JOURNAL OF NANO- AND ELECTRONIC PHYSICS
Vol. 8 No 4(2), 04085(5pp) (2016)
ЖУРНАЛ НАНО- ТА ЕЛЕКТРОННОЇ ФІЗИКИ
Том 8 № 4(2), 04085(5cc) (2016)

Enhanced Sensitivity of Surface Plasmon Resonance Sensor Based on Bilayers of Silver-Barium Titanate

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(Received 22 September 2016; published online 23 December 2016)

Surface plasmon resonance (SPR) sensors have been widely adopted with various fields such as physics, chemistry, biology and biochemistry. SPR sensor has many advantages like the less number of sensing samples required, freedom of electromagnetic interference and higher sensitivity. This research investigates the phase interrogation technique of a surface plasmon resonance sensor based on silver and thin film dielectric material of Barium titanate layers. Barium titanate ($BaTiO_3$) layer is adopted due to its excellent dielectric properties such as high dielectric constant and low dielectric loss. The numerical results demonstrate that the fusion of the proposed material $BaTiO_3$ layer into surface plasmon resonance sensor yields a higher sensitivity of 280 degree/RIU in comparison with surface plasmon resonance sensor without $BaTiO_3$ layer which shows only a sensitivity of 120 degree/RIU. As the thickness of this layer increases from 5 nm to 10 nm, the sensitivity is enhanced from 160 degree/RIU to 280 degree/RIU for a fixed metal layer of silver with a thickness of (70 nm).

Keywords: Surface plasmon resonance sensor, Phase interrogation, $BaTiO_3$, Ag.

DOI: 10.21272/jnep.8(4(2)).04085 PACS number: 73.20.Mf

1. INTRODUCTION

Surface plasmon is defined as oscillations of free electrons in the metal and when they are excited by optical radiation, they induce electromagnetic waves at the interface between the metal and dielectric media, which are called as surface plasmon waves. It can be excited by both *p*- and *s*-polarized electro-magnetic waves but no surface plasmon resonance would occur if excited only by *s*-polarized light. When excited by *p*-polarized light, at a suitable angle of incidence, the wave-vector of the incident light and wave vector of the surface plasmon waves would match, resulting in a resonance condition known as surface plasmon resonance (SPR) [1-4]. This phenomenon has been widely used as sensor technology, nonlinear optics, spectroscopy, optical modulators and microscopy, to name a few [1, 5-7].

During the last three decades, there has been an interest in the field surface plasmon resonance sensing technology due to its advantages like high sensitivity (it is a change of the reflected optical radiation intensity due to variations in the refractive index or the thickness of a sensing medium), real time detection with high accuracy [8, 4]. The sensitivity effects on the performance of SPR sensor because it demonstrates the SPR ability to detect the sample type and its concentration [9-11]. High sensitivity is attained by achieving a large shift of resonance angle in phase interrogation or resonance wavelength in wavelength interrogation with a small change of sample parameters such as refractive index, thickness and concentration [12-14]. In the last decade, Many studies have been carried out to improve the sensitivity of SPR sensor by utilizing an additional layer of semiconductor, dielectric or metamaterial. Sharmila et al. (2015) worked on analysis of the silicon layer to enhance the SPR sensor performance. They used an additional silicon nano-layer to increase the stability (it is a chemical stability of the metal film under extreme environment, which improves the SPR ability to obtain reliable optical signals and to perform long time measurements) and sensitivity (it is a change in the resonant angle due to change of the sample refractive index) of the SPR which enhances the evanescent field near the metal-sample interface in comparison with conventional SPR sensor [1].

Although Metamaterial have been exploited to enhance the detection accuracy of SPR sensor see, Sarika Pal et al. (2015), the sensitivity is still lower as compared to the SPR sensor based on an additional Tantalum Pentoxide (Ta_2O_5) dielectric layer [2]. Researches have focused on enhancing sensitivity of SPR sensor by using an oxides based on interrogation technique, as reported earlier by Sarika Singh et al (2013).

Beside sensitivity enhancement using this oxide layer, it is also used as a protective layer for the sensor from oxidation [3]. The semiconductor (Zno) thin layer was utilized as a protective layer and the performance of SPR sensor was improved as presented by Sarika Shukla et al (2015) [4].

In this paper, we numerically demonstrate the functionality of the SPR sensor by using a thin layer of $BaTio_3$ and demonstrate improved sensitivity to biosample as compared to conventional SPR sensor based on Ag layer.

2. BACKGROUND

SPR sensor is based on the excitation of the *p*-polarized light along the metal-dielectric interface and the principle is based on Attenuated Total Reflection

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(ATR) coupling method. Out of three different configurations, this work reports on two different configurations as illustrated in Fig. 1.

The first configuration is known as Kretschmann type in which case a thin metallic layer (denoted by *M*) with dielectric constant ε_2 is deposited directly on prism, the sensing medium which is a probe sample, and whose dielectric constant ε_1 is to be determined, is kept in contact with the metallic layer, *M*.

The second type is an Otto configuration in which the sensing medium is kept between the prism and thin film, and metal like gold (Au) or silver (Ag) for supporting the surface Plasmon wave. In the Otto configuration, is controlling the gap between metal and prism is cumbersome. Thus, Kretschmann configuration, shown in (a) is usually preferred used for bio-detection [15-16].

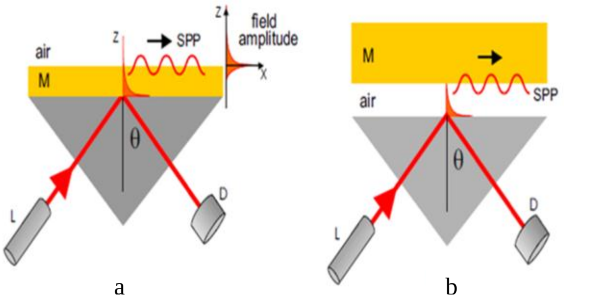


Fig. 1 – Surface plasmon sensor configuration (a) Kretschmann Type (b) Otto type. (L: Laser source, D: detector) [17]

As shown in Figure 2, when the *x*-component of light wave vector, in this case, $K \times \sin(\theta)$ matches to the wave vector of the surface plasmon wave, K_{SP} propagating along the metal-sensing medium interface, the resonance condition will be satisfied. In this geometry, light is incident on the prism-metal interface at a known incident angle. An evanescent wave propagating through the metal and excites surface plasmons on sensing medium. Depending on the probe material, the sensing medium can vary from water to liquid medium such as blood, urine, various protein, and DNA for example.

At resonance condition,

$$K_{x-component} = K_{sp} \quad (1)$$

Where *x*-component of the wave vector of incident light is $K_{x\text{-}component}$ and the surface Plasmon wave vector is K_{sp} and is given as:

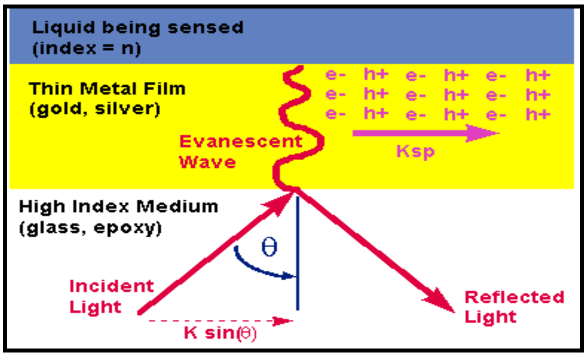


Fig. 2 – Schematic of surface plasmon resonance condition for SPR sensor [18]

$$K_{x-component} = K \times sin(\theta_{in}) = \\ = \frac{2\pi}{\lambda} n_p \times sin(\theta_{in}) \quad (2)$$

$$K_{sp} = \frac{2\pi}{\lambda} \times \sqrt{\frac{\varepsilon_1 \times \varepsilon_2}{\varepsilon_1 + \varepsilon_2}} \quad (3)$$

Where K is the complex-valued wave number, λ is the wavelength of the incident optical radiation/light, n_p is the refractive index of the prism, θ_{in} is the incident angle of the laser beam at the prism-metal interface and ε_1, ε_2 are the dielectric constants of the metal and the sensing medium respectively [5].

There are many parameters that determine the resonance condition such as wavelength of the incident light, incident angle, sensing layer thickness, and the refractive indices of both the metal as well as the sensing medium which is also a dielectric material. Under resonant this condition, the incident angle is known as resonant angle [4].

In actual measurement, the *p*-polarized light is incident on the prism-metal interface at a known incident angle and the light is reflected according to the total internal reflection theory. The reflected optical radiation is measured by using optical detector. In the phase interrogation mode, the reflected light is measured as a function of incident angle with at a fixed wavelength. In the Wavelength Interrogation mode, the reflected light is measured as a function of incident wavelength of the incident light with a fixed incident angle. A sharp dip in reflection is observed at the resonance angle in the Phase Interrogation mode or resonance wavelength in Wavelength Interrogation mode. The same measurement configuration can also be used to computer the refractive index of the probe material through varying resonance angle [15].

3. THEORETICAL MODEL FOR PROPOSED SURFACE PLASMON RESONANCE SENSOR

A schematic geometry of the proposed SPR Bilayers of Silver-Barium Titanate (Ag-$BaTiO_3$) sensor is shown in Fig. 3. It consists of; BK7 dielectric prism is used to achieve the resonance matching between the incident wave and surface plasmon wave where its refractive index depends on the wavelength of the incident light due to dispersion relation.

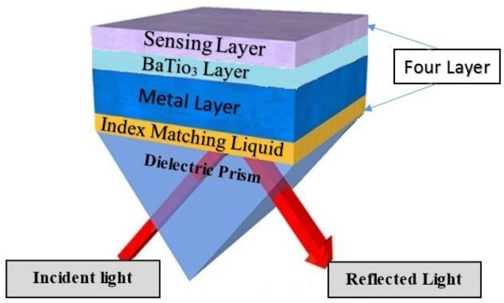


Fig. 3 – The proposed Ag-$BaTiO_3$ sensor configuration

Then, it is followed by an index matching liquid. A thin film metallic layer of Ag is deposited on it, for supporting the surface plasmon wave at the metal-dielectric interface whose dielectric constant also depends on the wavelength according to Drude model [19]. This metal layer is covered with $BaTiO_3$ dielectric layer to enhance the performance of the SPR sensor. The $BaTiO_3$ dielectric layer is made in contact with the sensing layer that has a refractive index in the range of 1.338-1.348.

To excite the surface plasmons in the metal layers, the laser source is utilized, and in this case, He-Ne laser with wavelength 632.8 nm is proposed. Any small change occurring within the sensing medium will cause large angular shift of the SPR curve. The sensitivity of the sensor is defined by the quantity of the reflected laser beam measured against the change in angle of the resonance peak incident angle. For a sensor with large sensitivity, a large change of the resonance angle at small change of refractive index is necessary and is defined by the equation as:

$$S = \frac{\Delta\theta_{res}}{\Delta n} = \frac{\theta_{res2} - \theta_{res1}}{n_2 - n_1} \tag{4}$$

Where $\Delta\theta_{res}$ is the change in the dip position of the reflected light at the resonance angle based on phase interrogation and Δn is the change of refractive index.

The minimum reflectivity of the SPR curve θ_{res} and λ_{res} based on the phase and wavelength interrogation respectively are illustrated in Fig. 4. The shift from λ_{res1} to λ_{res2}, or from θ_{res1} to θ_{res2} is based on the change in refractive index of the sensing medium induced using the phase and wavelength interrogation respectively. In order to study SPR phenomena, taking place within the SPR configuration, such as the Kretschmann configuration, three theoretical methods can be employed a) the field tracing method, b) the resultant wave method and c) the transfer matrix method. Among all three methods, the transfer matrix method is considered more accurate as it contains no approximations [20].

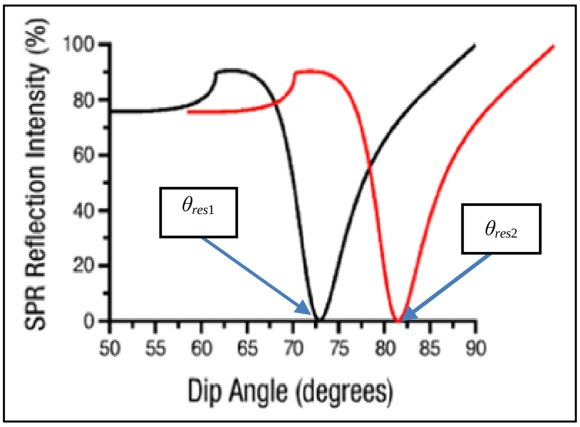


Fig. 4 – Surface plasmon resonance sensor curve [19]

For each layer, the phase shift and the admittance should be found in order to compute the transfer matrix and as follows:

$$\beta_j = \frac{2\pi}{\lambda} d_j \sqrt{n_j^2 - (n_p \sin(\theta_{in}))^2} \tag{5}$$

$$q_j = \frac{\sqrt{n_j^2 - (n_p \sin(\theta_{in}))^2}}{n_j^2} \tag{6}$$

Where q_j is the admittance, β_j is the phase shift, n_j is the refractive index and d_j is the thickness of layer j. The reflective index of the prism and the incident angle of the prism-metal interface are n_p and θ_{in} respectively [20].

Fig. 5 illustrates the structural form of the four layer surface plasmon sensor where several reflections resulting at each layer interface due to the incident light at prism-first layer interface, the accumulation of those reflections must be consider all for computing overall reflection/transmission computations. For a propagating wave through medium j towards medium $j+1$ is described by the transfer matrix as shown in Equation (7):

$$M_j = \begin{bmatrix} \cos(\beta_j) & -i\sin(\beta_j)/q_j \\ -iq_j\sin(\beta_j) & \cos(\beta_j) \end{bmatrix} \tag{7}$$

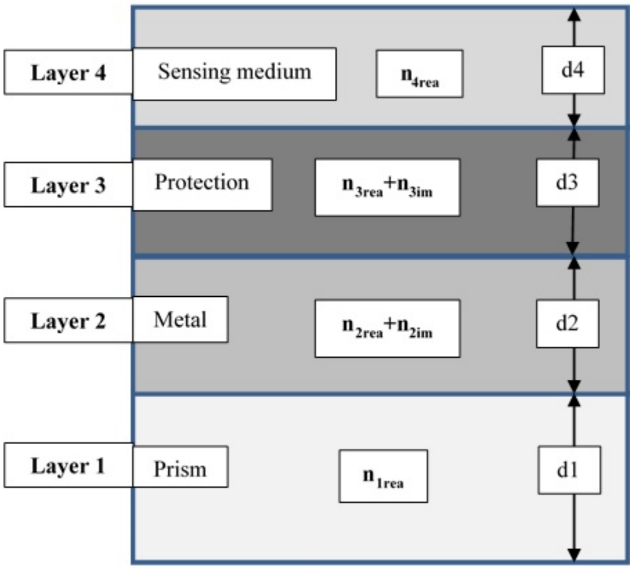


Fig. 5 – The proposed sensor layer architecture [19]

The overall transfer matrix of multilayer is calculated as a function of the transfer matrix M_j for each layer which described as:

$$M_{tot} = \prod_{j=2}^{m-1} \begin{bmatrix} m_{11} & m_{12} \\ m_{21} & m_{22} \end{bmatrix} \tag{8}$$

For a multilayer architecture consisted of m layers, the total reflection R and the transmission coefficients T are derived from:

$$R = \frac{(m_{11} + m_{12}q_m)q_1 - (m_{21} + m_{22}q_m)}{(m_{11} + m_{12}q_m)q_1 + (m_{21} + m_{22}q_m)} \tag{9}$$

$$T = \frac{2q_m}{(m_{11} + m_{12}q_m)q_1 + (m_{21} + m_{22}q_m)} \tag{10}$$

4. NUMERICAL RESULTS AND DISCUSSION

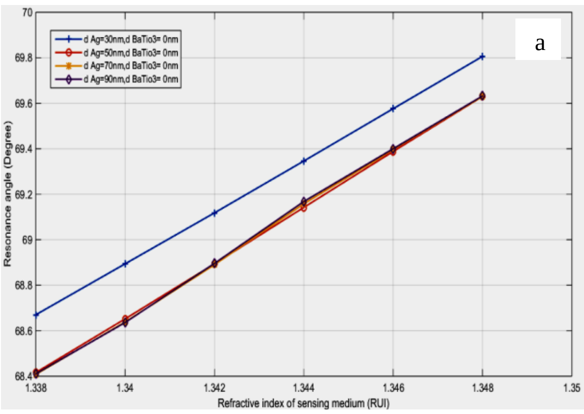
We have used a transfer matrix method to study the performance of the SPR sensor without using $BaTiO_3$ in compared with utilizing $BaTiO_3$ layer based on Matlab software. We have chosen the He-Ne laser with wavelength 633 nm as an excitation source, the optical properties of the layers as shown in Table 1, where the refractive index and extinction coefficients are depend on the operating wavelength due to the dispersion relation while the thickness of the layers has been selected for optimum performance. For a sensing medium, the refractive index various from 1.338 to 1.348 (in steps 0.002).

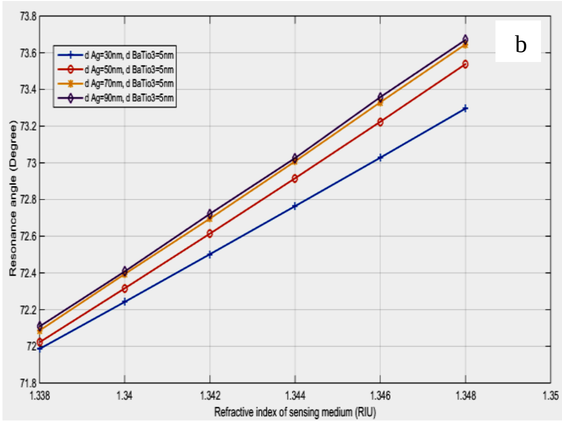
Fig. 6a shows that the resonance angles for each Ag layer thickness is raised proportionally to the value of the refractive index sensing medium. Refer to Equation 1, the increasing of the refractive index of the sensing medium for different thick Ag layers results in increasing of the real part of the wave vector of the surface plasmon wave K_{sp} which in turn the resonance angle at which the resonance condition is satisfied.

Table 1 – The optical properties of materials at 633 nm wavelength of light [21]

Material	Refractive index(n)	Extinction coefficient (k)	Thickness
BK7 prism	1.515	0	25 mm
Ag layer	0.056	4.276	30-90 nm
$BaTiO_3$ layer	2.4043	0	5-10 nm
Sensing medium	1.338-1.348	0	∞

On the other hand, the thickness of Ag layer also contributes to establishing the resonance condition at which the resonance angle is happening, because changing its thickness from 30 nm to 90 nm (in steps of 20 nm) will lead to a change of the dielectric constant of this layer. Fig. 6b and c illustrates the variation of the resonance angle with refractive index of the sensing medium for 30 nm, 50 nm, 70 nm and 90 nm Ag layers with preset 5 nm and 10 nm thick $BaTiO_3$ layer correspondingly. As well as, for each thickness of Ag and $BaTiO_3$ the resonance angle increases with increasing in refractive index of sensing medium. Whereas $BaTiO_3$ layer comes in contact with Ag layer as its thickness changes from 5 nm to 10 nm, the surface plasmon wave propagation real part constant of the wave vector is also changing and thus increasing the resonance angle of the proposed SPR sensor.





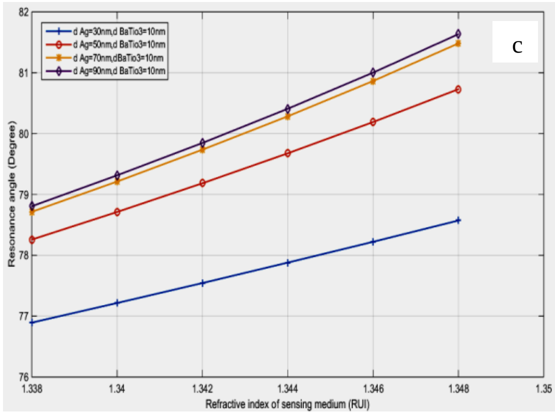


Fig. 6 – SPR senor's resonance angle disparities versus refractive index of sensing medium of various Ag layer thickness (30 nm, 50 nm, 70 nm and 90 nm) of (a) 0 nm, (b) 5 nm and (c) 10 nm thickness of $BaTiO_3$ layers

Fig. 7 shows the variations of sensitivity of the SPR sensor with a thickness of Ag layer for fixed 0 nm, 5 nm and 10 nm thick $BaTiO_3$ layer. It is obviously shown that the sensitivity of the sensor combining a $BaTiO_3$ layer is higher than without using $BaTiO_3$ layer due to the large shift of resonance angle within a small change of refractive index of sensing medium which causes higher sensitivity of the sensor. As the thickness of $BaTiO_3$ layer increases, a larger shift of resonance angle is resulting. Where the thickness of this layer is selected for achieving higher performance of SPR sensor.

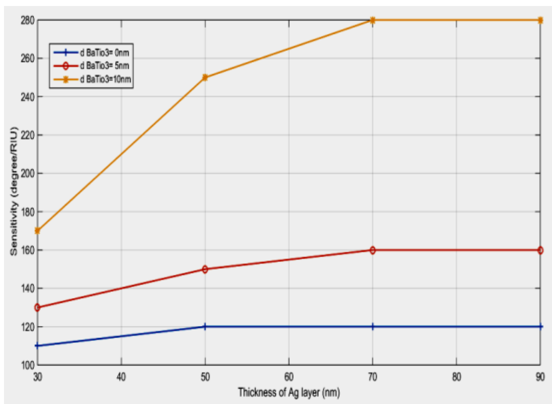


Fig. 7 – SPR sensor sensitivity variation against metallic layer (Silver) thickness for various $BaTiO_3$ thickness values (0 nm, 5 nm and 10 nm)

It can be viewed from Table 2 that the sensitivity of SPR sensor increases from 120 degree/RIU for Ag layer thickness of 70 nm and without using $BaTiO_3$ layer to a 280 degree/RIU for Ag layer thickness 70 nm, with a thin film of 10 nm $BaTiO_3$ layer.

Table 2 – The sensitivity of SPR sensor comparison for various thickness of Ag layer for fixed $BaTio_3$ layer thickness

Thickness of $BaTiO_3$ layer (nm)	Sensitivity (degree/RIU)			
	Thickness of silver layer (nm)			
	30	50	70	90
0 (without layer)	110	120	120	120
5	130	150	160	160
10	170	250	280	280

5. CONCLUSION

Theoretical and simulation investigations of the surface Plasmon resonance sensor with new proposed material $BaTiO_3$ added over Ag layer are conducted. The two layers of Ag-$BaTio_3$ have been simulated and analysis based interrogation technique. Besides that, $BaTiO_3$ is considered as a protection layer for preventing the metallic layer from oxidation, it increases the sensitivity of the SPR sensor. The sensitivity increases with an increase in the thickness of the $BaTiO_3$ layer for all thickness of Ag layer. The SPR sensor with two layers of 70 nm Ag-10 nm $BaTiO_3$ exhibits high sensitivity of 280 degree/RIU. Thin film $BaTio_3$ layer is used due to its high dielectric constant which realize large shift of resonance angle of the SPR curve within a small change of the refractive index sample, thus higher sensitivity can be achieved.

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