Introducing S-RILS and D-RILS as refractive index limited optical biosensors in multiple nanolayers

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Abstract

The amplitude and phase sensitivities of a sensor with a multiple nanolayers consisting of gold-silicon dioxide-gold (Au-SiO₂-Au) are investigated. The proposed sensor has the ability to limit to the certain regions of the sensing medium refractive index \( n_f \). The number of these regions can be increased by increasing the thickness of the silicon dioxide layer (d_{SiO₂}). The results show that for the d_{SiO₂} = 200 nm the sensitivity has a nearly Gaussian in shape which its position depends on the refractive index of the prism \( n_p \). This type of sensor is introduced as a singlet refractive index limited sensor (S-RILS) that is useful tool in the detection of the target sample optical properties with certain refractive indices. It is found that two nearly Gaussian curves are generated in the amplitude and phase sensitivities for the d_{SiO₂} = 450 nm, which is introduced as a doublet refractive index limited sensor (D-RILS). The optical properties of the mixture samples can be detected by D-RILS, in while the S-RILS is used to detect the optical properties of single samples. The efficiency of the sensors can be improved by adding the titanium or titanium-graphene layers. The results exhibit that the optimal amplitude and phase sensitivities occur for multiple nanolayers (Ti-Au-SiO₂-Ti-Au).

1. Introduction

Surface plasmon resonance (SPR) is a physical phenomenon which is generated by the interaction of P-polarized light with the interface of two environments with different optical properties [1–8]. The SPR sensors with a single resonance peak usually consist of three dielectric-metal-dielectric layers [9–15]. The multiple structures of metal-dielectric can excite the multiple resonance peaks [16–20].

Sensors with multiple structures have many applications for measurement of many parameters in many scientific researches in the field of identification of molecular information, selective optical filters, biochemical sensing applications and multiplexing detector arrays [18, 21–23]. The geometric structure and dielectric function of the multiplex arrays play an important role in the amount of sensitivity of the designed sensor [22, 24, 25]. Chen et al. have studied the excitation of multiple resonance modes for a 3D graphene metamaterial structure in the Terahertz region. They showed that the resonance modes depended on the geometric parameters[22]. Similarly, Tang et al. have investigated the THz modulator based on the symmetrical and asymmetric split ring structure which has been made of the graphene[26]. Their results are done based on intensity detection. The method of intensity detection in multiple sensors has low accuracy for the weight of the biomolecules less than 500 Da [1, 27–29]. Also, regulation ability has limited for this type of detection in these sensors. In order to increase biosensor performance with multiple resonance methods, the phase detection techniques are used. The theoretical studies have shown which the phase detection method has a sensitivity of around 10²–10⁶ orders more than the amplitude detection method in the refractive index measurement of sensing medium [30, 31]. So far several methods have been employed to monitor the phase measurements of SPR, such as shear interferometry, polarimetry, heterodyne detection, temporal and spatial phase modulation interferometry [32–42].
In the proposed method, a multilayer structure consisting of gold-silicon dioxide-gold is utilized. Throughout simulation using the angle scanning method, the amplitude detection and the phase detection information are extracted for sensing medium that its refractive index varies from 1 to 1.833 in steps of 0.005 at the wavelength of 850 nm. The results show that the width of the obtained peaks is strongly dependent on geometrical dimensions of the sensor and the materials type. Therefore, the proper dimensions of sensor layers are optimized to achieve better sensitivity. The results show that for 200 nm silicon dioxide layer, a single peak is appeared in the amplitude and phase sensitivity diagrams, which reaches the maximum over the range $\Delta n_s = 1.47 - 1.65$. With increasing the silicon dioxide thickness to 450 nm, the amplitude and phase sensitivities produce two separate peaks in two different ranges of the refractive index. It is found that the phase sensitivity diagram has the drastic changes with respect to the refractive index variations of the sensing medium compared to the amplitude sensitivity diagram. Therefore, the use of phase data can increase the measurement accuracy of biological samples. Also, the results show the sensitivity range is similar in both amplitude and phase methods, and maximum sensitivity of two peaks obtain in the range $\Delta n_{s1} = 1.06 - 1.36$ and $\Delta n_{s2} = 1.41 - 1.54$ respectively.

### 2. Sensor structure

Figure 1 shows an outline of the proposed biosensor containing titanium-gold-silicon dioxide-titanium-gold-graphene. By considering the kretschmann structure, the thickness of gold and silicon dioxide layers has been optimized for different structures. To evaluate the optical response of the proposed biosensor, we used the multilayer transfer matrix method using MATLAB software [43]. The dielectric constants of material are given in table 1 at wavelength of 850 nm. The structure A called as the reduced structure comprises from gold-silicon dioxide-gold. The structure B is obtained by adding a multi-nanometer titanium layer to the previous structure to enhance the adhesion strength of the gold layer to the substrate [44–47].

Finally, in structure C for increasing the molecular adsorption to the sensor surface [48–63], five-layer graphene with the 1.7 nm thickness was added to the structure B between the top gold and the sensing medium.

### Table 1. Dielectric constants of materials.

| Medium   | Dielectric constant ($\epsilon$) | Refractive index ($n$) |
|----------|---------------------------------|------------------------|
| Prism    | 2.9206                          | 1.7090                 |
| Titanium | $-5.4018 + 125.874$             | 3.2427 + i3.9896       |
| Gold     | $-28.269 + 11.7456$             | 0.16408 + i5.3194      |
| Silica   | 2.1097                          | 1.4523                 |
| Graphene | $6.6191 + 9.258$               | 3 + i1.5430            |

In the proposed method, a multilayer structure consisting of gold-silicon dioxide-gold is utilized. Throughout simulation using the angle scanning method, the amplitude detection and the phase detection information are extracted for sensing medium that its refractive index varies from 1 to 1.833 in steps of 0.005 at the wavelength of 850 nm. The results show that the width of the obtained peaks is strongly dependent on geometrical dimensions of the sensor and the materials type. Therefore, the proper dimensions of sensor layers are optimized to achieve better sensitivity. The results show that for 200 nm silicon dioxide layer, a single peak is appeared in the amplitude and phase sensitivity diagrams, which reaches the maximum over the range $\Delta n_s = 1.47 - 1.65$. With increasing the silicon dioxide thickness to 450 nm, the amplitude and phase sensitivities produce two separate peaks in two different ranges of the refractive index. It is found that the phase sensitivity diagram has the drastic changes with respect to the refractive index variations of the sensing medium compared to the amplitude sensitivity diagram. Therefore, the use of phase data can increase the measurement accuracy of biological samples. Also, the results show the sensitivity range is similar in both amplitude and phase methods, and maximum sensitivity of two peaks obtain in the range $\Delta n_{s1} = 1.06 - 1.36$ and $\Delta n_{s2} = 1.41 - 1.54$ respectively.
The optimum thicknesses of gold, silicon dioxide and titanium layers were obtained for three structures. The graphene layer partially modifies the optimum thicknesses values of the gold, silicon dioxide and titanium layers in structure B which can be neglected, but it affects the amplitude and phase sensitivity diagrams. We introduce the amplitude and phase sensitivity as follows:

\[
S_l = \max \left\{ \left| \frac{R_{n_l+\Delta n_s}(\theta) - R_{n_l}(\theta)}{\Delta n_s} \right| \right\},
\]

\[
S_\varphi = \max \left\{ \left| \frac{\varphi_{n_l+\Delta n_s}(\theta) - \varphi_{n_l}(\theta)}{\Delta n_s} \right| \right\}. 
\]

Also, for a better understanding of the phase detection, we use the phase difference between the sensing medium and the blank sample which is defined as follows [40]:

\[
\Delta \varphi = \max (|\varphi_{n_l}(\theta) - \varphi_{n_b}(\theta)|),
\]

Here, \(n_s\) changes from 1 to 1.833 in steps \(\Delta n_s = 0.005\) and \(n_b\) denotes the refractive index of blank sample and the air is chosen as blank. \(R_s(\theta)\) and \(\varphi_s(\theta)\) stand for the reflection and phase spectrum in terms of incident angle \(\theta\) at the refractive index \(n\).

3. Results and discussion

3.1. Investigation of the effect of silicon dioxide layer thickness on the sensor:

The simulation results show that for a fixed gold thickness (\(d_g = 50\) nm) in the structure A, the amplitude spectrum and phase spectrum of the proposed sensor undergo drastic changes as the silicon dioxide layer thickness varies. For a more detailed analysis of this phenomenon, the spacer layer thickness \(d_{SiO_2}\) was swept in the range of 200 to 450 nm with step 50 nm (figure 2(a)). As is seen in the figure, the obtained spectrum for the thicknesses below 200 nm have two separate dips (data not shown) that it was optimized for \(d_{SiO_2} = 200\) nm. Also, for the thickness in the range of 250 to 450 nm, the reflection spectrum has three separate dips and its optimum occurs in the thickness of 450 nm. Reflection spectra diagram was plotted for all three structures at optimum thicknesses of \(d_{SiO_2} = 200\) nm and 450 nm (figures 2(b) and (c)). As can be observed, the titanium layer reduces the depth of the resonant dips. To compensate this effect, the gold thickness has been optimized in the presence of titanium (further details in the next sections). The simulation studies show that by adding the graphene layer the changes in reflection spectrum is very negligible. The extracted sensitivity diagram of the sensor with \(d_{SiO_2} = 200\) nm which has one single local maximum and the sensor with \(d_{SiO_2} = 450\) nm which has two local maxima, are called as case (I) and case (II) sensors, respectively.

3.2. S-RILS sensor optimization:

To obtain the amplitude and phase sensitivity in structures A, B and C for \(d_{SiO_2} = 200\) nm, we have swept the gold thickness from 35 to 50 nm with step 5 nm. According to figure 3, the amplitude sensitivity diagrams for all three structures reach a maximum value only in a specified interval of the sensing medium refractive index. This result indicates that the proposed sensor detects the optical properties of the sensing medium at a certain interval. So we introduce this sensor as a singlet refractive index limited sensor (S-RILS). One of the important applications of S-RILS is the characterization of biological samples in a specific refractive index range. The maximum position of the amplitude and phase spectra can also be changed by manipulating the refractive index of substrate \(n_g\). By selecting the substrates with refractive indices of 1.5098 (BK7), 1.6567 (SF5), 1.7090 (SF10)
and 1.8301 (LASF9) for the structure C, the maximum position of the amplitude spectra shifts to 1.43, 1.50, 1.50 and 1.51 respectively (red curves in figures 3, 4). Therefore, the optical information of the target sample can be revealed by choosing appropriate substrate.

Also as shown in figure 3, adding the titanium and graphene layers can slightly decrease the peak height of the amplitude sensitivity diagram (SI). In figure 5, the phase sensitivity (Sj) versus the sensing medium refractive index diagram has been extracted for the three structures A-C over the range 1-1.8 in steps 0.005. It is seen that for structure A, the Sj sensitivity diagram has three peaks for the 35 nm gold thickness which were represented by symbols I, II and III. With the increase of the gold layer thickness, the position of the peaks shifts and their numbers decrease. In figure 5 for structure B by adding of the titanium layer, the Sj diagram is evolved and the sensitive range to the RILS refractive index reduces. Also the number of peaks in the phase sensitivity diagram decreases as titanium layer is added to structure A. By adding titanium and graphene layers simultaneously an important evolution in the Sj spectrum occurs.

According to figure 5 for structure C, the sensitive range to the singlet refractive index in S-RILS decreases with respect to the two previous structures and finally reduces to a single Gaussian peak. Owing to the amplitude sensitivity diagrams in figure 3, the structures A-C in the phase sensitivity diagrams that have a similar Gaussian-shaped spectrum in thicknesses of dg = 35 nm, dg = 40 nm and dg = 50 nm, respectively, are introduced as optimized S-RILS. The variations of the sensor phase with respect to the blank sample Δϕmax has been extracted in figure 5. It is seen that by adding titanium or titanium-graphene, similar changes to Sj diagrams is detected.

### 3.3. D-RILS sensor optimization:

The amplitude and phase sensitivity spectra for all three structures A, B and C in the proposed sensor show two separate peaks as the silicon dioxide layer thickness is increased to 450 nm. Therefore, the sensor limited to the refractive index reveals the optical properties of the sensing medium at two different refractive index intervals. To optimize this sensor, as in the previous case, the gold thickness was swept in the range of dg = 35 nm to dg = 50 nm. In figure 6, the amplitude sensitivity spectra were plotted for all three structures A-C in different thicknesses of the gold layer. It is also observed, by adding titanium or titanium-graphene layers the sensitivity spectrum is similar to structure A.

The phase sensitivity spectrum was plotted for case (II) sensor in different conditions in figure 7. The results show that the number of peaks in Sj diagram decreases by adding titanium or titanium-graphene layers to
structure A. The number of phase jump has also decreased in the $\Delta \phi_{\text{max}}$ sensitivity spectrum (see figure 7 for $\Delta \phi_{\text{max}}$).

In the final optimized sensor containing titanium layer, the $\Delta \phi_{\text{max}}$ and $S_j$ evolution in terms of the sensing medium refractive index have two Gaussian-shaped peaks for the optimum thickness of $d_g = 40$ nm in the refractive index ranges of $\Delta n_1 = 1.06 - 1.36$ and $\Delta n_2 = 1.41 - 1.54$ (see figure 7 for B structure). The important application of the doublet refractive index limited sensor (D-RILS) is to detect and characterise the mixed biosamples and the optical information separation of target sample. The proposed sensing structure of D-RILS has the ability to separate two refractive indices in dichroic materials or in polarization-sensitive mixed materials. Therefore, by using polarimetry technique, the D-RILS can be applied as a powerful mechanism in separate detection of biomaterials.

In figure 8, the $S_I, S_j$, and $\Delta \phi_{\text{max}}$ sensitivity diagrams were plotted for all three structures A-C with the optimized gold thicknesses of 50 nm, 40 nm and 40 nm, respectively. Owing to the height of peaks in figure 8(a) show that the $S_I$ sensitivity for structures A and B are better than structure C. Further comparison exhibits that structure B has a higher separation power with respect to structure A due to the dip value (see points $P_1$ and $P_2$ in 8(a)). Also the height of the peak for structure B in both the $S_I$ and $\Delta \phi_{\text{max}}$ sensitivities reaches its maximum value compared to two other structures for the range $\Delta n_{12}$. Adding the graphene layer results in the decrease of the peaks height and behaves similar to structure A. Therefore, the B-type structure will be introduced as the most optimum sensor of D-RILS, providing the lack of use graphene.
4. Conclusion

By using transfer matrix method in MATLAB and by choosing a TM incident light at the wavelength of 850 nm, the amplitude and phase sensitivities have been investigated in a multiple nanolayers consisting of gold-silicon dioxide-gold. Functionality of the obtained sensitivities diagrams show a drastic dependence on the thickness of the middle layer of silicon dioxide. The results indicate that by choosing \( d_{SiO_2} = 200 \) nm, the S-RILS is restricted to one region of the sensing medium refractive index, so that \( S_n, S_j \), and \( \Delta \phi_{\text{max}} \) diagrams are only maximized in the range of \( n_s = 1.4 - 1.6 \). This type of sensor is used to detect samples that lie in the interval of the above refractive index, whereas in conventional monolayer sensors, the sample detection is performed over a large refractive index range. Therefore, S-RILS can be used to extract the optical information of the target sample within a certain range of refractive index. With increasing the thickness of the silicon dioxide layer to 450 nm, the amplitude and phase sensitivities of the D-RILS sensor are maximized in two different intervals of refractive indices \( n_s = 1.06 - 1.36 \) and \( n_s = 1.41 - 1.54 \) which will be able to detect the optical properties of the mixed samples. The advantage of the D-RILS sensor with respect to the S-RILS sensor is that it can simultaneously detect the optical information of the compounds existing on the mixed sample. Therefore, D-RILS is introduced as a separator detector of physical properties in mixed samples.

The performance of the above sensors can be increased by adding titanium or titanium-graphene layers. Adding a titanium layer (structure B) causes the increase of the gold layers adhesion to the substrate, adding the titanium-graphene layer (structure C) as well, the reactivity strength of biological materials with the sensor surface increases due to graphene. The gold thickness was optimized in the presence of titanium or titanium-
graphene layers in order to improve the amplitude and phase sensitivities of the sensor. The optimum values of the gold thicknesses for structures A, B and C were obtained 50, 40 and 40 nm, respectively. The peaks height in the sensitivity diagrams of structure B reaches its maximum value compared to the other structures A and C.

According to schematic diagram of multiple plasmon resonance biosensor (figure 1), the reflected electromagnetic fields from two gold layers can interfere with each other. Therefore, by varying the distance of these two layers (thickness of the silicon dioxide), constructive and destructive interference produces in the reflected light. As shown in figures 3 and 5 for the $d_{SiO_2} = 200 \text{ nm}$ a constructive interference is generated, consequently a single peak is observed in amplitude and phase sensitivities. As a result of destructive interference in the $d_{SiO_2} = 450 \text{ nm}$, a double peak occurs at the amplitude and phase sensitivities in figures 6 and 7. This physical effect in the proposed sensor works based on Fabry–Perot interferometry.

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