Research Article

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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_2) 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_2 and graphene layers have optimized for better sensitivity. The proposed biosensor Al-Co-WS_2-graphene structure displayed an excellent sensitivity of 300°/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_2 and WX_2, namely Molybdenum disulfide (MoS_2), Molybdenum diselenide ($MoSe_2$), Tungsten disulfide (WS_2), Tungsten diselenide (WSe_2) 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 $cm^2V^{-1}S^{-1}$) 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 π 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^2/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_2 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_2-graphene coated on Au leads to improved SPR's sensitivity due to graphene's biomolecule absorption capabilities and WS_2's high carrier mobility (Anower *et al.* 2018). Rahman *et al.* developed a fibre-based SPR biosensor with a WS_2 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_2 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 WS2-graphene-based hybrid structures coated on silver-indium-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_2-graphene with excellent sensitivity. The 2D layers - WS_2 and graphene are used to increase the light absorption capability of SPR sensor, which is more conducive to obtain higher sensitivity.

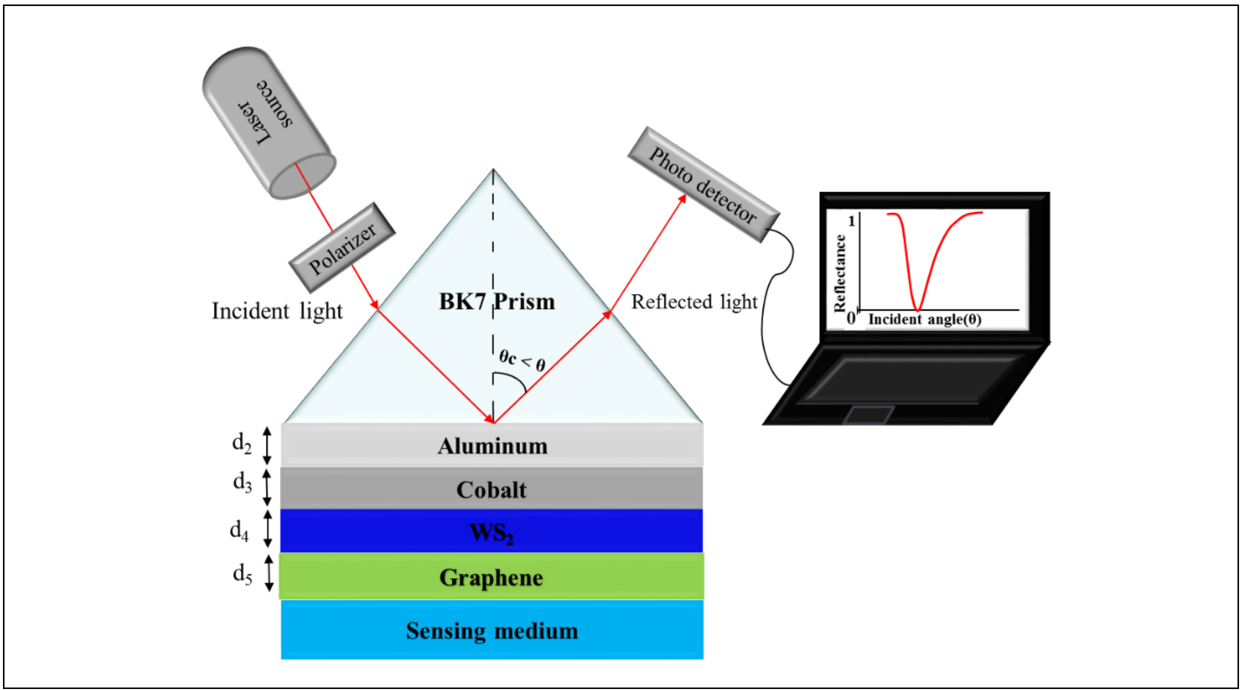

Fig. 1: Schematic illustration of the proposed Al-Co-WS_2-Graphene configuration

2. THEORETICAL MODEL

2.1 The Present Sensor Structure

The proposed Al-Co-WS_2-graphene SPR biosensor's schematic structure is shown in Fig. 1. The biosensor comprises of six layers: BK7 prism, aluminium, cobalt, WS_2, 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 (ε_m), using:

$$\varepsilon_m(\lambda) = \varepsilon_{mr} + \varepsilon_{mi} = 1 - \frac{\lambda^2 \lambda_c}{\lambda_p^2(\lambda_c + i\lambda)} \quad \text{......... (1)}$$

The collision wavelength (λ_c) and the plasma wavelength (λ_p) for Al and Co are shown in Table 1 (Maharana *et al.* 2013; Shukla *et al.* 2016). The fourth layer is WS_2 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_s) is 1.33.

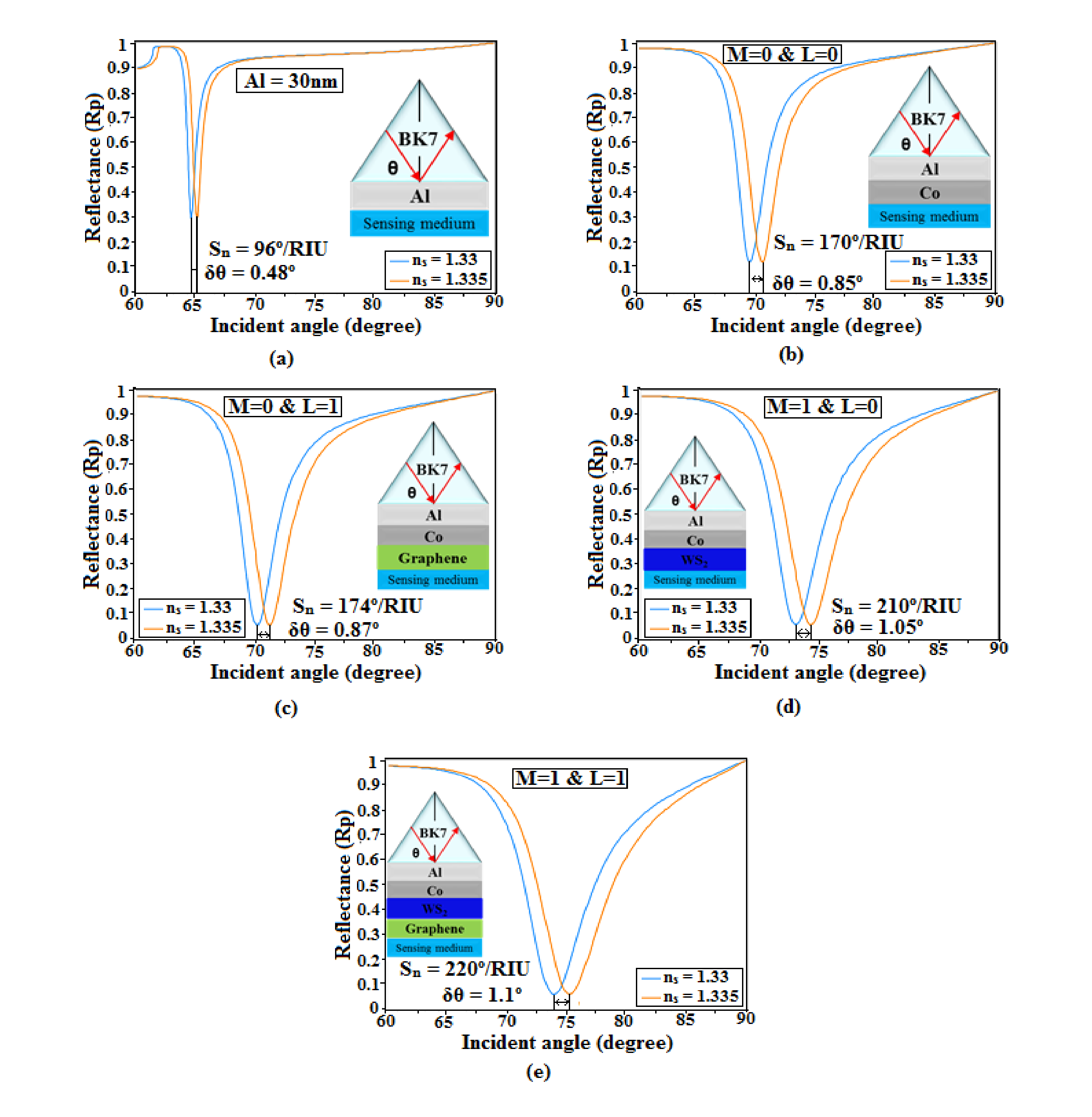

Fig. 2. Reflectance vs. Incident angle at δn_s = 0.005 for different structures: (a) metal (Al = 30 nm), (b) bimetal (Al = 30 nm, Co = 10 nm), (c) Al = 30 nm, Co = 10 nm and L = 1, (d) Al = 30 nm, Co = 10 nm and M = 1 and (e) Al = 30 nm, Co = 10 nm, M = 1 and L = 1

Table 1. Collision and plasma wavelength of Al and Co

Parameter	Al	Co
λ_c (m)	2.4511 x 10^{-5}	3.3578 x 10^{-5}
λ_p (m)	1.0657 x 10^{-7}	3.1215 x 10^{-7}

Table 2. Thickness and Refractive index of 2D materials at λ = 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 (δn_s) and is given by (Lin *et al.* 2016),

$$S_n = \frac{\delta\theta_{SPR}}{\delta n_s} \qquad \text{……………...........} \qquad (2)$$

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_2 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 (δn_s = 0.005) was attained with a corresponding resonance angle shift of $\delta\theta$ = 0.48° and a sensitivity of S_n = 96°/RIU. Fig. 2 b has shown that when using Al-Co bimetallic layer without WS_2 and graphene layers (M=0 and L=0), the sensor has given a large resonance angle shift ($\delta\theta$ = 0.85°) and high sensitivity (S_n= 170°/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_2 (M=0 & L=1), retaining the other parameters, resulted in an increase in resonance angle change to $\delta\theta$ = 0.87° with the sensitivity of S_n = 174°/RIU. From Fig. 2 d, it was evident that when plating a single layer of WS_2 on Al-Co bimetallic layer, the sensor provided a longer resonance angle shift of $\delta\theta$ = 1.05° with a high sensitivity of S_n= 210°/RIU. This clearly revealed that the WS_2 layer exhibited a sensitivity greater than the graphene layer. Fig. 2 e shows that by using WS_2 and graphene simultaneously, the shift in resonance angle was greatly improved as $\delta\theta$ = 1.1° and the sensitivity was increased to S_n = 220°/RIU. Hence, it was evident that the proposed sensor containing a monolayer of WS_2 and a monolayer of graphene can obtain the greatest sensitivity.

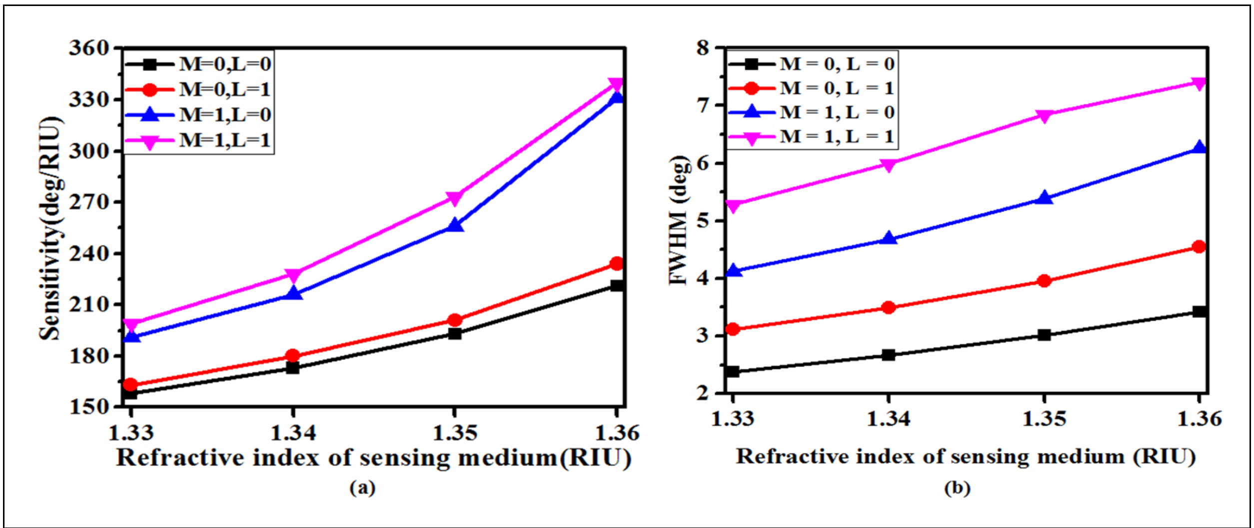

Fig. 3: (a) Sensitivity vs. Refractive index and (b) FWHM vs. Refractive index

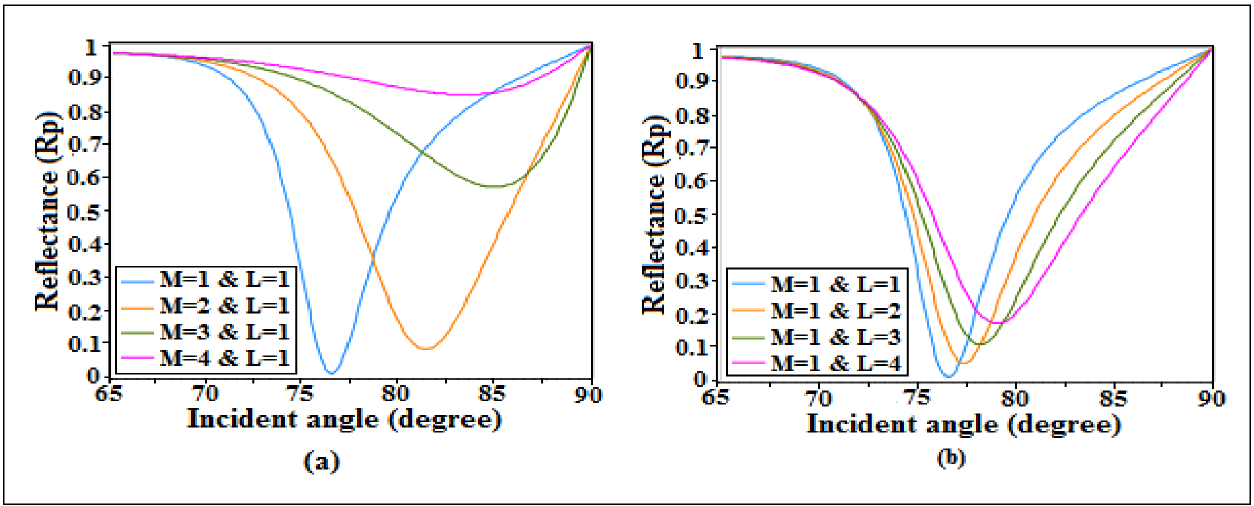

Fig. 4: (a) Reflectance vs. Angle of incidence for various numbers of WS_2 layers with 1-layer of graphene (L = 1) at n_s = 1.33 and (b) Reflectance vs. angle of incidence for various numbers of graphene layers with fixed 2-layer of WS_2 (M = 2) at n_s = 1.33

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

Wavelength (nm)	Configuration	Sensitivity (°/RIU)	Reference
633	SF10-Au-Si-WS_2	155.68	Ouyang *et al.* 2016
633	BK7-AU-MoS_2-Au-graphene	182	Lin *et al.* 2016
633	BK7-air-MoS_2-Al-MoS_2-graphene	190.36	Wu *et al.* 2017
633	BK7-Ag-franckeite-graphene	196	Gan *et al.* 2019
633	BK7-AU-Si-MoS_2-Au-graphene	210	Shushama *et al.* 2017
632.8	BK7-Ag-ITO-WS_2-graphene	213.2	Han *et al.* 2020
633	BK7-Au-MoS_2-Ni-graphene	229	Nisha *et al.* 2019
633	BK7-Ag-Si- MXene	231	Kumar *et al.* 2021
633	CaF_2-ZnO-Au-BlueP/MoS_2	235	Singh *et al.* 2021
633	BK7-Ag-BP-WS_2	237	Wu *et al.* 2017
633	BK7-Ag-BP-WSe_2	279	Wu *et al.* 2017
633	BK7-Al-Co-WS_2- graphene	300	Proposed

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 π-π stacking property. Further, for the sensor holding a single layer of WS_2 in the absence of graphene (M=1 & L=0), the sensitivity has linearly improved from 191°/RIU to 331°/RIU. Thus, the WS_2 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_2 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_2 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° to 3.42° (M=0 and L=0), 3.11° to 4.55° (M=0 and L=1), 4.12° to 6.26° (M=1 and L=0) and 5.28° to 7.41° (M=1 and L=1). That the suggested design comprising of Al-Co-WS_2-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_2 (M=2) and graphene (L=1) will have extraordinary sensitivity of 300°/RIU and high resolution, as evident from Table 3.

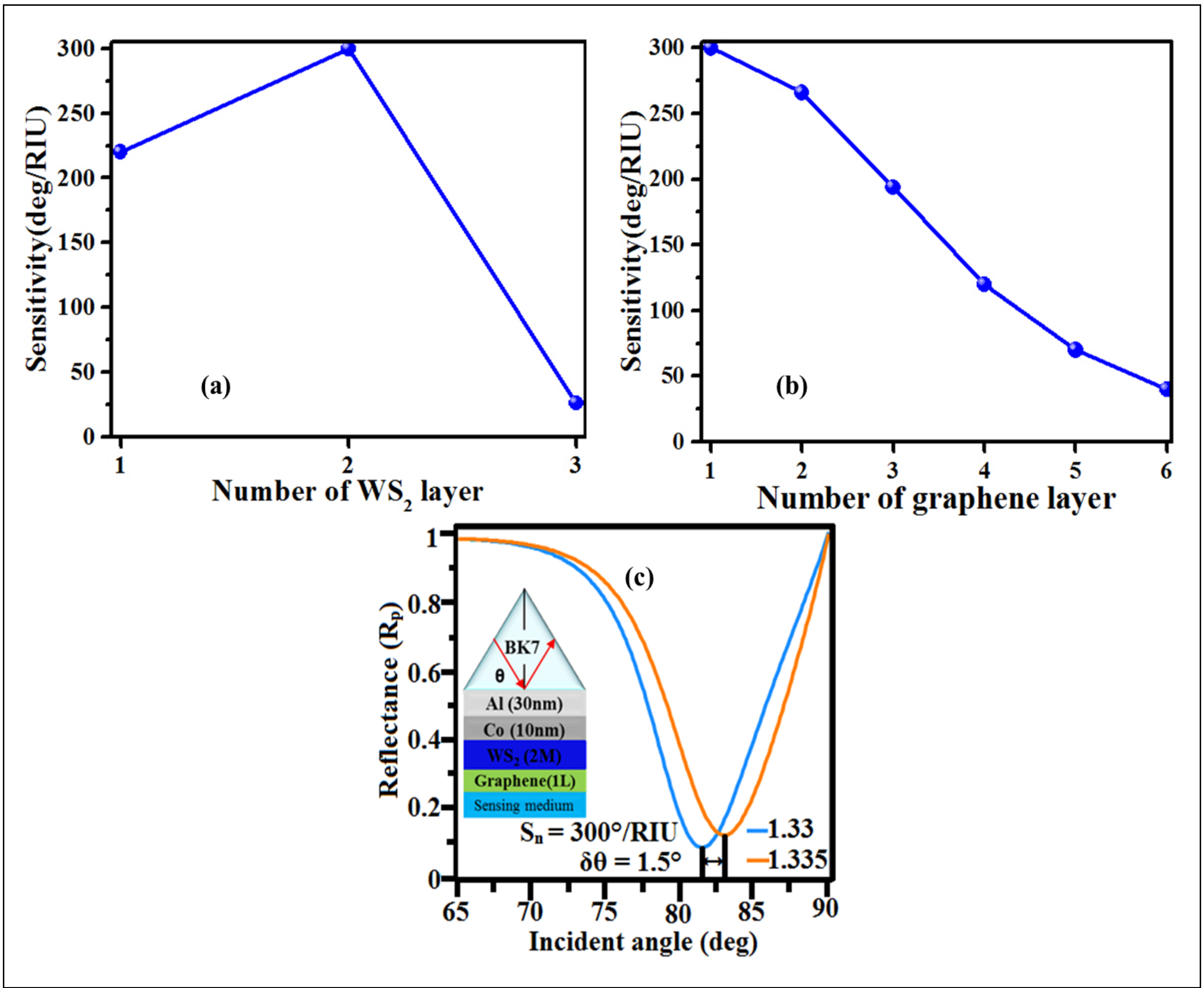

Fig. 5 (a) Sensitivity vs. number of WS_2 layers under the condition of monolayer graphene (L = 1), b) Sensitivity vs. number of graphene layers with optimized 2-layer WS2 (M = 2) and c) Reflectance spectra for Al (30 nm)-Co (10 nm)-2M-1L based proposed structure

Furthermore, the sensor performance for various numbers of WS_2 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_2 and graphene layers. It was found that the resonance angle change for WS_2 layers was better than the graphene layer because the real part of dielectric constant is high for WS_2. Moreover, WS_2 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_2 layers increases compared to graphene. When the resonance curve widens, it becomes challenging to identify 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_2 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_2 (M=1) to two layers of WS_2 (M=2); with further increase in WS_2 layers, the sensitivity decreased because of the decrease in light consumption rate. As a result, the two-layer configuration of WS_2 (M=2) was optimized, which resulted in the highest sensitivity of 300°/RIU. Then, the graphene layer was optimized with the two-layer configuration of WS_2. 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_2 (M=2) and graphene (L=1) has an extraordinary sensitivity of 288°/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_2 and graphene layers on the Al-Co bimetallic layer was analyzed to further enhance the sensitivity. The 2D materials (WS_2 and graphene) were used not only to increase the sensitivity, but also to protect the bimetallic layers of Al-Co. The number of WS_2 and graphene layers were optimized for better results. The proposed Al-Co-WS_2-graphene model exhibits the highest sensitivity of 300°/RIU, with 2 layers of WS_2 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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