Sensitivity Enhancement of Silver Based Surface Plasmon Resonance Sensor via Graphene-Dielectric Composite Structure

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Abstract

In this paper, a silver based surface plasmon resonance (SPR) sensor with graphene and dielectric layer was presented. The influences of dielectric layer and graphene on sensitivity and other sensing properties were theoretically calculated and then comprehensively discussed. The refractive index sensitivities for composite silver film based SPR sensors with graphene and dielectric layer could be increased by 29% and 288% than that of monolayer silver film based SPR sensor, respectively. Further, the sensitivity could be enhanced by 202% when combining graphene and dielectric layer together. Considering the high adsorptive capacity of graphene for biochemical molecules, the composite silver film with both dielectric layer and graphene would have great potential application in biochemical sensing fields. Further, BSA protein was successfully used to verify the biochemical sensing ability of proposed SPR sensor. The shift of resonance angle is nearly 3.1 folds than that of monolayer silver based SPR sensor.

Keywords Surface plasmon resonance sensor; Graphene; Dielectric layer; Sensitivity; BSA detection
Introduction

Silver based surface plasmon resonance (SPR) sensor has been widely studied since its more excellent sensing performance than that of traditional gold based SPR sensor [1, 2]. However, its poor chemical stability limits the further application. In order to protect silver film in stability, these structures of silver/gold double layer [3-5], silver/metal oxide layer [6, 7], silver/organic molecular layer [8, 9] and silver/other dielectric layer [10-12] have been employed. In order to ensure the stability of the silver film, a thick dielectric layer is usually required, which will limit the optimization of sensing performance in terms of thickness.

Two-dimensional nanomaterials have also been widely used in the field of SPR sensor due to their excellent optical and electrical properties, such as optical band transparency and high specific surface area [13-15]. The composite structure based on silver and two-dimensional nanomaterials has also attracted much attention [16, 17]. The typical silver/graphene structure and silver/TMDCs (transitional metal dichalcogenides) structure have not only improved the refractive index (RI) sensitivity of the silver based SPR sensor, but also could improve its stability. Wu et al. theoretically calculated the RI sensitivity of the silver/BP (black phosphorus)/graphene/TMDCs based SPR biochemical sensor, and the introduction of two-dimensional nanomaterials such as BP, graphene or TMDCs improved the RI sensitivity with more than 200 deg/RIU(refractive index unit) [18]. Further, Xia et al. used genetic algorithm to optimize the sensing structure, and when the thicknesses of silver film and BP are 49 nm and 12 layers (approximately 6 nm), the RI sensitivity could reach as high as 400 deg/RIU [19]. Wu et al. also improved the stability and sensitivity of the silver based SPR sensor via introducing several layers of two-dimensional Ti3C2Tx MXenes, and theoretical calculations showed that the RI sensitivity was 28.4% higher than that of the conventional silver based SPR sensor [20].

Graphene is a special two-dimensional nanomaterial, due to its high specific surface area, a theoretical value of 2630 m²/g, which is conducive to increasing the adsorption capacity of biomolecule [21-23]. Szunerits et al. successfully modified the
graphene surface with biotinylated bovine serum protein (BSA), and the adsorption capacity is 5 folds higher than that of gold membrane [21]. Owing to its high density and excellent optical properties, the introduction of graphene would provide the possibility of realizing high stability and high sensitivity for silver film based SPR sensing [4, 24, 25].

In summary, the introduction of dielectric layer and two-dimensional nanomaterials can improve the stability and sensitivity of the silver based SPR sensor [26]. However, few research investigates their effects on sensing performance when considering both dielectric layer and two-dimensional nanomaterials (it is worth noting that the dielectric has not an imaginary part of RI and the RI of the two-dimensional nanomaterials is complex in the paper). Further, since a certain thickness of dielectric layer is required to protect silver film better and the defects in two-dimensional nanomaterials reduces the actual protection effect, we presented a structure of graphene/dielectric/silver/BK7 based silver SPR sensor to enhance stability. The introduction of graphene can protect both the silver film and dielectric layer, and make it possible to optimize the sensing structure of silver based SPR sensor.

**Theoretical Modeling of the Proposed Sensor**

In this proposed composite silver film based SPR sensor, the Kretschmann configuration via BK7 prism [9] is used to excite SPR mode, as shown in Fig. 1. The transfer matrix method is employed to calculate its reflectivity versus incident angle spectrum based on Fresnel reflection theory [27]. And then, its sensing performance would be calculated based on these angular spectra. In order to enhance the stability of silver film, a 2-nm Cr layer is deposited on the BK7 glass as an adhesion layer before the deposition of silver film [9]. Thus, the Cr film is added in our sensing structure. The wavelength of incident light with transverse magnetic (TM) mode is set as 633 nm, and thus the corresponding calculation parameters of different layers, including refractive index and thickness, are shown in Table 1. The thickness of graphene monolayer is 0.34 nm.
**Fig. 1** Schematic diagram of proposed silver based SPR sensor with graphene/dielectric/silver/BK7 structure

| Layer                | Refractive index         | Thickness (nm) |
|----------------------|--------------------------|----------------|
| BK7 glass            | 1.515 [28]               | ∞              |
| Cr                   | 3.1395+i3.3152 [29]      | 2              |
| Silver film          | 0.056206+i4.2776 [30]    | 10–90          |
| Dielectric layer     | 1.35~5                   | 0–30           |
| Graphene             | 3.0+i1.1491 [31]         | 0–12 layers    |
| Sensing medium (water) | 1.33+Δn               | ∞              |

The evaluation indexes for sensing performance we used are RI sensitivity, figure of merit (FoM), penetration depth, field enhancement and RI range. The RI sensitivity is given as [32]:

$$S = \frac{\Delta \theta_R}{\Delta n}$$

(1)

where S represents the RI sensitivity, $\Delta \theta_R$ is the difference of resonance angle with the RI change of sensing medium, that is $\Delta n_s = 1.335 - 1.33 = 0.005$ RIU. Based on
the obtained RI sensitivity, the FoM is [33]:

\[ \text{FoM} = \frac{S}{2 \times \text{LHWHM}} \] (2)

where LHWHM is the left half width at half maximum (LHWHM). Since the SPR angular spectrum is asymmetric and the LHWHM is easier to be obtained than that of the right half width at half maximum, the LHWHM is adopted rather than full width at half maximum. The electric field intensity decays in the sensing medium and the penetration depth is defined as the distance at where the electric field intensity is 1/e of that at the sensor/sensing medium interface. The electric field intensity enhancement factor (EF) in this paper is the averaged electric field intensity enhancement within 20 nm from the sensor/sensing medium interface into the sensing medium, that is:

\[ \text{EF} = \frac{\int |E|^2 dz}{|E_0|^2 \int dz} \] (3)

where E is electric field intensity, z is normal distance and E₀ is electric field intensity at the BK7/Cr interface. For simplicity, the field enhancement would be used instead of EF in the following paragraphs.

The BSA protein was used for sensing application of the optimized silver based SPR sensor. Assuming homogeneous adsorption of BSA molecule on the sensor’s outer surface, the RI of the closely packed BSA monolayer could be calculated to be \( n_{\text{BSA}} = 1.44 + 5000/\lambda^2 \) and \( \lambda \) is the wavelength of incident beam [34]. It was calculated to be 1.452 when the wavelength of incident light is 633 nm. The RI of the BSA adlayer (\( n_{\text{ad}} \)) was calculated by using the Lorentz-Lorenz equation described below [34, 35]:

\[ \frac{n_{\text{ad}}^2 - 1}{n_{\text{ad}}^2 + 2} = \frac{n_{\text{BSA}}^2 - 1}{n_{\text{BSA}}^2 + 2} f_{\text{BSA}} + \frac{n_s^2 - 1}{n_s^2 + 2} (1 - f_{\text{BSA}}) \] (4)

where \( f_{\text{BSA}} \) presents the volume fraction of adsorbed BSA in the monolayer adlayer.

**Sensing Effects of Multilayer Structure with Dielectric and Graphene**

Before optimizing the graphene/dielectric/silver/BK7 based SPR sensor, the structure of graphene/silver/BK7 and dielectric/silver/BK7 were calculated for comparison.
For calculating the structure of graphene/silver/BK7, the number of graphene layers is set as 0 ~ 12 layers and other parameters are shown in Table 1. The results in Fig. 2 indicate that the thickness of silver film and the layers of graphene have significant influences on the RI sensitivity, figure of merit, penetration depth and average field enhancement. As shown in Fig. 2(a), the RI sensitivity increases firstly with increasing the thickness of silver film and then keeps almost unchanged when the thickness is over 40 nm. However, the conclusion is not applicable when the number of graphene is over 6 layers and the RI sensitivity would increase first and then decrease as the thickness of silver film increases. Thus, in order to obtain a greater RI sensitivity, the optimal thickness range of silver film is 40 ~ 80 nm. With increasing graphene layers, the RI sensitivity also shows a significant upward trend at the optimal thickness of silver film, as shown in Fig. 2(a), and the highest RI sensitivity could reach approximately 140 deg/RIU, while the RI sensitivity for monolayer silver SPR sensor is about 116 deg/RIU. Other sensing properties, such as figure of merit, penetration depth and average field enhancement factor all have an optimal thickness range for silver film as shown in Fig. 2(b), 2(c) and 2(d), respectively. Different from RI sensitivity, they all showed a significant decreasing trend with the increase of graphene layers, which might be related to the increased angular peak width and decreased coupling efficiency at the resonance angle with the increase of graphene layers, as shown in Fig. 2(e), the reflectivity vs incident angle spectrum. That means, the increased number of graphene layers would widen the spectrum of reflectivity vs incident angle and decrease the intensity of transmitted beam.
Fig. 2 The effect of different graphene layers and silver film thickness on the sensing performance without dielectric layer, (a) RI sensitivity, (b) figure of merit, (c) penetration depth away from sensor/sensing medium interface, (d) average field enhancement factor, (e) the changes of angular spectrum with 60-nm silver film, and RI sensitivity (f) with larger change of graphene layers

Pay attention that, Fig. 2(e) also shows that the resonance angle has a red shift with increasing graphene layers, which might be the reason why the RI sensitivity was improved when adding the number of graphene as shown in Fig. 2(a). Further, we enlarged the variation range of graphene layer and calculated the corresponding RI sensitivity at different thicknesses of silver film as shown in Fig. 2(f). One can easily find that the RI sensitivity climbs up and then declines with respect to the number of graphene layer and there is a cut-off number of graphene layer. And the highest RI sensitivity is about 150 deg/RIU for graphene/silver/BK7 based SPR sensor with 40-nm silver film and 20-layer graphene.
In order to clarify the influence of dielectric layer on the sensing performance, relevant theoretical calculation based on the multilayer reflection theory and the transmission matrix method was also carried out, as shown in Fig. 3. According to the above theoretical calculation results, the thickness of silver film and the number of graphene are set as 60 nm and 12 layers, respectively. And the optical parameters of the dielectric layer are thickness and refractive index, which are 0 ~ 30 nm and 1.35 ~ 5 RIU, respectively, as shown in Table 1.

![Fig. 3](image)

**Fig. 3** The influence of the dielectric layer with different thickness and RI on the sensing performance, (a) RI sensitivity, (b) figure of merit, (c) penetration depth in sensing medium, (d) average field enhancement factor, with the 60-nm silver film and 12-layer graphene

For calculating the RI sensitivity, the RI difference of sensing medium is also set as $\Delta n_s = 0.005$ RIU. Fig. 3 indicates that the RI sensitivity, figure of merit, penetration depth and average field enhancement factor almost all have a certain symmetrical relation with the thickness and RI of the dielectric layer. That means, the influences of the RI and thickness of dielectric layer on sensing properties are not isolated. The blank area in Fig. 3(a) and 3(b) means that SPR mode is nearly not generated at those
situations or the difference of resonance angles is nearly zero and even negative when
\[ \Delta n_s = 0.005 \text{ RIU}. \] Thus, those cut-off conditions are ignored in our later discussions. It
can be obviously found that the RI sensitivity is positively correlated, while the other
properties are negatively correlated with the RI or thickness of dielectric layer. The RI
sensitivity is up to about 160 deg/RIU and is improved by 38\% compared with that of
monolayer silver film based SPR sensor. And the corresponding reduction rate of FoM
and penetration depth are about 22\% and 19\%, respectively. Therefore, optimization of
dielectric layer structure can significantly improve the RI sensitivity of silver based
SPR sensor.

Fig. 4 The effect of different graphene layers and silver film thicknesses on the sensing
performance, the dielectric layer thickness is 5 nm and the RI is 3 RIU, (a) RI sensitivity, (b)
figure of merit, (c) penetration depth and (d) average field enhancement factor

As shown in Fig. 4, considering the presence of dielectric layer, for example, when
the thickness is 5 nm and RI is \( n = 3 \), we also theoretically calculated the relationship
between its sensing performance and the silver film thickness or the number of
graphene layers. It can be obviously seen that there is also an optimal range for silver
film thickness, that is 40 ~ 70 nm, which is shorter than that without dielectric layer,
that is 40 ~ 80 nm. The largest RI sensitivity can reach nearly 200 deg/RIU, which is 72% higher than that of the monolayer silver film based SPR sensor. Therefore, the graphene layer is more suitable as a protective or adsorbed layer and the optimized dielectric layer has a more obvious improvement on the RI sensitivity. Further, we also calculated RI sensitivity when the number of graphene layer is zero or monolayer via changing RI and thickness of dielectric layer, as shown in Fig. 5(a) and 5(b). The largest RI sensitivities for those cases are at least 450 and 350 deg/RIU, respectively.

![Graph showing variation of RI sensitivity](image)

**Fig. 5** Variation of RI sensitivity with respect to RI and thickness of dielectric layer for zero-layer graphene (a) and monolayer graphene (b), resonance angle (c) and RI sensitivity (d) depending on the RI of sensing medium for different sensing structures

The results obtained above could be summarized as that, both graphene and dielectric layer can enhance the RI sensitivity although the other evaluation indexes would be weakened and the former is more suitable as a protective or adsorbed layer and the latter has a more obvious improvement on the RI sensitivity.
Four Typical Silver Based SPR Sensor

Based on the above discussions, four typical silver based SPR sensor would be discussed latter. The RI of different layers could be found from Table 1 and other calculating parameters of main layers are as shown in Table 2. It should be emphasized that Si material was selected as the dielectric layer and its imaginary part of RI was ignored.

Table 2 The parameters of different layers for calculating angular spectra

| Structures               | Main layers                  | Parameters          |
|--------------------------|------------------------------|---------------------|
| Silver/BK7               | Silver film                  | 55 nm               |
| Graphene/silver/BK7      | Silver film                  | 60 nm               |
|                          | Graphene                     | 12 layers           |
| Dielectric layer         | Silver film                  | 55 nm               |
| (Si)/silver/BK7          | Dielectric layer (Si)        | 3.846 RIU, 5 nm     |
|                          | Silver film                  | 55 nm               |
| Graphene/Si/silver/BK7   | Dielectric layer (Si)        | 3.846 RIU, 5 nm     |
|                          | Graphene                     | 3 layers            |

Fig. 5(c) and 5(d) show the resonance angle and RI sensitivity for the four typical sensing structures at different RI of sensing medium. The RI sensitivity is also calculated when RI difference of sensing medium is 0.005 RIU. The sensing range is also an important performance evaluation parameter for the application of SPR sensor. The whole sensing range combined with linear and nonlinear intervals can be ordered by silver/BK7 > graphene/silver/BK7 or Si/silver/BK7 > graphene/Si/silver/BK7 as shown in Fig. 5(c). Fig. 5(d) also obviously indicates that the linear sensing range decreases when the silver film was protected with dielectric layer or graphene.

Fig. 6(a) shows that the RI sensitivity is 116 deg/RIU for conventional monolayer silver based SPR sensor in water environment. Fig. 6(b) demonstrates that the RI sensitivity would be improved 22.4% that is 142 deg/RIU, when introducing a 12-layer graphene on the silver film. It is demonstrated that the RI sensitivity could reach 206
deg/RIU when the thickness of Si protected layer is 5 nm, as shown in Fig. 6(c). Further, as shown in Fig. 6(d), the RI sensitivity as high as 240 deg/RIU was obtained for the graphene/Si/silver/BK7 based SPR sensor.

**Fig. 6** Four typical silver based SPR sensing structures with their angular spectra and RI sensitivity simulated by Fresnel calculations. (a) Silver/BK7 structure, (b) graphene/silver/BK7 structure, (c) Si/silver/BK7 structure, (d) graphene/Si/silver/BK7 structure

Fig. 7 shows the electric filed distributions for four typical sensing structures. It is obviously that their electric field distributions are not continuous at every interface since the SPR phenomenon is excited under TM mode. The field intensity at Cr/BK7 interface was used as a reference filed intensity for calculating field enhancement factor. As shown in Fig. 7, the electric field intensity is nearly exponential attenuation in the sensing medium. It is also demonstrated that the existence of dielectric layer or graphene would significantly decrease the electric field intensity in the silver film and sensing medium. Especially, the graphene has a greater impact than the dielectric layer and it might be ascribed to its imaginary part of RI.
**Fig. 7** Electric field enhancement distributions for the four typical silver based SPR sensing structures. (a) Silver/BK7 structure, (b) graphene/silver/BK7 structure, (c) Si/silver/BK7 structure, (d) graphene/Si/silver/BK7 structure

**Application of Biosensing with BSA Protein**

The biosensing application of optimized composite silver film based SPR sensor was carried out based on Fresnel reflection theory for BSA protein. Assuming adsorbed BSA molecules from sensing medium formed a monolayer on sensing surface as shown in insets of Fig. 8(a) and 8(b). Fig. 8(a) and 8(b) also show that the resonance angle is red shift with the increase of fBSA for both silver/BK7 and graphene/dielectric/silver/BK7 based SPR sensors. Fig. 8(c) presents shifts of the resonance angle vs fBSA for the typical four silver based SPR sensors. It is obviously that their detection sensitivities have an order of graphene/dielectric/silver/BK7 > dielectric/silver/BK7 > graphene/silver/BK7 > silver/BK7. The shift of resonance angle for graphene/dielectric/silver/BK7 SPR sensor is 1.2, 2.1, and 3.1 folds larger than those of dielectric/silver/BK7, graphene/silver/BK7 and silver/BK7 SPR sensors, respectively.
Fig. 8 Variation of reflection spectra with respect to the surface coverage of BSA molecules (fBSA) adsorbed on (a) the silver film (silver/BK7 structure) and (b) the graphene surface (graphene/dielectric/silver/BK7 structure), (c) shifts of the resonance angle vs fBSA for the typical four silver based SPR sensors

Conclusion
In this paper, we comprehensively discussed the influence of dielectric layer and graphene on silver based SPR sensor, the former factors are its refractive index and thickness and the latter is its number of layers. Graphene is a special two-dimensional nanomaterial, and thus the results are suitable for other two-dimensional nanomaterials, such as TMDCs and MXenes. The RI sensitivity could be obviously improved via introducing dielectric layer or graphene, while the other evaluation indexes, such as figure of merit, penetration depth, electric field enhancement and sensing range, are weakened. The RI sensitivity climbs up and then declines with respect to the number of graphene layer or RI and thickness of dielectric layer. The RI sensitivities for composite silver film based SPR sensor with monolayer silver, graphene/silver, dielectric/silver and graphene/dielectric/silver could reach about 116, 150, 450 and 350
deg/RIU, respectively. Further, graphene is more suitable as a protective or adsorbed layer and the optimized dielectric layer has a more obvious improvement on the RI sensitivity. The shift of resonance angle for graphene/dielectric/silver/BK7 SPR sensor is 1.2, 2.1, and 3.1 folds than those of dielectric/silver/BK7, graphene/silver/BK7 and silver/BK7 SPR sensors, respectively.

**Author Contribution** G.W. contributed to this work in the modeling, calculations, and data analyzing. G.W. and L.H. contributed to this work in the data fitting. G.W. and L.H. prepared the paper. All authors read and approved the contents.

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**Data Availability** The datasets used and/or analyzed during the current study are available from the corresponding author on reasonable request.

**Declarations**

**Ethics Approval** This is a theoretical study, and no ethical approval is required.

**Consent to Participate** Informed consent was obtained from all individual participants included in the study.

**Consent for Publication** The work described has not been published before. The work is not under consideration for publication elsewhere. Its publication has been approved by all co-authors.

**Competing Interests** The authors declare no competing interests.

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