and zinc sulfide interface, and intensity decreases at zinc sulfide and SiO_2 . It indicated that field intensity is more when light absorption is more significant. Further electric field intensity increases at the black phosphorus and sensing layer interface. It shows that greater absorption of light to have significant SPW excitation. A proposed electric field plot has been designed for a single layer of ZnS and BP.

Fig. 9: Electric field distribution at resonance angle of SP wave in the proposed sensor

A tabular comparative analysis of this work with earlier literature is being compared here with the help of fundamental parameters with operating wavelength and design configuration of table 3.

Table 3: Comparative analysis of our proposed work with existed researches

Ref.	Configuration	Sensitivity (Deg/RIU)	FWHM (Deg)	DA	Q
This work	BK ₇ /Ag/ZnS/SiO ₂ /BP	664.6	3.323	0.3009	200
[10]	MgF ₂ /Rh/Ag/Si/graphene	220	10.204	0.0980	21.56
[32]	SF10/ZnO/Au/MoS ₂ /graphene	101.58	6.723	0.1487	15.11
[33]	BK ₇ /Ag/BP/WSe ₂	279	-	-	-
[34]	BK ₇ /Au ₁ /MoS ₂ /Au ₂ /graphene	182	9.475	-	19.21
[35]	BK ₇ /Ag/Tin selenide (SnSe)	178	-	-	-
[36]	SF10/Ag/BP/graphene/affinity/PBS solution	91.54	-	3.61	54.81

Conclusion

We proposed a new SPR optical biosensor in this study that has a high sensitivity. The suggested biosensor's configuration includes a ZnS layer to improve sensitivity and protect the nanocomposite layer from oxidation. Furthermore, our findings demonstrate that using black phosphorus as a biological diagnosis component can be helpful for sensing. We looked examined how different types of prisms affected the sensitivity of this proposed SPR optical biosensor. The results reveal that when the refractive index of the prism increases, the sensitivity of the suggested

SPR biosensor diminishes. As a result, we chose BK7 as the coupling prism in our biosensor design. Our calculations also suggest that the proposed biosensor with the maximum sensitivity requires a SiO₂ layer with a thickness of 50 nm that contains a silver metal layer with a thickness of 45 nm. Finally, we demonstrated that the best-proposed biosensor has six layers of ZnS on both surfaces of the nanocomposite layer, with a maximum sensitivity of 664.6°/RIU.

Code Availability: Not applicable.

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Availability of data and material: No data available.

Authors' contributions

BK formulated the problem statement wherein giving the theoretical background and mathematical modelling for SPR biosensor. He also helped in drafting and finalizing the manuscript.

YT provided the theoretical background to biosensing and the importance of Optical Biosensing. He also helped in finalizing the design of the proposed sensor.

AU worked towards the complete manuscript, formatting, and finalizing the manuscript.

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

Ethics approval

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

Consent to participate

I am willing to participate in the work presented in this manuscript.

Consent for publication

The author has given their consent to publish this work.

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