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Design and Analysis of Graphene Based SPR Biosensor Using Ellipsometry Method

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Abstract- Two-dimensional nanomaterials have attracted increasing attention for enhancing surface plasmon resonance (SPR) biosensors application. In this work, we use the graphene layer to improve the sensitivity of the SPR biosensors based on conventional Kretschmann configuration. We employ WS_2 and MoS_2 2D materials as an interlayer to enhance the sensitivity of Au/Graphene biosensor in angle interrogation method. Transfer matrix method (TMM) is used to analyze the characteristics of the device. Results show that using WS_2 in Au/Graphene structure increases sensitivity about 12.64%, which is higher than MoS_2. Combining graphene based SPR and ellipsometry as a highly sensitive, label-free, real-time, and versatile method can be used to measure a very small concentration of biomolecules which leads to 170-fold enhancement compared to angle interrogation method.

Keywords: surface plasmon resonance; graphene; transition metal dichalcogenides (TMD); Ellipsometry

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1. Introduction

SPR based biosensors have attracted great attention due to high sensitivity, reliability, label-free, and ability to real-time detection. Due to poor interaction between biomolecules with conventional Kretschmann SPR chip, the graphene layer was introduced as BRE [1]. Graphene, a 2D-material with honey-comb structure, has π-π stacking interaction with biomolecules, so it provides a highly sensitive sensor and improves the efficiency of conventional prism/Au chip. Furthermore, due to effective charge transfer in Au/Graphene interface, electric field enhancement is generated. Also, 2D transition metal dichalcogenides (TMD) were proposed as interlayer to enhance the sensitivity of biosensors [2]. However, the phase measurement of these structures has not been investigated in detail yet. In this work, highly sensitive surface plasmon resonance enhanced ellipsometry have been used to probe the phase response of graphene based Kretschmann SPR structures. Then, the calculated sensitivity of these structures in angle interrogation method has been compared to ellipsometry results.

2. Theory

Figure 1a shows a SPR sensor that is based on the Kretschmann configuration. In the interlayer portion, WS_2 and MoS_2 are used. The reflectance (R) of the structure shown in Figure 1a can be calculated for TM polarized light with the N-Layer model [1, 2]. For TM polarized light, the reflectance is shown by R as a function of matrix elements,

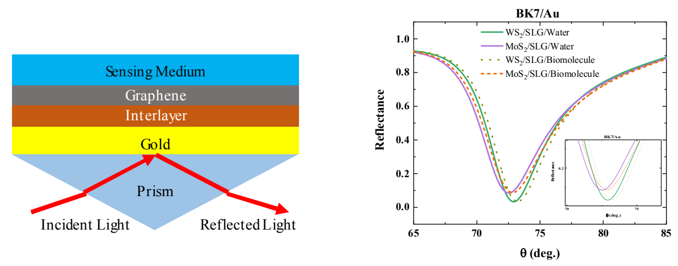
$R = \left| \frac{(M_{11} + M_{12}q_N)q_1 - (M_{21} + M_{22}q_N)}{(M_{11} + M_{12}q_N)q_1 + (M_{21} + M_{22}q_N)} \right|^2$, given by $M_{ij} = \left(\prod_{k=2}^{N-1} M_k \right)_{ij}$, $i, j = 1, 2, ...$ in which

$$M_k = \begin{bmatrix} \cos\beta_k & -i \sin\beta_k / q_k \\ -iq_k \sin\beta_k & \cos\beta_k \end{bmatrix}, \ \beta_k = d_k \left(\frac{2\pi}{\lambda_0} \right) (\varepsilon_k - n_1^2 \sin^2\theta)^{1/2}, \text{ and } q_k = \frac{(\varepsilon_k - n_1^2 \sin^2\theta)^{1/2}}{\varepsilon_k}, \quad (1)$$

where, λ_0 is the wavelength of incident TM-polarized light, which is considered 633 nm, θ is the incident angle, n_k and d_k are the refractive index (RI) and the thickness of the kth layer,respectively, with $k=2$ to N-1. The first layer is BK7 (n=1.5151) prism ($k=1$) or SF10 (n=1.7231) prism. The Nth is the analyte defined as n_w=1.33 (water medium) and changes to n_{bio}=1.332 as biomolecule and immobilizer. The RI of Au is calculated with Drude-Lorentz model, which is 0.1378+3.6196i at 633 nm. The thickness of monolayer graphene, MoS_2, and WS_2 is 0.34 nm, 0.65 nm, and 0.8 nm, respectively. Moreover, their corresponding RI at 633 nm are 3+1.1487i, 5.0805+1.1723i, and 4.8937+0.3123i, respectively. The reflectance is related to change in RI of sensing medium, and dip in reflectance shows resonance angle (Figure. 1b). As RI of sensing medium increases with Δn, the dip in reflectance shifts to a higher value that can be considered as Δθ. The sensitivity is defined by $S = \frac{\Delta\theta}{\Delta n}$. For ellipsometry measurement, we use the same approach that is proposed in [3].

3. Results and Discussion

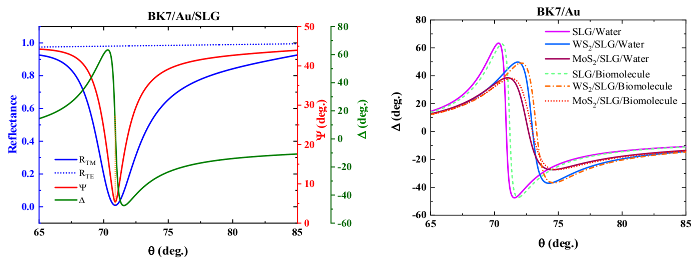
Figure 1b shows reflectance results for WS_2/SLG (Single Layer Graphene) and MoS_2/SLG in water and biomolecules medium on the BK7/Au substrate. Results show that using MoS_2 and WS_2 (Fig. 1b) as interlayer improves the sensitivity of biosensor in angle interrogation method. In the resonance angle, there is a linear and abrupt change in phase, which provides a very high sensitive measurement that can be used to monitor very slight changes in the sensing medium (Figure 2a). Figure 2b shows the phase response of SLG, WS_2/SLG, and MoS_2/SLG in water (Solid lines) and biomolecules (Dash, Dot, Dash-Dot lines respectively) medium on BK7/Au substrate. Table 1 gives us information about different structures used in this work. From Table 1, BK7/Au/ WS_2/SLG has the best sensitivity in angle interrogation (156 °/RIU). So, adding MoS_2 and WS_2 as an interlayer enhances the sensitivity, which has two main reasons. Firstly, adding an additional layer leads to an increase in the slope of θ-n_d dispersion, so increasing RI of medium leads to Δθ gives higher value compared to the absence of interlayer. Secondly, effective charge transfers in Au/ WS_2 or MoS_2/Graphene, lead to larger electric field enhancement, which results in higher sensitivity to sensing medium. Table 1 shows that phase response of all structures provides much higher sensitivity to measure small change in sensing medium and Au/SLG structure has the best sensitivity. In Au/SLG structure, the ellipsometry technique provides the sensitivity enhancement of more than 170 fold compared to angle interrogation. So, without adding any layer, high sensitive approach with a simple Au/Graphene SPR structure can be achieved.



(a) (b)

Figure 1. (a) The schematic of the proposed five-layer SPR structure. (b) SPR curve for BK7/Au/WS_2 or MoS_2/Water or Biomolecule.

Table 2 shows the results of using SF10 prism instead of BK7, which has a lower sensitivity in angle interrogation. Ellipsometry results show that SF10 prism provides slightly lower phase sensitivity in graphene based structures. Although by adding TMD nanomaterials, SF10 based SPR chips show better phase response compare to BK7 based SPR chip.



(a) (b)

Figure 2. (a) Reflectance of TM (solid blue line) and TE (blue dashed line) light, Ψ (red), and Δ (olive) for BK7/Au/SLG orange vertical line shows that in resonance angle abrupt change in phase occurs. (b) Ellipsometry results for different structures.

Table 1 Simulation results with BK7/Au substrate

Structure	Θ_{SPR} Water (W)	Θ_{SPR} Biomolecule (Bio)	$\Delta\theta_{SPR}$	Sensitivity (°/RIU)	FWHM (W)	FWHM (Bio)	Δ
SLG	70.898°	71.175°	0.277°	138.5	4.303°	4.371°	47.158°
Bilayer Graphene	71.288°	71.571°	0.283°	141.5	4.826°	4.874°	27.736°
Trilayer Graphene	71.697°	71.986°	0.289°	144.5	5.264°	5.310°	17.835°
WS_2/SLG	72.917°	73.229°	0.312°	156	5.450°	5.478°	24.248°
MoS_2/SLG	72.529°	72.833°	0.304°	152	5.778°	5.799°	12.362°

Table 2 Simulation results with SF10/Au substrate

Structure	Θ_{SPR} Water (W)	Θ_{SPR} Biomolecule (Bio)	$\Delta\theta_{SPR}$	Sensitivity (°/RIU)	FWHM (W)	FWHM (Bio)	Δ
SLG	56.397°	56.545°	0.148°	74	2.269°	2.282°	47.105°
Bilayer Graphene	56.614°	56.765°	0.151°	75.5	2.798°	2.815°	27.100°
Trilayer Graphene	56.840°	56.993°	0.153°	76.5	3.091°	3.109°	17.804°
WS_2/SLG	57.501°	57.659°	0.158°	79	3.230°	3.248°	25.600°
MoS_2/SLG	57.303°	57.459°	0.156°	78	3.535°	3.554°	12.589°

4. Conclusion

2D TMD materials like WS_2 and MoS_2 have been used as an interlayer to enhance Au/Graphene sensitivity. BK7/Au/WS_2/SLG chip shows the most sensitivity in angle interrogation with an enhancement of about 12.64% compared to BK7/Au/SLG. In angle interrogation using, BK7 prism leads to higher sensitivity compared to SF10 prism, but with using TMD materials as an interlayer SF10 based structures have shown better sensitivity in phase measurement. Ellipsometry results show BK7/Au/SLG has the best phase response. Using the ellipsometry technique as a non-destructive, high sensitive measurement provides sensitivity enhancement more than 170 fold compares to the angle interrogation.

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