Theoretical study of modulated multi-layer SPR device for improved refractive index sensing

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Abstract. In the present work, a theoretical investigation of Surface Plasmon Resonance (SPR) properties of a multilayer film (Au-SiO₂-Au) coated on a glass prism is being carried out. In this multilayer structure, each interface corresponds to multiple SPR modes. To obtain the maximum reflection dips in the SPR modes, the thickness of SiO₂ layer is optimized by varying it from 100-600 nm. Our calculation also reveals that SPR mode corresponding to Au-ambient interface is very sensitive to the changes in the surrounding medium, least affecting other SPR modes. The sensing performance of the proposed nano-plasmonic sensor is theoretically calculated using bulk refractive index sensing. Such multilayer SPR sensing device has advantages over conventional SPR devices in terms of their bulk sensitivity and self-referencing, claiming itself as a potential candidate for the development of highly sensitive biological sensor.

1. Introduction
Over the last thirty years, Surface Plasmon Resonance (SPR) is becoming an increasingly important phenomenon as it has been found that label-free detection technique is suitable and reliable for clinical analysis and bio-molecular interactions [1-3]. Cost-effectiveness, high sensitivity, and possibility in-situ real time measurement are main characteristics of SPR sensing [4]. Among all the SPR techniques, prism coupling technique is the most commonly used method for the excitation of surface plasmons (SPs) [5]. SPs are collective oscillations of free electrons in the surface of metallic films such as gold, silver, and aluminum. Gold is utilized as the metal layer in most cases because it is highly chemically stable, inert in nature and higher sensing resolution at visible wavelength range as compared to other metals (like silver and aluminium etc.). The sensitivity of the SPR device depends on the optimization of metal thickness and utilization of multi-layer of alternative metal and dielectric layer [6-9]. In this work, we demonstrate a novel SPR sensor with SiO₂ layer sandwiched in between two gold layers of thickness 50 nm. Here, we optimized the thickness of gold layer as well as the SiO₂ layer by varying the thickness. It is observed that, the multi-layer structure has higher sensitivity as compared to the traditional single layer SPR sensor and a broad detecting range of refractive index, which could be used in biochemical analysis, disease diagnosis and monitoring.

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2. Theory

The reflectance of a multi-layered structure can be measured by using transfer matrix method for N-layer model to obtain the expression for Fresnel reflection coefficient for p-polarised incident light. As shown in the generalized equation for the \( k \)'th layer of a multi-layer structure, in the arbitrary medium layer is denoted by dielectric constant \( \varepsilon_k \), thickness \( d_k \), refractive index \( n_k \) and permeability \( \mu_k \). The tangential fields at the first boundary \( Z=Z_1=0 \) are related to those at the final boundary \( Z=Z_{N-1} \) as given by \([10,11]\)

\[
M = \sum_{k=2}^{N-1} M_k = \begin{bmatrix} M_{11} & M_{12} \\ M_{21} & M_{22} \end{bmatrix}
\]

(1)

with

\[
M_k = \begin{bmatrix} \cos \beta_k & -i \sin \beta_k \\ -iq_k \sin \beta_k & \cos \beta_k \end{bmatrix}
\]

(2)

where \( \beta_k \) is the phase factor given by

\[
\beta_k = \frac{2\pi d_k}{\lambda} \left( \varepsilon_k - n_k^2 \sin^2 \theta \right)^{\frac{1}{2}}
\]

(3)

and

\[
q_k = \frac{\left( \varepsilon_k - n_k^2 \sin^2 \theta \right)^{\frac{1}{2}}}{\varepsilon_k}
\]

(4)

The reflection coefficient for a transverse magnetic wave is given as,

\[
r_p = \frac{(M_{11} + M_{12}q_k)q_1 - (M_{21} + M_{22}q_k)}{(M_{11} + M_{12}q_k)q_1 + (M_{21} + M_{22}q_k)}
\]

(5)

and reflectance for p-polarised light is given by,

\[
R_p = |r|^2
\]

(6)

3. Results and Discussions

For the present study, freely available MATLAB code with slight modification was used to simulate angle dependent SPR spectra for single layer metal (gold) and multiple layers in configuration of metal- dielectric- metal (MDM, gold-SiO\(_2\)-gold) placed on the top of a prism (refractive index \( n=1.5 \)) [12]. These structures are schematically depicted in Figure 1(a) and (b).

![FIGURE 1](image-url)

FIGURE 1. Schematic diagram of (a) Single layer SPR device and (b) Multi-layer SPR device (gold-SiO\(_2\)-gold).
In this context, the main aim was to optimize the thickness of the gold layer for a single layer system and then we have to compare its sensitivity with multilayer of MDM. At first, the SPR curves at 670 nm wavelengths were simulated for varying the gold thickness from 10 nm to 60 nm with step size of 10 nm and the prism was coated with gold with the surrounding medium being air (refractive index n=1).

**FIGURE 2.** (a) Optimization of thickness of a single layer SPR device of gold at prism material, (b) SPR reflectance spectra of the single layer gold with changing various analytes having different refractive index from 1.33-1.38 and (c) Reflectance dip shift of angle as a function of refractive index.

In Figure 2(a), it is found that the resonance angles of the SPR curves for different metal thickness as well as the contrast of reflected intensities are varying. The optimum thickness is considered to be the one which has the minimum contrast, or the point where the reflectance curves tend towards zero at resonance condition. It is utmost clearly visible that 50 nm thickness of gold is the best calculated optimum thickness for resonance condition. Figure 2(b) shows the SPR spectra of a single gold layer of 50 nm thickness for the detection of analytes with Refractive Indices (RI) ranging from 1.33 to 1.38. The SPR dip is red shifting depending on the increase in RI of analytes. The refractive indices (1.33-1.38) led to shift of the SPR dip shown in the spectra at the corresponding angles. However, the calculated bulk refractive index sensitivity was found to be 174 degree/RIU (Refractive Index Unit) which was obtained from the linear fitting of plot between angular dip shifts versus refractive index [shown in Figure. 2(c)].

Later, we introduce a multilayer SPR device of dielectric (SiO$_2$, relative permittivity ($\varepsilon_r$) = 3.9) layer in between the two gold coated layers on the prism to study its sensitivity over a conventional single layer device. In Figure. 3(a) the corresponding SPR spectra observed with two SPR dips in reflectance spectra. Apparently, these two SPR dips observed due to the excitation of free electrons (surface plasmons) at two interfaces, one being the gold-SiO$_2$ interface and the other is due to gold-air interface. We have also plotted the SPR curves by varying the SiO$_2$ thickness from 100 nm to 600 nm with step size of 50 nm in the same figure, with a fix gold layer thickness of 50 nm as optimized above. Further, 400 nm thickness of the SiO$_2$ layer corresponds to the minimum reflectance dip as found from Figure. 3(a) and more sharper having minimum full width half maximum (FWHM). Hence, this is considered as the optimum thickness. In Figure. 3(b), we have plotted the SPR spectra for varying analytes refractive index from 1.33-1.38 on the optimum gold-SiO$_2$-gold structure on prism. It is clearly observed that with the increase in refractive index of analytes, both the reflectance dips show red shifts. However, the total shifts in the first SPR dip which is of 0.10 degree is very less as compared to the second SPR dip which is total shifted 11 degree angular shift and is shown in Figure. 3(c). For self-referencing purpose, the first angular shift is kept fixed and we consider the shifting of the second SPR dip. Further, the calculated bulk refractive index sensitivity of the second SPR dip was found to be 211 degree/RIU (Refractive Index Unit) which was obtained from linear
fitting of the plot between SPR dip shifts versus refractive index in case of multilayer device, as shown in Figure. 3(d).

FIGURE 3. (a) SPR reflectance spectra for varying thickness of SiO$_2$ sandwiched between two 50 nm of gold layer on prism material. This spectra is used for optimization of SiO$_2$ thickness that is 400 nm, (b) SPR reflectance spectra of the multi-layer (gold-SiO$_2$-gold: 50 nm-400 nm-50 nm) with changing various analytes having different refractive index from 1.33-1.38 with self-referencing mode at 49 degree, (c) dip shifting response curve for two dips with respect to change of refractive index of analytes and (d) SPR dip shift of angle as a function of refractive index of a multi-layer SPR device.

TABLE 1. Calculation of SPR dip shift in angle versus the refractive index of analytes with single and multi-layer structure.

| Layer                                      | Refractive Index |
|--------------------------------------------|------------------|
|                                            | 1.33  | 1.34  | 1.35  | 1.36  | 1.37  | 1.38  |
| Single Layer (50 nm gold)                  | 71°    | 72°    | 74°    | 75°    | 77°    | 80°    |
| Multi-layer (50 nm gold-400 nmSiO$_2$-50 nm gold) | 74°    | 75°    | 77°    | 80°    | 82°    | 84°    |
The SPR dip in angle corresponding to the refractive index of analytes for both single layer and multi-layer structure is shown in Table 1. It is clear from the table that, the SPR dip shift in angle with multi-layer structure is comparably more than the single layer structure.

4. Conclusion
In this work, we have reported numerical tests about the sensitivity of plasmonic sensors. The multi-layer modulation technique can have enhanced sensitivity as compared to the conventional single layer device. Globally, utilization of this technique can help to fabricate a label free biological sensor with the benefits of self-referencing.

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