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# Hybrid Heterostructures for SPR Biosensor

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## Abstract

Surface plasmon resonance (SPR) based biosensors have been enormously studied in the last decade for their better sensitivity. In recent years hybrid heterostructures are getting popularity to implement these SPR biosensors for their superior sensing capability. This chapter demonstrates the details of SPR technology with two recently studied prism-based hybrid heterostructures. These heterostructures are made up of conventional SPR biosensors with two additional layers of recently invented transition metal dichalcogenides, platinum di-selenide (PtSe<sub>2</sub>), and highly sensitive 2D material, tungsten di-sulfide (WS<sub>2</sub>). Angular interrogation method is discussed to investigate the sensing capabilities of the sensors which prove the superiority of the Ag-PtSe<sub>2</sub>-WS<sub>2</sub> structure. The sensing capability of this structure has been found at least 1.67 times higher than that of the conventional non-hybrid structures, respectively, with comparable FOM and QF. A comparison table has been provided at the end of this chapter which also shows the impressive performance of the hybrid heterostructures for SPR biosensors. Proper demonstration with a suitable example of this chapter will emphasize the potential use of hybrid heterostructure based SPR biosensors in prospective medical diagnostics and biomedical detection applications.

**Keywords:** biosensor, hybrid heterostructure, sensitivity, surface plasmon resonance

## 1. Introduction

Surface plasmon resonance (SPR) biosensors have become one of the most promising, standard, and affordable technology due to prompt research and expansion of SPR phenomenon in the last two decades. Nowadays, SPR sensors are broadly implemented for numerous biological and biochemical analytes identification and characterization due to its high sensitivity, real-time monitoring, level free detection assay, small sample size, and reusable sensor chip [1–5]. To be detailed, the SPR biosensors are adopted to agriculture and food quality monitoring [6], security and safety analysis [7], in need of medical diagnostics, environmental monitoring, bio-imaging [8–10], cancer detection [11, 12], DNA hybridization [13, 14], enzyme detection [15], protein-protein, protein-DNA, and protein-virus hybridization [16, 17], microorganisms identifying [18], industrial appliance's condition monitoring, temperature monitoring [19], gas sensing [20, 21], chemical and biochemical analysis [22, 23], pharmaceutical and biological molecule analysis [24, 25], oil condition monitoring [26], and so on. In the year 1902, Wood [27] first

observed unexpected optical power attenuation characteristic at the time of measuring the reflection of light from metallic gratings. This phenomenon occurs due to absorbance and conversion of photon energy to surface plasma wave (SPW) which is the result of combined oscillation of excited electrons called surface plasmon polaritons (SPPs). This oscillating electron consumes maximum energy at a certain wavelength for a specific angle of incidence of light which is called resonance condition. That is why this phenomenon is named surface plasmon resonance (SPR). In 1968, Otto [28] and Kretschmann [29] introduced attenuated total internal reflection (ATR), which encouraged scientists and researchers to concentrate on the implementation of SPR sensing technology practically. In 1982, the SPR sensing technique was first demonstrated by Nylander and Liedberg [4, 30] for the practical application of gas sensing. After that, SPR sensing technology has been getting ceaselessly developing consideration from the scientific and academic network. In 1990, the SPR sensing instrument was first commercially produced and introduced to the market by Biacore AB. Since then a considerable number of manufacturers e.g. IBIS Technologies B.V., Graffinity pharmaceuticals, GWC Technologies, Bio-Red, AutoLab, Farfield Sensors, Genoptics Bio Interactions, Microvacuum, Biosensing Instrument, and SPR Navi have launched their SPR instruments to the market [17, 31].

Different optical techniques are currently proposed for sensing purposes, including Raman scattering based sensors [32, 33], grating coupled sensors [34, 35], prism coupled sensors [36, 37], optical fiber-based sensors [38, 39], planar waveguide-based sensors [40, 41] etc. The optical biosensors basically work with the measurement of change in input incident light and detected light at the output terminal. To be specific, the change in phase, amplitude, wavelength, frequency, or polarization of light is measured at the output terminal of the sensors and the changes in these parameters are observed. Among them, the commonly used technique is observing the reflected light angle where maximum light is attenuated. This method is called angular interrogation approach with attenuated total internal reflection (ATR) that is applied usually in prism coupled devices. The performance of an optical sensor is basically measured in terms of its sensitivity, detection accuracy or detection limit, the figure of merits (FOM) and quality factor (QF), etc. The researchers and scientists are continuously working for the improvement of the performances of the SPR sensors [31, 42–44].

In SPR biosensors, the most crucial parameters determining the characteristics of the sensors are plasmonic materials. Materials with adequate free electrons at their valence bands can be used as plasmonic materials. To be specific, metals e.g. gold (Au), aluminum (Al), silver (Ag), copper (Cu), etc. are a good candidate to be used as a plasmonic material [45, 46]. Al and Cu have not gained much interest to be used because of their high damping nature, prone to oxidation, corrosion, and interband transition characteristics. But Silver (Ag) can be nominated as a potential candidate for SPR sensors as it attributes outstanding optical properties, such as no interband transfer at the visible light frequency, small optical damping, and sharper resonance peak [46–48], etc. Using Ag in SPR sensors, better sensitivity can be captured, but it shows poor chemical stability as it creates brittle oxide layers with liquid analyte [49]. Some researchers have reported that applying bimetallic layer on the Ag surface can resolve this problem [50, 51]. On the other hand, Au is more chemically stable compared to Ag and free of corrosion and oxidation problems. But, gold offers a slightly higher damping loss and widen SPR curve that restricts the detection accuracy and figure of merits (FOM) of the sensors [52]. The sensitivity of Au-based sensors is also slightly lower because of the low biomolecular adsorption characteristics of the gold surface. In order to improve the sensitivity of the sensors, researchers recommended various approaches in which the application

2D Materials Library	Graphene Family	Graphene	hBN 'white graphene'	BCN	Fluorographene	Graphene oxide
	2D chalcogenides	MoS <sub>2</sub> , WS <sub>2</sub> , MoSe <sub>2</sub> , WSe <sub>2</sub> PtSe <sub>2</sub> , SnSe <sub>2</sub>		Semiconducting dichalcogenides: MoTe <sub>2</sub> , WTe <sub>2</sub> , ZrS <sub>2</sub> , ZrSe <sub>2</sub> , and so on	Metallic dichalcogenides: NbSe <sub>2</sub> , NbS <sub>2</sub> , TaS <sub>2</sub> , TiS <sub>2</sub> , NiSe <sub>2</sub> and so on	
					Layered semiconductors: GaSe, GaTe, InSe, Bi <sub>2</sub> Se <sub>3</sub> and so on	
2D oxides	Micas, BSCCO	MoO <sub>3</sub> , WO <sub>3</sub>	Perovskite-type: LaNb <sub>2</sub> O <sub>7</sub> , (Ca,Sr) <sub>2</sub> Nb <sub>3</sub> O <sub>10</sub> , Bi <sub>4</sub> Ti <sub>3</sub> O <sub>12</sub> , Ca <sub>2</sub> Ta <sub>2</sub> TiO <sub>10</sub> and so on		Hydroxides: Ni(OH) <sub>2</sub> , Eu(OH) <sub>2</sub> and so on	
	Layered Cu oxides	TiO <sub>2</sub> , MnO <sub>2</sub> , V <sub>2</sub> O <sub>5</sub> , TaO <sub>3</sub> , RuO <sub>2</sub> and so on			Others	

**Figure 1.** 2D materials library where blue shaded materials are stable at ambient condition, green-shaded are probably stable, pink shaded are unstable at ambient condition but stable at inert condition. The gray shaded materials are 3D but can be exfoliated down to monolayers [60, 61].

of hybrid structures (multilayer structures) are widely used [53–56]. Various 2D materials are used in the hybrid configuration of SPR based sensors. A single atom thick carbon nanostructure (graphene) is often applied on the top of the plasmonic materials to avoid oxidation problems and increase the performance of the sensors because of its chemical inertness and high adsorption characteristics [57, 58]. There are also some other nanomaterials e.g. graphene oxides, graphene carbon nitride (g-C<sub>3</sub>N<sub>4</sub>), transition metal dichalcogenides (TMDs: MoS<sub>2</sub>, MoSe<sub>2</sub>, WS<sub>2</sub>, WSe<sub>2</sub>, PtSe<sub>2</sub>, SnSe<sub>2</sub>, etc.), transition metal chalcogenides (NbSe<sub>3</sub>, TaSe<sub>3</sub>), transition metal oxides (TMOs: LaVO<sub>3</sub>, LaMnO<sub>3</sub>), Black phosphorene (BP), hexagonal boron nitride (hBN), group IV elements [59, 60] and so on which are summarized in the **Figure 1**.

This chapter mainly focuses on the recent trends applied for enhancing the performance of the Kretschmann configuration based prism coupled SPR sensors and their potential applications. The fundamental theory of SPR phenomena is presented first. Then, the method of angular interrogation utilizing attenuated total internal reflection and the performance measuring parameters of the SPR sensors are narrated. Finally, with their compressive architectures, recent developments of the prism coupled SPR sensors are discussed.

## 2. Principle of SPR phenomena

Metals are composed of positively charged nuclei with a lot of free electrons in their conduction band (surface of the metal). If an external electric field is applied close to the metal surface, free electrons are dislocated, resulting in an electric dipole [61]. A longitudinal oscillation has resulted from such electron transportation in a metal surface known as surface plasmons (SPs) [49]. To support the generated SPs a metal and dielectric interface is needed [46] whereas excitation of these SPs leads to an enhanced electromagnetic field resulting in a collective oscillation of free electrons or electron plasma [46, 61, 62]. The basic principle of the construction of SPR based sensors lies in the generation and propagation of electromagnetic waves called surface plasmon wave (SPW) due to the interaction of irradiating electric fields and the generated fields for dislocation of the electrons between the metal-dielectric interface [4]. The SPWs can only be produced by the incidence of a transverse magnetic (TM-) or plane (p-) polarized field as Maxwell's equations supports no solution for transverse electric (TE-) polarized case [46]. Furthermore, the fact that electron oscillation means resistive losses. Thus, when an optical field appears at the metal-dielectric boundary, the SPW produces due to

optical absorption of exponentially decaying evanescent waves. Mathematically, when the wave vector of the SPW is equal to the propagation constant of the irradiating lightwave, maximum absorption of evanescent field is observed leading to a strong SPW generation [63, 64]. This condition is called resonance condition. The propagating evanescent wave can be characterized by propagation constant  $\beta_{ev}$  as follows [10, 65]:

$$\beta_{ev} = \frac{2\pi}{\lambda} n \times \sin \theta \quad (1)$$

Where  $\lambda$ ,  $n$ ,  $\theta$  indicate the incident light wavelength, refractive index of the medium, and angle of incident of light at the metal surface, respectively. The equation as follows characterizes the SPW [66]:

$$\beta_{SPW} = \beta_f \sqrt{\frac{\epsilon_m \epsilon_d}{\epsilon_m + \epsilon_d}} \quad (2)$$

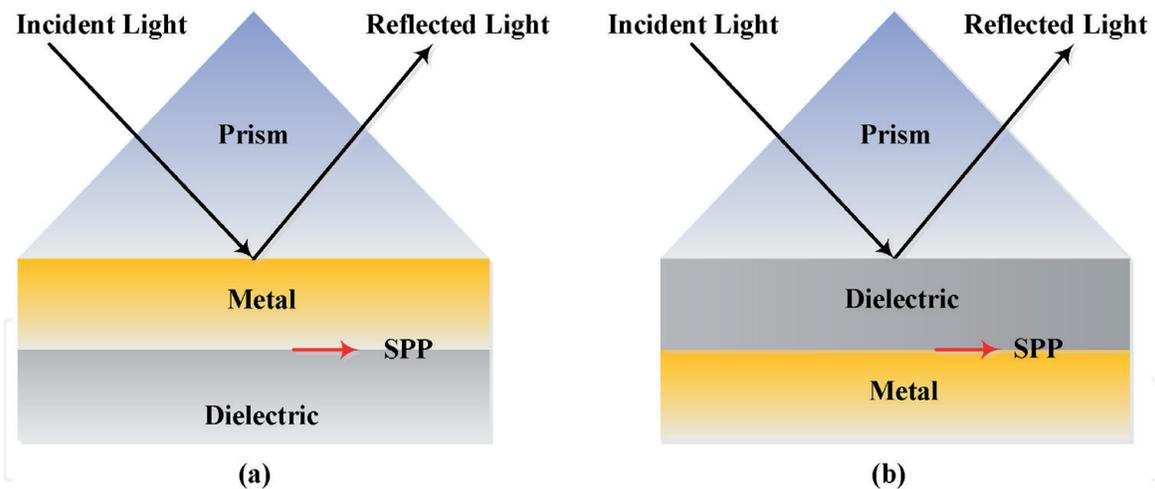
Where  $\beta_f = \frac{2\pi}{\lambda}$ , is the wave vector of light at free space. Also,  $\epsilon_m$  and  $\epsilon_d$  indicate the dielectric constants of plasmonic material and dielectric medium, respectively. Eq. (2) can also be rewritten in terms of the refractive index as follows [31]:

$$\beta_{SPW} = \frac{2\pi}{\lambda} \sqrt{\frac{n_m^2 n_d^2}{n_m^2 + n_d^2}} \quad (3)$$

Where the RI of plasmonic material ( $n_m$ ) and sensing medium ( $n_d$ ) are related to the dielectric constants as  $n_m^2 = \epsilon_m$  and  $n_d^2 = \epsilon_d$ , respectively. The resonance condition is located in SPR based sensor, where the propagation constant of incident light ( $\beta_{ev}$ ) is matched with the SPW's wave vector  $\beta_{SPW}$  [67]. From Eqs. (1) and (3), it can be related that by controlling the incident angle for a particular frequency of light, the resonance condition can be achieved. This method is called the angular interrogation method. Similarly, the light wavelength can be modified to achieve the SPR condition for a particular angle of incident light for the wavelength interrogation method [68]. At this condition, the electrons start to resonate triggering the generation of SPW on the metal-dielectric interface where a sharp loss peak called the SPR point appears. The SPR point is extremely responsive to the refractive index (RI) of the surrounding medium where a minor change in RI of the dielectric (sensing) medium shifts the SPR point to a new state [69].

### 3. Angular interrogation approach and performance parameters of the sensor

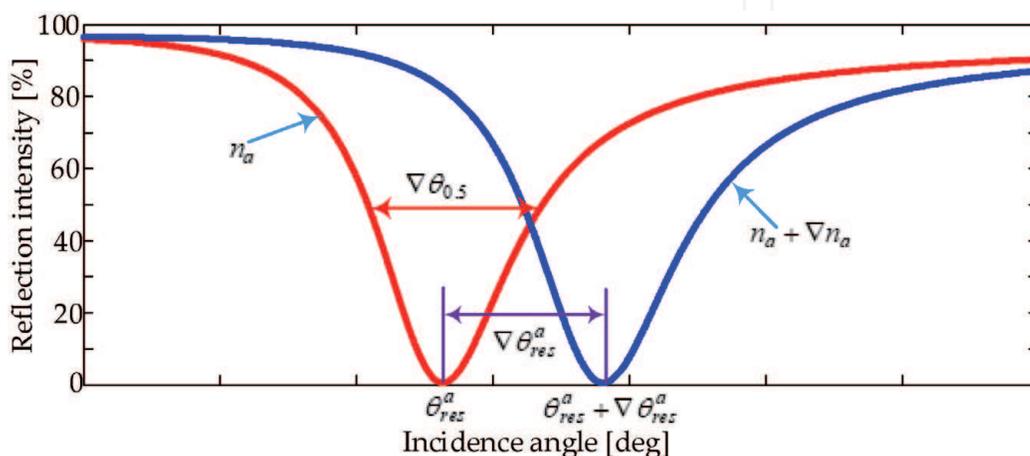
Due to its outstanding performance characteristics, commercial standardization, and ease of manufacturing technology, the angular interrogation method using ATR has become more popular today among various SPR based sensors. When light is directly coupled to the metal-dielectric interface, due to a mismatch of momentum, the SPs are not sufficiently excited to generate SPWs [70]. Researchers have suggested several special arrangements called Otto configuration [71], Kretschmann configuration [72, 73] as visualized in **Figure 2** to alter the momentum of the photon to couple with the SPPs leading to propagation of SPW. In prism



**Figure 2.** Special Arrangements [74] e.g. (a) Kretschmann configuration, and (b) Otto configuration to match the momentum of incident photon and SPW.

based Otto configuration, there is a distance where a dielectric layer with a smaller RI is used between the prism and metal sheet on which the light is employed. On contrary, Kretschmann configuration the metallic layer is in direct contact with the prism. Among them, the Kretschmann configuration is the most popular solution to ensure the coupling of the strongest evanescent wave passing through the metal and generate SPW [53, 74–76]. In the Kretschmann configuration, the light is incident at the metal-dielectric interface through a high index prism [77].

Usually, the incident light bounces back from the interface while the evanescent field is induced by a portion of light penetrating through the metal. For a particular sensor configuration and light frequency, the momentum of the evanescent field is aligned with the wave vector of SPW at a specific angle called resonance angle [76]. Maximum light is coupled to the oscillating electrons at this resonance condition, leading to minimum reflection. If the reflected light is plotted concerning the incident angle, then a resonance dip of reflection spectrum is observed called SPR point which is highly responsive to the RI of the sensing medium. By interrogating this SPR point the analyte can be detected easily. The performance measuring parameters e.g. sensitivity, detection accuracy, FOM, and QF should be as high as possible to eliminate false positive detection. The sensitivity of the sensor operating on the angular interrogation approach depends on the change in the SPR point or resonance angle with a change in RI of the sensing medium. **Figure 3** illustrates the



**Figure 3.** Illustration of the SPR curve variation due to change in sensing medium RI.

SPR curve variation due to change in sensing medium RI where the resonance point is found at  $\theta_{res}^a$  and  $\theta_{res}^a + \nabla\theta_{res}^a$  for sensing medium RI of  $n_a$  and  $n_a + \nabla n_a$ . Due to change in RI of  $\nabla n_a$  the shift in SPR is observed as  $\nabla\theta_{res}^a$ . Thus, the sensitivity ( $S_a$ ) of the sensor with the angular interrogation approach can be measured as [78]:

$$S_a = \frac{\nabla\theta_{res}^a}{\nabla n_a} \quad (4)$$

A sensor's detection accuracy, which depends on the width of the SPR curve, determines how quickly and accurately the SPR point can be measured by the sensor. It is inversely proportional to the width of SPR. If  $\nabla\theta_{0.5}$  is the width of the SPR curve corresponding to 50% reflection then the detection accuracy (D.A.), FOM, and QF can be defined as [15, 79, 80]:

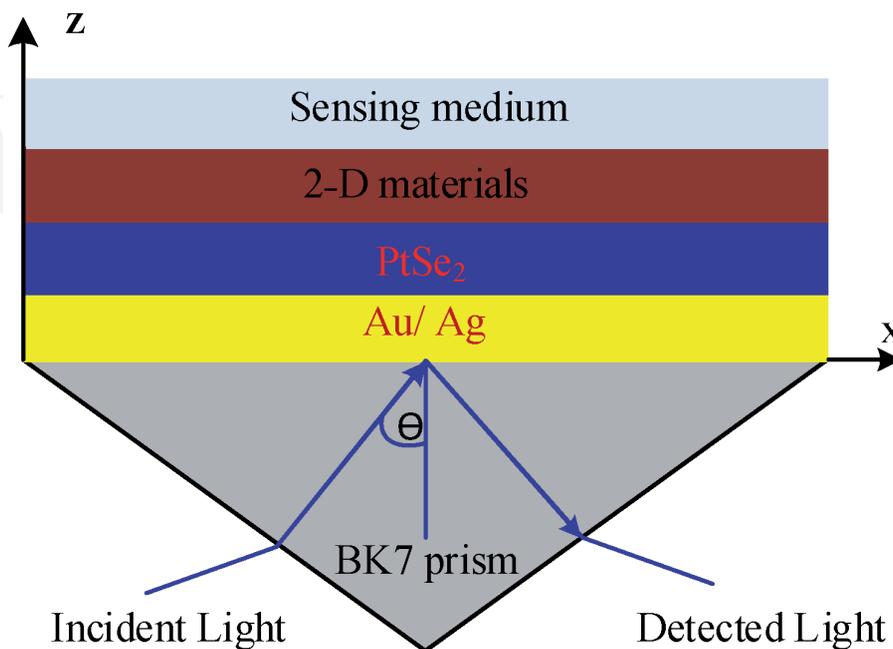
$$D.A. = \frac{1}{\nabla\theta_{0.5}} \quad (5)$$

$$FOM = \frac{\nabla\theta_{res}^a / \nabla n_a}{\nabla\theta_{0.5}} = S_a \times D.A. \quad (6)$$

$$QF = \frac{\nabla\theta_{res}^a}{\nabla\theta_{0.5}} \times S_a \quad (7)$$

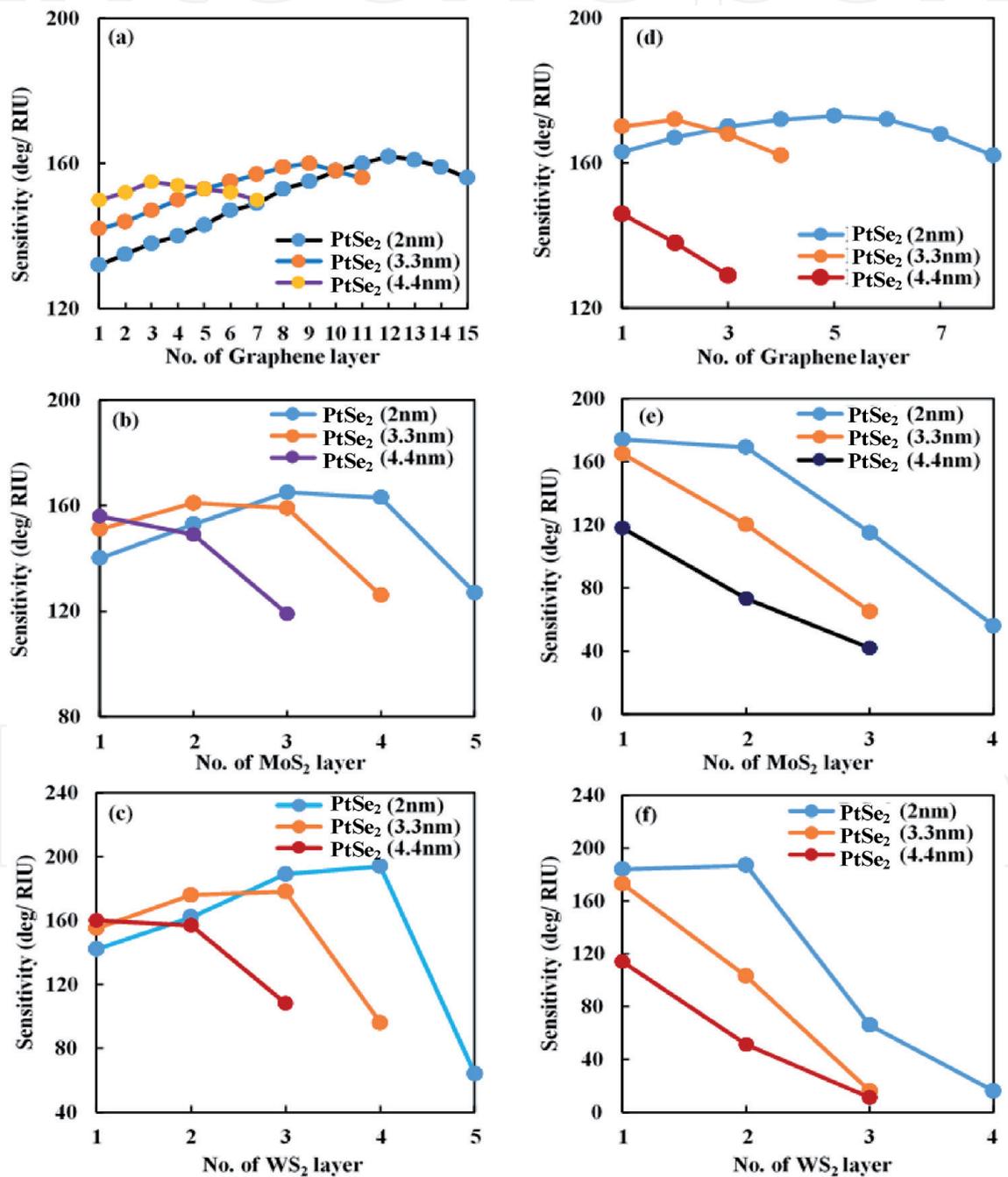
#### 4. Recent trends to enhance the performance of the SPR sensors

Nowadays, the prime concern of scientists, researchers, and academicians are to enhance the performance of the SPR based sensor. To date, several attempts have



**Figure 4.** Schematic Illustration of SPR biosensor employing hybridization of 2D materials with Ag/Au [86].

been reported to attain highly sensitive sensors where the use of bimetallic coating and hybridization of numerous 2D materials along with plasmonic materials are the most popular approach to accommodate the angular interrogation approach. Benaziez S. et al. [81] reported a sensor where Ag is considered as an SPR active material. They showed that the addition of mostly used 2D material graphene on Ag surface enables to reduce the oxidation problem as well as increase the sensitivity up to 9.3%. Yet, the detection accuracy of the sensor is slightly reduced. Also, Rouf H. K. and Haque A. [82] proposed a hybrid structure using InP and Ti with the Ag-Au bimetallic configuration. Their sensor shows maximum sensitivity of 70.90 deg/RIU. Similarly, Mishra S. K. and their team [83] have demonstrated a configuration with excellent sensor sensitivity of 229 deg/RIU. They used a rarely used material



**Figure 5.** Sensitivity variation due to change in the thickness of PtSe<sub>2</sub>, and number of (a) Graphene layer (b) MoS<sub>2</sub> layer; (c) WS<sub>2</sub> layer for BK7/Ag (50 nm)/PtSe<sub>2</sub>/2D materials (Graphene/MoS<sub>2</sub>/WS<sub>2</sub>) hybrid structure; and number of (d) Graphene layer, (e) MoS<sub>2</sub> layer, and (f) WS<sub>2</sub> layer for BK7/Au (50 nm)/PtSe<sub>2</sub>/2D materials (Graphene/MoS<sub>2</sub>/WS<sub>2</sub>) hybrid structure [86].

Rhodium (Rh) with Ag to realize bimetallic configuration. Also, they used a silicon layer on the bimetallic layer to lessen the limitations of Ag. Likewise, N. Mudgal et al. [3] proposed a four-layer hybrid structure that consists of Au, molybdenum disulfide ( $\text{MoS}_2$ ), h-BN (hexagonal boron nitride), and graphene to detect urine glucose. The structure can enhance the sensor sensitivity up to 194.12 deg/RIU with the detection accuracy of 16.04/RIU. In the same way, Hailin Xu et al. [84] proposed an optical sensor with the graphene-Al-graphene sandwich structure where graphene prevents the oxidation issue of Al as well as enhances the sensor sensitivity 3.4 times more than only Al-based sensor. Besides, Wang M. et al. [85] suggested a sensor consisting of graphene, Tungsten disulfide ( $\text{WS}_2$ ), and Au-Ag bimetallic film. They observed that hybridization of single layer graphene and  $\text{WS}_2$  with Au-Ag bimetallic nanostructure leads to sensitivity up to 182.5 deg/RIU which is superior to Au-only based sensor. Incorporating the advantages of hybrid structure and bimetallic configuration, very recently Rahman M. et al. [86] also proposed a new configuration of SPR biosensors utilizing the newly emerged TMDC ( $\text{PtSe}_2$ ) embedded 2D materials as illustrated in **Figure 4**.

In this configuration, a heterostructure of  $\text{PtSe}_2$ /2D material (e.g., graphene,  $\text{MoS}_2$ ,  $\text{WS}_2$ ) has been employed to realize the hybrid configuration whereas BK7 prism is used as a coupler that increases the momentum of the evanescent wave to match with the wave vector of the SPW. The sensor comprises a thin layer (50 nm) of Au or Ag as an SPR active material between the prism coupler and  $\text{PtSe}_2$ /2D material heterostructure. A monochromatic He-Ne laser source having a wavelength of 633 nm have been incorporated to excite the SPPs. The sensor parameters are altered and optimized varying the thickness of  $\text{PtSe}_2$  and number 2D material's layer to get better performance where the results are revealed in **Figure 5**.

The effects of alteration of different parameters of  $\text{PtSe}_2$ , and 2D materials have been analyzed comprehensively and two new sensors have been introduced with excellent performance characteristics. The details of optimized design parameters and performances are listed in **Table 1**. As well, **Table 2** shows the performance comparison of different SPR biosensors based on Kretschmann configuration with a hybrid structure.

Sl. no.	Proposed SPR sensors with optimized structural parameters	Operating range of sensing medium RI	FOM [ $\text{RIU}^{-1}$ ]	QF [deg/RIU]	Sensitivity [deg/RIU]
01.	Ag/ $\text{PtSe}_2$ / $\text{WS}_2$	Thickness of Ag (nm)	1.33-1.38	17.64	34.22
		50			
		02			
		Thickness of $\text{PtSe}_2$ (nm)			
		02			
		Number of $\text{WS}_2$ Layers			
		04			
02.	Au/ $\text{PtSe}_2$ / $\text{WS}_2$	Thickness of Au (nm)	1.33-1.38	15.72	29.39
		50			
		02			
		Thickness of $\text{PtSe}_2$ (nm)			
		02			
		Number of $\text{WS}_2$ Layers			
		02			

**Table 1.**

*Details of optimized design parameters and results of the proposed SPR biosensors [86].*

Ref.	Configuration of the sensors	Sensitivity (deg/RIU)
[83]	Prism/Air/Titanium (Ti)/Ag/Au/InP	70.90
[84]	Prism/Rh/Ag/Si	229
[3]	Prism/MoS <sub>2</sub> /h-BN/graphene	194.12
[85]	Prism/Ag/Au/WS <sub>2</sub> /graphene	182.5
[86]	Prism/Ag/PtSe <sub>2</sub> /WS <sub>2</sub>	194
[86]	Prism/Au/PtSe <sub>2</sub> /WS <sub>2</sub>	187
[87]	Prism/Au/Black Phosphorous (BP)	180
[2]	Prism/Au/Graphene/MoS <sub>2</sub>	89.29
[1]	Prism/Au/MoS <sub>2</sub>	75.34
[88]	Prism/ZnO/Ag/Au/graphene	66
[89]	Prism/Au/MoS <sub>2</sub> /WS <sub>2</sub> /WSe <sub>2</sub>	142
[90]	Prism/Au/MoS <sub>2</sub> /Au film/graphene	182
[55]	Prism/MoS <sub>2</sub> /aluminum (Al) film/MoS <sub>2</sub> /graphene	190.83
[91]	Prism/Ag/PtSe <sub>2</sub>	162
[91]	Prism/Au/PtSe <sub>2</sub>	165

**Table 2.**  
 Sensitivity comparison of Kretschmann configuration based SPR biosensors comprising hybrid structures.

## 5. Conclusion

This chapter provides a detailed description of the surface plasmon resonance phenomenon with the recent trends that are being applied in the advancement of SPR based sensors where the application of hybrid structures as well as bimetallic configurations are found to be potential techniques to enhance the sensor performances. Besides, it demonstrates different 2D materials applied for sensing capability enhancement of the hybrid SPR biosensors. Also, two 5 layer prism based hybrid heterostructures (Prism-Au-PtSe<sub>2</sub>-WS<sub>2</sub> and Prism-Ag-PtSe<sub>2</sub>-WS<sub>2</sub>) have been comprehensively discussed here to show the effectiveness of hybrid technology.

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