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RESEARCH PAPER

Graphene-Perovskite Based Surface Plasmon Resonance Biosensor

Nasih Hma Salah[1, *]

[1]Department of Physics, College of Science, Salahaddin University-Erbil, Kurdistan Region, Iraq

A B S T R A C T:
In this article, a highly sensitive surface plasmon resonance (SPR) based biosensor is proposed, Kretschmar's configuration with angular interrogation as the most successful and popular technique has been used, a laser with 664 nm is impinged on a device that consists of a stratified medium that includes perovskite ($CH_3NH_3PbI_3$), a transfer matrix method for a p-polarized coherent and collimated incident laser beam is used to calculate the reflectivity, and this proposed device is more sensitive for a medium with refractive indices between 1.34 to 1.40. In order to obtain a high enough sensitivity, four different configurations have been brought to the reader's attention. The device has been introduced with the aid of some important quantities to measure the sensitivity, figure of merit (FoM), full width at half-maximum (FWHM) and detection accuracy (DA), their results respectively are $\sim 153.58^o/\text{RIU}$, $\sim 17\ \text{RIU}^{-1}, \sim 10^o\ (\text{at } n_s = 1.34)$ and $\sim 0.1\ \text{Deg}^{-1} (\text{at } n_s = 1.34)$. Finally, these performances are far better than the first simple configuration, and the simulation results are implemented in Matlab codes.

KEY WORDS: Surface plasmon resonance (SPR), perovskite, Kreschmann's configuration, attenuated total reflection (ATR), angular interrogation and sensitivity enhancement.
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1. INTRODUCTION:

A surface plasmon resonance (SPR) sensor is a popular tool for physical, chemical, and medical sensing today because of its ease of use, high sensitivity, real-time detection, and absence of labelling requirements (Patil et al., 2019, Shankaran et al., 2007, Wang et al., 2022). SPR is now a recognized instrument for biosensing, including electrochemical analysis, molecular detection, and DNA-protein, protein-protein, and protein-drug interactions (Khan et al., 2022, Couture et al., 2013). One of the top methods for quick in-situ detection of a variety of biological targets has evolved from SPR measurements of biomolecular interactions on the surface of gold thin films (Boozer et al., 2006, Stahelin, 2013). The SPR sensing technology has developed a name over the last few decades for the detection of chemical and biological analytes, as well as for medical diagnostics and environmental monitoring (Shankaran et al., 2007, Haes and Duyne, 2004).

*** Corresponding Author:**
Nasih Hma Salah
E-mail: nasih.hmasalah@su.edu.krd
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A surface plasmon wave (SPW) propagates along the metal-dielectric interface as a consequence of the SPR sensing principle, an optical phenomenon in which a p-polarized light beam meets a set of resonance requirements, excites a charge density oscillation, and causes an SPW(Hma Salah, 2015, Tudos, 2008). The attenuated total reflection (ATR) design proposed by Kretschmann forms the basis for most SPR sensor structures (Lee*2, 2010).
If the resonance requirement is fulfilled, a thin metal film is deposited on the base of a light-coupling prism, and the p-polarized incident light excites an SPW along the metal-dielectric interface(Salah et al., 2012). Angular sensitivity, which is described as the variation of resonance angle with respect to the change in refractive index of the sensing sample, is one of the most crucial factors describing the performance of an angle modulated SPR sensor. Previous research has shown that, an effective way to increase

sensitivity is to use different metals and their combinations in an SPR sensor(Zhao et al., 2015, Gupta and Sharma, 2005). Because of their excellent optical performance in the visible and near infrared regions, gold (Au), silver (Ag), and copper (Cu) are three highly active metals for exciting SPR in a sensor (Chen and Lin, 2019b). Although silver is strongly oxidizable, it has a narrower SPR curve (less FWHM) than gold because the real part of silver's dielectric constant is slightly larger than gold's one(Chen and Lin, 2019b). However, Gold is the most commonly used because gold SPR sensors have high sensitivity and stable chemical characteristics. It should be mentioned that, Copper is a promising material in the SPR sensing field because it is inexpensive and has reduced optical losses in the visible and near infrared ranges (Kravets et al., 2014, Rifat et al., 2015).

The graphene-based SPR biosensor is more sensitive than the conventional metal film-based SPR biosensor due to the high adsorption of biomolecules on graphene and the excellent optical properties of graphene (Wang et al., 2017). Using an extra graphene layer on the metal surface is another effective method for overcoming the oxidation of Ag and Cu films(Kravets et al., 2014). Choi et al. According to research on the angle-scanned variable-wavelength SPR biosensor, the angular shift is lower at a longer wavelength (890 nm), but the dip and signal-to-noise ratio are better (Choi et al., 2011). However, the sensitivity of graphene-based SPR biosensors with different metal layers at different wavelengths in the visible region has yet to be studied and compared(Chen and Lin, 2019a, Nurrohman and Chiu, 2021).

The most common configuration for plasmon excitation is Kretschman's configuration, where resonance is achieved by the maximal dip in SPR angle relative to the reflection intensity (RI)(Salah, January 1, 2019). Here, it considers the angle interrogation case for a constant wavelength of the incident light source. Improving the performance value of SPR-based biosensors is a significant research issue in light of the current situation and requirements(Wang et al., 2022).

The implementation of two-dimensional heterostructures or Perovskite materials in conventional metal prism-based biosensors can improve SPR biosensor performance, according to theoretical analysis and a review of the relevant literature (Dhibi et al., 2021, Akjouj and Mir, 2022, Nurrohman and Chiu, 2021). Importantly, the dielectric constant of the nano film layer is strongly linked with the maximum sensitivity. Increases in the real component of the dielectric constant result in larger maximum values of sensitivity. In this research, it has been numerically illustrated the functionality of the perovskite nano-film-based SPR and shown improved sensitivity to biological samples as compared to traditional SPR sensors based on Au or Ag layers. In addition, we propose a novel sensing configuration made of Ag-Au bimetallic layers, Perovskite, and graphene with higher sensitivity and resolution, as shown in Fig. (1). To decrease the FWHM, an Ag-Au bimetallic layer is used. Perovskite and graphene can increase light absorption, which is very beneficial for obtaining a high-sensitivity SPR signal. Furthermore, the graphene layer has a high specific surface area and can detect aromatic compounds selectively. The sensitivity and FWHM of an SPR sensor are investigated systematically using Fresnel equations and the transfer matrix method.

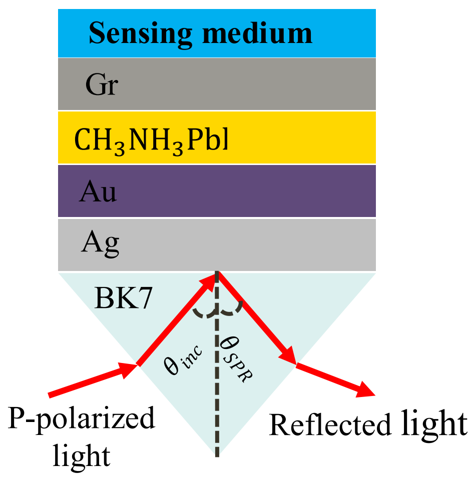


Figure (1): 2D schematic diagram of proposed SPR sensor.

2. THEORY

Light is supposed to enter the system through the first medium (n_1), which is typically a glass prism, and be reflected from the final medium (n_N), which is the sensed layer. Between these two media, one could stack multiple layers of Silver, gold, Perovskite, graphene, etc. to generate a plasmon resonance and enhance the system's sensitivity and affinity (Kurihara and Suzuki, 2002, Xue-feng and Li, 2010, Maharana and Jha, 2012). This structure is shown graphically in Fig. (2):

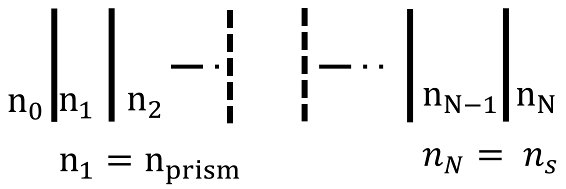


Figure (2): Notation for multilayer system.

To test how well the proposed SPR biosensor works, the well-known transfer matrix method is used on an N-layer system made of metal and graphene layers (Ohta and Ishida, 1990, Chen and Lin, 2019a, Yamamoto, 2002). The prism and the sample are thought to be the incident and rising mediums, respectively. The N-layer structure's characteristic matrix (M) for p-polarized incident light and the total reflectivity of the system are calculated based on Fresnel theory, which is written as follows:

$$M = \prod_{k=2}^{N-1} M_k = \begin{bmatrix} M_{11} & M_{12} \\ M_{21} & M_{22} \end{bmatrix} \quad (1)$$

Where, M_k is given by

$$M_k = \begin{bmatrix} \cos\beta_k & \frac{i \sin(\beta_k)}{q_k} \\ -iq_k \sin\beta_k & \cos\beta_k \end{bmatrix} \quad (2)$$

here, N is the number of layers, β_k and q_k are the phase difference and the phase factor, respectively, that are characterized with the refractive index of prism n_1 and incident light angle θ_{in}, are represented as:

$$q_k = \frac{1}{\epsilon_k}(\epsilon_k \mu_k - n_1^2 \sin^2 \theta_{in})^{\frac{1}{2}}, \qquad \beta_k = \frac{2\pi}{\lambda} d_k \epsilon_k q_k \quad (3)$$

The total reflection coefficient for p-polarized incident light is given by:

$$r_p = \frac{(M_{11} + M_{12}q_N)q_1 - (M_{21} + M_{22}q_N)}{(M_{11} + M_{12}q_N)q_1 + (M_{21} + M_{22}q_N)} \quad (4)$$

where, q_1 and q_N are the phase factor of prism and sensing medium, respectively, as it has been schematically shown in Fig. (2).

The reflectivity (R_p) for the p-polarized light are obtained as

$$R_p = |r_p|^2$$

The parameters, such as the minimum reflectivity R_{min}, sensitivity, FWHM, and FoM, are analyzed to assess the performance of the suggested SPR biosensor. The reflectance curve of the sensor can be used to determine these performance parameters. The minimum reflectivity R_{min} in a typical reflectance curve is achieved at the resonance angle θ_{SPR}, which varies with the variation of the sensing medium's refractive index. The ratio of the difference in the resonance angle θ_{SPR} to the variation in the refractive index of the sensing medium determines the angular sensitivity S of an SPR sensor. The angular sensitivity is given by

$$S = \frac{\Delta\theta_{SPR}}{\Delta n}$$

By calculating the full width at half maximum of the reflectance drop, also known as the FWHM, is equivalent to

$$FWHM = \Delta\theta_{0.5}$$

Thus, the value of FoM can be calculated by

$$FoM = \frac{S}{FWHM}$$

The detecting layer's changing refractive index as a result of the analyte's binding will change the resonant angle, which is the angle with the lowest reflectivity. The sensitivity of an SPR sensor can be described as follows:

$$S(\theta) = \frac{\Delta\theta}{\Delta n} = \frac{d\theta}{dn}$$

Where $\Delta\theta$ and Δn are respectively the variation of resonance angle and refractive index of the sample.

2.1 CHARACTERISTICS AND PARAMETERS OF THE SPR SENSOR

The quality factors of an SPR sensor are crucial in determining how sensitive the device is to detect minute changes on the system's surface where biomolecules adsorb. Sensitivity (S), figure of merit (FoM), and detection precision (DA) are the three key parameters that determine an SPR sensor's effectiveness. The reflectance curve is a function of the variation of the index of refraction (n) with respect to the resonance angle (θ_{SPR}) in the sensing medium. The performance variables are specified as:

Table 1: Materials with its optical constants and thickness.

Layers	Materials used	Refractive index for λ = 664 nm		Thickness/ nm	References
		Real part (*n*)	Imaginary part (*k*)		
Layer 1	Prism (BK7)	1.5141	-	-	(Eid et al., 2021, Hma Salah, 2015)
Layer 2	Silver (Ag)	0.049	4.5185	28	(Maharana et al., 2014)
Layer 3	Perovskite	2.4852	0.25917	5	(Leguy et al., 2015, Ball et al., 2015)
Layer 4	Gold (Au)	0.139	3.7376	3	(Kumar Maharana et al., 2013)
Layer 5	Graphene	3.0	1.21	3 layers	(Salah et al., 2014, Bruna and Borini, 2009)

$$\text{Sensitivity} = \left[\frac{\text{change in resonance angle } (\Delta\theta)}{\text{variationin refractive index } (\Delta n)}\right]; (\text{Unit:}^{o}/\text{RIU})$$

$$\text{FoM} = \left[\frac{\text{Sensitivity}}{\text{FWHM}}\right]; (\text{Unit: } 1/\text{RIU})$$

$$\text{DA} = \left[\frac{1}{\text{FWHM}}\right]; (\text{Unit: } 1/\text{Deg})$$

2.2 METHODS AND TUNING PARAMETERS

A laser (664 nm) is utilized as an excitation source, which impinges on the setup consisting of stratified medium. The thickness of the metals can be scanned to optimize the sensitivity of SPR biosensor (Lin and Chen, 2019) and the definitions and optical characteristics for the proposed biosensor are tabulated in Table (1). Materials that consist of the stratified medium are arranged in such a way that they give high sensitivity, according to the order, the former is always the prism and the last is the sensing medium. All the materials included between the prism and the sensing medium are tabulated in Table (2). There are four different structures, and depending on the sensitivity of the fourth structure, the satisfactory agreement values are; sensitivity, FoM, R_{min}, θ_{SPR}, DA, FWHM and $\Delta\theta_{SPR}$.

Table 2: The possible structures of the SPR instrument.

Structures	Materials used
1st Structure	Prism (BK7)/ Ag
2nd Structure	Prism (BK7)/ Ag)/ Perovskite
3rd Structure	Prism (BK7)/ Ag / Perovskite / Gold (Au)
4th Structure	Prism (BK7)/ Ag/Perovskite/Au/ Graphene

3. RESULTS AND DISCUSSION

Due to its low refractive index at the specified frequency, BK7 prisms provide excellent sensitivity in addition to other parameters like FWHM and angular shift. Meanwhile, the surface of the prism is coated with layers of these metals. Figure 3 illustrates the Sensitivity and R_{min} of Ag, Au, Perovskite, and graphene layers. As it is depicted in Fig. (3a), the variation of Sensitivity and R_{min} with respect to Ag layer thickness is in good agreement at 28 nm, for Au layer 3 nm is the best choice, as shown in Fig. 3(b), for Perovskite layer thickness 5 nm is the best value, the graph is shown in Fig. 3(c), and for Graphene layer 3 which makes the device more sensitive, it is depicted in Fig. 3(d).

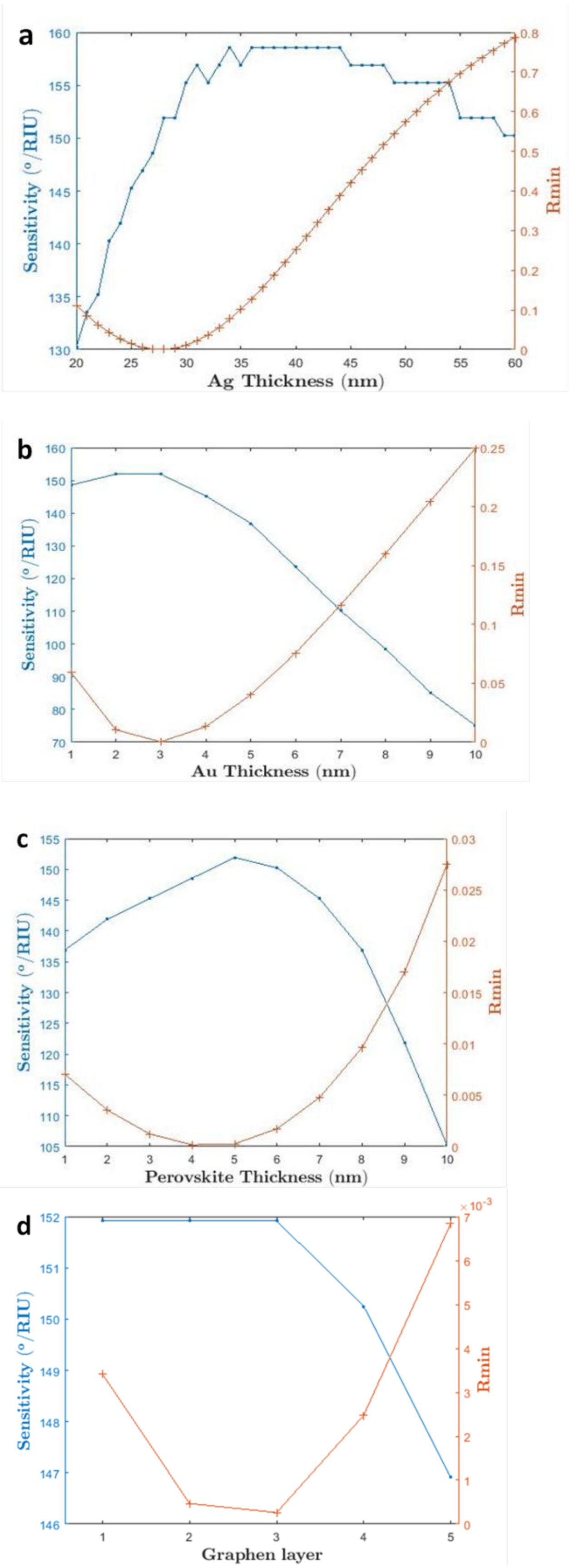
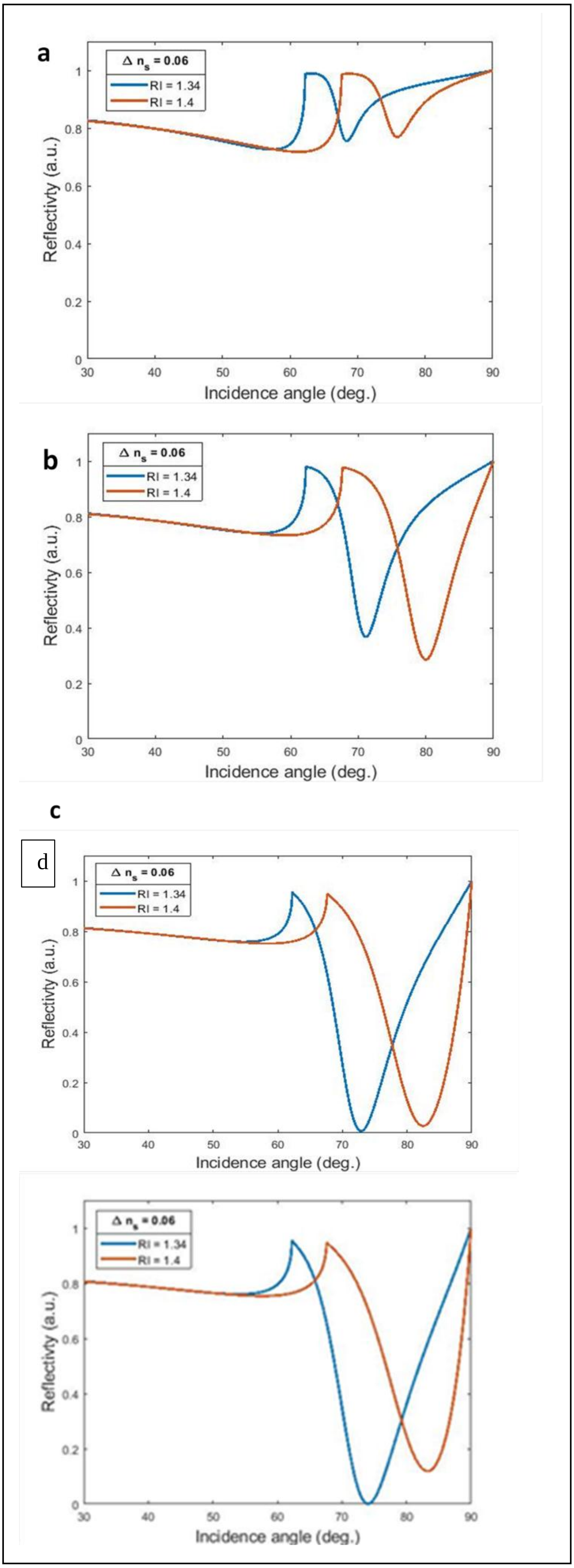


Figure (3): the variation of Sensitivity and R_min with respect to material thickness, for Ag layer (a), for Au layer (b), for Perovskite (c) and for Graphene layer (d).

these metal layers, so it is first determined what the ideal thickness should be for the graphene-perovskite based SPR biosensor (see Table 1 for more information). High sensitivity and low reflectivity (R_{min}) are vital characteristics for the SPR biosensor for improved performance. The sensitivity at minimum reflectivity has a value roughly equivalent to the maximum sensitivity (Jain et al., 2007, Wang et al., 2017). Accordingly, in order to investigate the performance of the suggested SPR biosensor with two distinct metal layers (Au, Ag), the optimal thickness of the Au and Ag layers are determined using monochromatic light at 664 nm (Salah et al., 2012). It should be mentioned that, the energy conservation principle states that when the reflectivity hits a minimum value, the maximum energy from the incident light can be coupled to the surface plasmon polaritons, increasing the sensitivity and resolution of an SPR biosensor (Bryan-Brown et al., 1991, Marusov et al., 2011, Chien and Chen, 2004). As a result, four structures of SPR biosensors have been proposed, as tabulated in Table (2). The first structure was just a silver layer deposited on a BK7 prism, the Perovskite layer was added to the previous one to make structure number two. Structure three is 3 nm Au on top of structure number 2. Finally,
structure number four was BK7, 28 nm silver, 5 nm Au, 3 nm perovskite, and 3 layers of graphene. Figure 4 displays the Reflectivity with respect to the incident angle. Therefore, to achieve a distinct resonance drop in the resonance curve, the metal layer thicknesses are optimized. Figure (4) demonstrates that for a given optimum thickness of the four SPR sensors with different structures, resonance angles vary. The resonant angles are 67^{o} and 68^{o} for the first structure and have sensitivities of 107, as shown in Fig. 5 (a). Meanwhile, as it can be seen in Fig. 5 (c) for structure 4 the resonant angles are 72^{o} and 74^{o} with sensitivities more than 140 by adjusting the refractive index of the analyte by $\Delta n = 0.06$.
Based on the data, we can conclude that a single metallic film is insufficient to achieve the SPR sensor's high sensitivity and resolution. The sensitivity of the SPR sensor improves with growing perovskite, Au layers, and graphene.



Graphene layers can not only increase the sensitivity of an SPR sensor, but also prevent the metal layer from oxidizing and improve the adsorption of biological and chemical molecules (Kravets et al., 2014, Choi et al., 2011, Hma Salah, 2015). We focus on the sensor's efficacy when plasmonic layers are coated with three different structures. The sensitivity verses RI of sensing medium curves of the SPR sensors to determine the optimal structure has been plotted, as shown in Fig. (5). This figure depicts the sensitivity of a graphene-based SPR biosensor with optimal thicknesses of Ag, Perovskite, and Au layers as a function of refractive index. In general, as depicted in this figure, the proposed sensor is more sensitive at some restricted values of the sensing refractive indices, but the values are near as compared to the third configuration, it important to mention that the graphene has significant role in SPR biosensor(Karki et al., 2023).

With the aqua system's refractive index above 1.34, the fourth structure has the highest sensitivity of above 140 °/RIU for restricted refractive index. As can be seen, the sensitivity of the suggested sensors increases as the refractive index increases.

In addition to that, the performance parameters are computed, and they are presented in Table (2). The silver 28 nm, perovskite 5 nm, the gold 3 nm and the three layers of graphene are opted for. All three (1, 2 and 3) estimated structures sensitivities are greater than the sensitivity of a sensor’s first structure made from only Ag film deposited on a BK7 prism, as shown in Fig. (5).

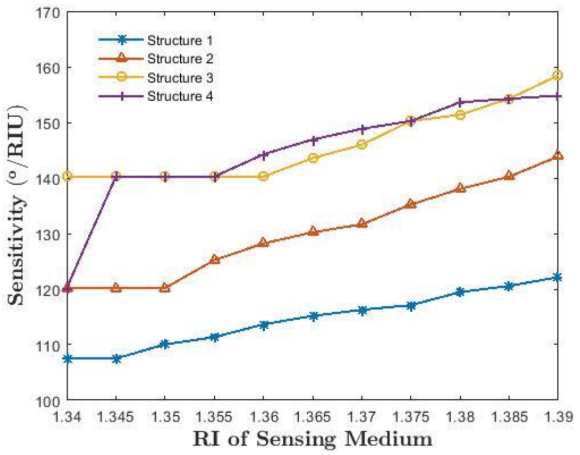


Figure (4): The variation in sensitivity as a function of refractive index for four different proposed SPR structures.

The proposed sensor's minimum reflection light positions, resonance angle, angular shifts, and FWHM are displayed in Table (3). Also, the parameters that are feasible, such as sensitivity, FoM and DA are listed in Table (4). We investigated the suggested sensor response for various refractive index values based on the optimal thicknesses of all layers, including silver, Au, and Perovskite, and the optimal number of graphene sheets.

Finally, optimized parameters of the proposed sensor are graphed with respect to the refractive index of the sensing medium that is restricted between 1.34 and 1.40. In order to simplify results, all graphs are compared to make brevity and distinguishable values in each refractive index, so one can find the sensor based requested parameters (i.e., the region of high performance), the results are summarized in Fig. (6).
A tabular comparison of the proposed and previously published studies on SPR sensors is also provided in Table (5) It is obvious that this work exhibits the greatest sensitivity. We think that the proposed SPR device, which is based on BK7/Ag/Au/Perovskite/Graphene structure, can be widely utilized in the field of chemical and biological sensing technologies due to its exceptional performance.

Table 4: The second set of optimized values of performance parameters are summarized.

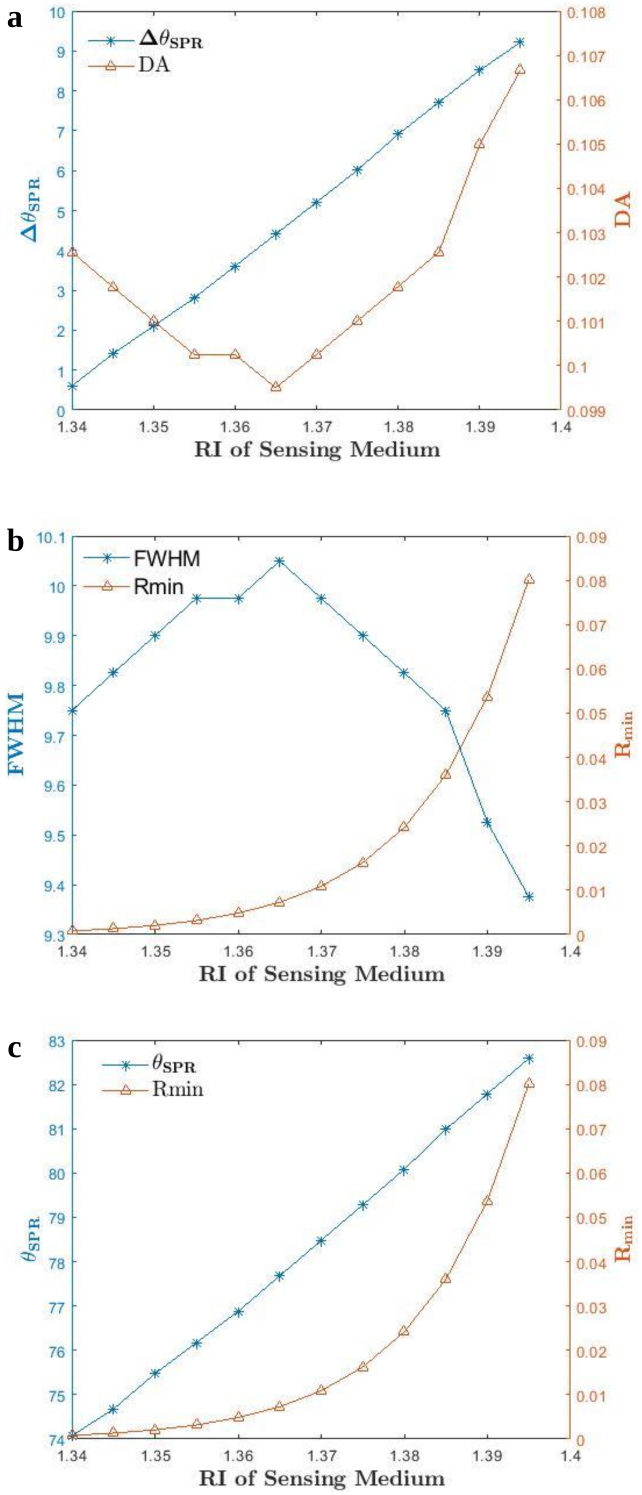
Structure	**Sensitivity**	**FoM**	**DA**	
	$\Delta n_s = 0.06$	$\Delta n_s = 0.06$	$n_s = 1.34$	$n_s = 1.4$
1st Structure	124.1514	10.34	0.101	0.083
2nd Structure	146.9115	23.04	0.180	0.156
3rd Structure	160.2671	18.10	0.119	0.113
4th Structure	153.5893	16.78	0.102	0.109

Table 5: Comparison of prior and present work

Structures	**Sensitivity (deg./RIU)**	**References**
2S2G/Ag/Au/Graphene	36.08	**(Wang et al., 2021)**
SF10 /Au/Graphene	62.30	**(Wu et al., 2010)**
SF10/Au/Si/Graphene	134.60	**(Verma et al., 2011)**
BK7/Ag/Au/ Perovskite/Graphene	153.58	**Proposed work**

Table 3: The first set of optimized values of performance parameters are summarized.

Structure	θ_{SPR}		$\Delta\theta_{SPR}$	R_{min}		**FWHM**	
	$n_s = 1.34$	$n_s = 1.4$	$\Delta n_s = 0.06$	$n_s = 1.34$	$n_s = 1.4$	$n_s = 1.34$	$n_s = 1.4$
1st Structure	68.42	75.87	7.45	0.75	0.77	9.83	12.00
2nd Structure	71.16	79.98	8.81	0.37	0.29	5.55	6.38
3rd Structure	72.87	82.49	9.62	0.0077	0.0278	8.40	8.85
4th Structure	74.07	83.29	9.22	0.0008	0.1188	9.75	9.15



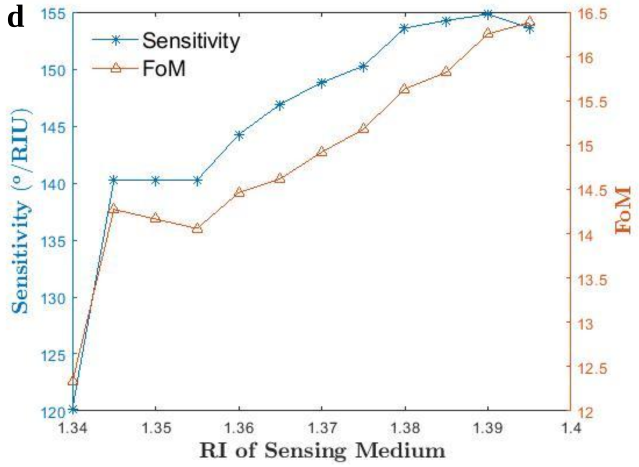


Figure (5): Performance parameters graphs for the proposed sensor based on data analyzed for restricted refractive index of sensing medium (1.34 to 1.40).

4. CONCLUSION

In this article, a SPR biosensor is proposed with the aid of perovskite, Au, Ag and graphene, arranged as a stratified medium in Kreschmann's configuration. Materials are sorted to give the performance parameter of a good biosensor. Adding Perovskite active materials to the biosensor encourage the researchers to provide cost effective devices with more sensitivity, in another side, having graphene with the perovskite does not reduce the sensitivity, which is a significant result for those who have problem with graphene. For the sake of brevity, angular interrogation is the most popular technique used to measure the higher sensitivity of the fourth structure. A sensing medium with limiting values of refractive index from 1.34 to 1.40 is possible to use with this device. We provide some performance parameters like angular sensitivity, FoM and Δθ_SPR are respectively have the values of 〖153.58〗^o/RIU, 17 (1/RIU), 9^o at Δn_s=0.06, and also, FWHM, DA, Rmin and θ_SPR respectively have the values 〖9.75〗^o, 0.1 (1/Deg), 0.0008 (a.u.) and 74^o at specific aqueous solutions (RI=1.34).

Conflict of interest

The author declares no conflict of interest.

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