

# Sensitivity Enhancement of Surface Plasmon Resonance-based Biosensor using Aluminium-Cobalt-Tungsten Disulfide-Graphene Heterostructure

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## ABSTRACT

An attempt has been made to enhance the sensitivity of a high-sensitive surface plasmon resonance (SPR) biosensor with an aluminium-cobalt bimetallic layer covered by a tungsten disulfide-graphene heterostructure. A thin layer of cobalt coated on an aluminium layer contributed substantially to increase the sensor performance. The use of Al and Co metals instead of noble metals like Ag and Au reduced the cost of the sensor. Further, tungsten disulfide (WS<sub>2</sub>) layers were employed to improve sensitivity and protect the bimetal Al-Co from becoming oxidized, whereas graphene served as the biomolecule trapping medium. The number of WS<sub>2</sub> and graphene layers have optimized for better sensitivity. The proposed biosensor Al-Co-WS<sub>2</sub>-graphene structure displayed an excellent sensitivity of  $300^{\circ}$ /RIU, convenient for sensing biomolecules.

Keywords: Surface Plasmon Resonance; Biosensor; Sensitivity; Cobalt; 2D materials.

#### 1. INTRODUCTION

Surface plasmon resonance (SPR) biosensor is a sensing technological marvel, owing to its fast detection, high accuracy, stable performance and high sensitivity, with several applications in many sectors including DNA hybridization (Rahman et al. 2017), drug discovery (Rich et al. 2002), environmental monitoring (Mauriz et al. 2007), medical diagnostics (Yonzon et al. 2004), food safety (Zhou et al. 2019), enzyme detection (Pumera, 2011), plasmonic detectors (Ooi et al. 2012) and immune sensor (Ramanaviciene et al. 2000). In the SPR biosensors, the angular interrogation-based Kretschmann configuration is customarily used owing to the ability to function without an intermediary between the prism and metal layer with utilization of monochromatic light to gain high signal-to-noise (Kretschmann et al. 1968). The SPR biosensor frequently uses the plasmonic materials gold or silver (Mishra et al. 2015; Maharana et al. 2013). Gold is the most desirable material as it has a good prohibition to oxidation and erosion, is chemically stronger and has excellent optical behavior (Maurya et al. 2015). However, Au produces wider resonance curve that affects the analyte identification accuracy. Moreover, the biomolecular binding potential of Au is weak (Dash et al. 2014). Conversely, SPR sensor utilising Ag layer shows better resolution, but sensitivity is weaker when compared with Au (Wang et al. 2017). However, silver is chemically susceptible because of its oxidized nature (Verma et al. 2015; Klantsataya et al. 2015). Nowadays, bimetallic blends, oxide layers and 2D materials are recommended to solve the Ag oxidation trouble (Dash and Jha, 2014; Sharma et al. 2007; Jha et al. 2009; Fouad et al. 2016; Kravets et al. 2014; Suresh et al. 2021). However, these approaches have a limited range of sensitivity and are not suitable for durable plasmonic applications. Aluminium (Al) is a low-price metal than Au and Ag. Al provides a fine SPR curve, but it oxidizes very easily (Jha et al. 2009). Recently, Maharana et al. suggested that the oxidization issue of Al can be overcome by coating Au layer on Al (Maharana et al. 2013). Maharana also proposed that using a graphene layer over aluminium can improve sensor sensitivity while also protecting Al from oxidation (Maharana et al. 2014). Recently, cobalt (Co), a ferromagnetic metal, has gained attention owing to its notable magneto-optical properties and magnetic properties (Yu et al. 1968; Gilliot et al. 2006; Maheswari et al. 2022). Shukla et al. suggested that the magnetic metal cobalt used in SPR sensors reduces the cost and enhances the sensitivity (Shukla et al. 2016).

Transition metal dichalcogenides, TMDCs, (MoX<sub>2</sub> and WX<sub>2</sub>, namely Molybdenum disulfide (MoS<sub>2</sub>),



Molybdenum diselenide (MoSe<sub>2</sub>), Tungsten disulfide (WS<sub>2</sub>), Tungsten diselenide (WSe<sub>2</sub>) and graphene have recently been recognized as the best 2D materials for improving sensitivity (Verma et al. 2011; Lin et al. 2016; Ouyang et al. 2017; Wu et al. 2017). Graphene possesses large charge carrier mobility (106 cm<sup>2</sup>V<sup>-1</sup>S<sup>-1</sup>) which encourages the strong coupling at the metal-graphene interface that improves the electric field (Mayorga-Martinez et al. 2015). Wu et al. demonstrated that simply coating graphene (10-layer) on Au layer increases sensitivity by 25% when compared to the standard sensor that only uses Au layer (Wu *et al.* 2010). Graphene's  $\pi$ integration is used to particularly capture aromatic molecules. Its hexagonal lattice structure prevents any gas molecule from passing through the biosensor; moreover, biomolecule adsorption is increased thanks to graphene's larger surface area (2630 m<sup>2</sup>/g), resulting in enhanced SPR sensor performance (Atta et al. 2015; Song et al. 2010; Bunch et al. 2008). Likewise, TMDCs have outstanding features such as large surface area, atomically thin nature, flexibility, adequate carrier mobility, chemical steadiness, good biocompatibility and layer based (optical and electronic) properties (Luo et al. 2016; Ambrosi et al. 2015; Ballif et al. 1996; Fortin et al. 1982; O'Brien et al. 2014). Wu et al. revealed that using different 2D materials on Ag layer can increase the sensitivity and can act as a defensive layer that prohibits oxidation of the metal film (Wu et al. 2017). Nisha et al. reported that the SPR sensor contains MoS<sub>2</sub> sandwiched between the metal layers with a covering single layer of graphene can enhance the sensitivity to 229°/RIU (Nisha et al. 2019). Anower et al. suggested that WS<sub>2</sub>-graphene coated on Au leads to improved SPR's sensitivity due to graphene's biomolecule absorption capabilities and WS<sub>2</sub>'s high carrier mobility (Anower et al. 2018). Rahman et al. developed a fibre-based SPR biosensor with a WS<sub>2</sub> layer placed between silver and graphene to enhance the sensitivity (Rahman et al. 2019). Wang et al. reported that covering Ag-Au bimetallic layer with graphene and WS<sub>2</sub> layers improves SPR's sensitivity.  $WS_2$  is the ideal option since it can more efficiently enhance light absorption and graphene can particularly capture aromatic compounds (Wang et al. 2017). Han et al. demonstrated that an SPR biosensor with WS2graphene-based hybrid structures coated on silverindium-tin oxide layers exhibits a maximum sensitivity of 213.2°/RIU (Han et al. 2020).

In this work, we proposed a new configuration composed of Al-Co-WS<sub>2</sub>-graphene with excellent sensitivity. The 2D layers - WS<sub>2</sub> and graphene are used to increase the light absorption capability of SPR sensor, which is more conducive to obtain higher sensitivity.



Fig. 1: Schematic illustration of the proposed Al-Co-WS<sub>2</sub>-Graphene configuration

## **2. THEORETICAL MODEL**

# 2.1 The Present Sensor Structure

The proposed Al-Co-WS<sub>2</sub>-graphene SPR biosensor's schematic structure is shown in Fig. 1. The

biosensor comprises of six layers: BK7 prism, aluminium, cobalt, WS<sub>2</sub>, graphene, and the sensing layer. In this case, p-polarized light with 633 nm wavelength is passed through the prism/metal interface and then the reflected light is sensed and examined by the photodetector. The first layer, BK7 prism, has a

refractive index ( $n_{BK7}$ ) of 1.5151 (Wu *et al.* 2017). The second layer, Al, is placed over the BK7 prism. The third layer, Co, is coated on Al. The Drude model is used to calculate the dielectric constant of metal layers ( $\varepsilon_m$ ), using:

The collision wavelength ( $\lambda_c$ ) and the plasma wavelength ( $\lambda_p$ ) for Al and Co are shown in Table 1 (Maharana *et al.* 2013; Shukla *et al.* 2016). The fourth layer is WS<sub>2</sub> and the fifth layer is graphene whose refractive indices are shown in Table 2 (Wu *et al.* 2017). Lastly, the sensing medium is the sixth layer and its refractive index (n<sub>s</sub>) is 1.33.



Fig. 2. Reflectance vs. Incident angle at  $\delta n_s = 0.005$  for different structures: (a) metal (Al = 30 nm), (b) bimetal (Al = 30 nm, Co = 10 nm), (c) Al = 30 nm, Co = 10 nm and L = 1, (d) Al = 30 nm, Co = 10 nm and M = 1 and (e) Al = 30 nm, Co = 10 nm, M = 1 and L = 1

Tabl	le 1. (	Collision	and p	lasma	wavel	lengti	h of	A	and	Co
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Parameter	Al	Со
$\lambda_{c}(m)$	2.4511 x 10 <sup>-5</sup>	3.3578 x 10 <sup>-5</sup>
$\lambda_{p}(m)$	1.0657 x 10 <sup>-7</sup>	3.1215 x 10 <sup>-7</sup>

Table 2. Thickness and Refractive index of 2D materials at  $\lambda$  = 633 nm

2D materials	Thickness of monolayer (nm)	Refractive index		
$WS_2$	0.80	4.9 + 0.3124i		
Graphene	0.34	3.0 + 1.1491i		

# 2.2 Sensitivity

Sensitivity is the crucial parameter of the SPR sensor that refers to the variation of the resonance angle  $(\delta \theta_{SPR})$  produced by the smallest modification in the refractive index of the sensing medium ( $\delta n_s$ ) and is given by (Lin *et al.* 2016),

$$S_n = \frac{\delta \theta_{SPR}}{\delta n_s} \tag{2}$$

#### **3. RESULTS AND DISCUSSION**

The performance of the suggested SPR biosensor was examined using the transfer matrix method for numerical calculations. In order to improve the sensitivity of the proposed SPR biosensor, the thicknesses of Al (30 nm) and Co (10 nm) layers were

optimized. Moreover, the effects of WS<sub>2</sub> and graphene layers on the sensitivity of the suggested biosensor were investigated. Fig. 2 a has shown the reflectance curve for the sensor with 30 nm aluminium metal layer. It has been observed that when including the biomolecules in the sensing medium, a slight alteration in the refractive index of the sensing medium ( $\delta n_s = 0.005$ ) was attained with a corresponding resonance angle shift of  $\delta\theta = 0.48^{\circ}$  and a sensitivity of  $S_n = 96^{\circ}/RIU$ . Fig. 2 b has shown that when using Al-Co bimetallic layer without WS<sub>2</sub> and graphene layers (M=0 and L=0), the sensor has given a large resonance angle shift ( $\delta\theta = 0.85^\circ$ ) and high sensitivity  $(S_n = 170^{\circ}/RIU)$ . It was clear that when using Co, the sensitivity reaches higher values. In Fig. 2 c, it was noted that the inclusion of a single layer of graphene and lacking of WS<sub>2</sub> (M=0 & L=1), retaining the other parameters, resulted in an increase in resonance angle change to  $\delta\theta = 0.87^{\circ}$  with the sensitivity of  $S_n =$ 174°/RIU. From Fig. 2 d, it was evident that when plating a single layer of WS<sub>2</sub> on Al-Co bimetallic layer, the sensor provided a longer resonance angle shift of  $\delta \theta =$  $1.05^{\circ}$  with a high sensitivity of S<sub>n</sub>=  $210^{\circ}/\text{RIU}$ . This clearly revealed that the WS<sub>2</sub> layer exhibited a sensitivity greater than the graphene layer. Fig. 2 e shows that by using WS<sub>2</sub> and graphene simultaneously, the shift in resonance angle was greatly improved as  $\delta \theta = 1.1^{\circ}$  and the sensitivity was increased to  $S_n = 220^{\circ}/RIU$ . Hence, it was evident that the proposed sensor containing a monolayer of WS<sub>2</sub> and a monolayer of graphene can obtain the greatest sensitivity.



Fig. 3: (a) Sensitivity vs. Refractive index and (b) FWHM vs. Refractive index



Fig. 4: (a) Reflectance vs. Angle of incidence for various numbers of  $WS_2$  layers with 1-layer of graphene (L = 1) at  $n_s = 1.33$  and (b) Reflectance vs. angle of incidence for various numbers of graphene layers with fixed 2-layer of  $WS_2$  (M = 2) at  $n_s = 1.33$ 1.33

Wavelength (nm)	Configuration	Sensitivity (°/RIU)	Reference
633	SF10-Au-Si-WS <sub>2</sub>	155.68	Ouyang et al. 2016
633	BK7-AU-MoS <sub>2</sub> -Au-graphene	182	Lin et al. 2016
633	BK7-air-MoS <sub>2</sub> -Al-MoS <sub>2</sub> -graphene	190.36	Wu et al. 2017
633	BK7-Ag-franckeite-graphene	196	Gan et al. 2019
633	BK7-AU-Si-MoS <sub>2</sub> -Au-graphene	210	Shushama et al. 2017
632.8	BK7-Ag-ITO-WS <sub>2</sub> -graphene	213.2	Han et al. 2020
633	BK7-Au-MoS <sub>2</sub> -Ni-graphene	229	Nisha et al. 2019
633	BK7-Ag-Si- MXene	231	Kumar et al. 2021
633	$CaF_2\text{-}ZnO\text{-}Au\text{-}BlueP/MoS_2$	235	Singh et al. 2021
633	BK7-Ag-BP-WS <sub>2</sub>	237	Wu et al. 2017
633	BK7-Ag-BP-WSe <sub>2</sub>	279	Wu et al. 2017
633	BK7-Al-Co-WS <sub>2</sub> - graphene	300	Proposed

Table 3. Comparison between the proposed sensor and other SPR sensors published previously

Furthermore, to study the sensitivity of the biosensor corresponding to the refractive index (RI) of sensing layer variations in the limits of 1.33 to 1.36, sensitivity vs. RI was plotted in the limits of 1.33 to 1.36, as illustrated in Fig. 4 a. It was observed that for the sensor structure of M=0 and L=0, the sensitivity linearly has increased from 158°/RIU to 221°/RIU. For the sensor consisting of the graphene layer (M=0 and L=1), the sensitivity has linearly improved from 163°/RIU to 234°/RIU. Thus, the graphene enriches the sensitivity and absorbs biomolecules more effectively since it has a greater surface-to-volume ratio and  $\pi$ - $\pi$  stacking property. Further, for the sensor holding a single layer of WS<sub>2</sub> in the absence of graphene (M=1 & L=0), the

sensitivity has linearly improved from 191°/RIU to 331°/RIU. Thus, the WS<sub>2</sub> layer contributes to augment the sensitivity than the graphene layer because of greater light absorption efficiency and higher real part of the dielectric constant. Moreover, it was observed that the sensor formation with a single layer of WS<sub>2</sub> and graphene layers (M=1 and L=1) has exhibited the greatest sensitivity which linearly rises from 199°/RIU to 340°/RIU, corresponding to the variation in the RI of the sensing medium from 1.33 to 1.36. From Fig. 3 a, it was clear that the proposed biosensor simultaneously consisting of a monolayer of WS<sub>2</sub> and graphene layers greatly rise the sensitivity when compared with the customary SPR sensor. Similarly, FWHM vs. RI of the

sensing medium was plotted in Fig. 3 b, with other factors unchanged. It was noted from the SPR curve that FWHM improved from  $2.38^{\circ}$  to  $3.42^{\circ}$  (M=0 and L=0),  $3.11^{\circ}$  to  $4.55^{\circ}$  (M=0 and L=1),  $4.12^{\circ}$  to  $6.26^{\circ}$  (M=1 and L=0) and  $5.28^{\circ}$  to  $7.41^{\circ}$  (M=1 and L=1). That the suggested design comprising of Al-Co-WS<sub>2</sub>-graphene hybrid structure has

FWHM as small as a traditional SPR sensor with pure gold. Hence, the proposed biosensor consisting of Al (30 nm), Co (10 nm), WS<sub>2</sub> (M=2) and graphene (L=1) will have extraordinary sensitivity of  $300^{\circ}/\text{RIU}$  and high resolution, as evident from Table 3.



Fig. 5 (a) Sensitivity vs. number of WS<sub>2</sub> layers under the condition of monolayer graphene (L = 1), b) Sensitivity vs. number of graphene layers with optimized 2-layer WS2 (M = 2) and c) Reflectance spectra for Al (30 nm)-Co (10 nm)-2M-1L based proposed structure

Furthermore, the sensor performance for various numbers of WS<sub>2</sub> and graphene layers added on Al (30 nm)-Co (10 nm) bimetallic layer were examined (Fig. 4 a and b). It was observed that the sensitivity increased with an increasing number of WS<sub>2</sub> and graphene layers. It was found that the resonance angle change for WS<sub>2</sub> layers was better than the graphene layer because the real part of dielectric constant is high for WS<sub>2</sub>. Moreover, WS<sub>2</sub> possesses a high imaginary part of dielectric constant, which causes more electron energy loss. Because of this, the SPR curve width increases as the number of WS<sub>2</sub> layers increases compared to graphene. When the resonance angle, providing

low accuracy. A random placement of  $WS_2$  and graphene layers has an effect on the sensor's performance. Therefore, it is necessary to optimize the number of  $WS_2$  and graphene layers.

Fig. 5 a illustrates that the sensitivity was a function of the increasing WS<sub>2</sub> layer and a single layer of graphene (L=1), while the other factors were kept constant. It was found that the sensitivity increased from a single layer of WS<sub>2</sub> (M=1) to two layers of WS<sub>2</sub> (M=2); with further increase in WS<sub>2</sub> layers, the sensitivity decreased because of the decrease in light consumption rate. As a result, the two-layer configuration of WS<sub>2</sub> (M=2) was optimized, which resulted in the highest

sensitivity of  $300^{\circ}$ /RIU. Then, the graphene layer was optimized with the two-layer configuration of WS<sub>2</sub>. From Fig. 5 b, it is obvious that a single layer of graphene resulted in the maximum sensitivity.

It can be concluded that the proposed biosensor comprising of Al (30 nm), Co (10 nm), WS<sub>2</sub> (M=2) and graphene (L=1) has an extraordinary sensitivity of  $288^{\circ}$ /RIU, highly beneficial for biological applications and medical diagnosis.

# **4. CONCLUSIONS**

In this study, an attempt has been made to enhance the sensitivity of a SPR biosensor with an aluminium-cobalt bimetallic layer covered by a tungsten disulfide-graphene heterostructure. It is noticeable that the bimetallic blend of Al-Co raises the sensitivity of the suggested sensor. Moreover, the inclusion of WS<sub>2</sub> and graphene layers on the Al-Co bimetallic layer was analyzed to further enhance the sensitivity. The 2D materials (WS<sub>2</sub> and graphene) were used not only to increase the sensitivity, but also to protect the bimetallic layers of Al-Co. The number of WS<sub>2</sub> and graphene layers were optimized for better results. The proposed Al-Co-WS<sub>2</sub>-graphene model exhibits the highest sensitivity of 300°/RIU, with 2 layers of WS2 and a monolayer of graphene. Thus the proposed structure of the SPR sensor has the promise of a new and effective approach to biosensing.

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## **CONFLICTS OF INTEREST**

The authors declare that there is no conflict of interest.

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