

Sensitivity enhancement of metamaterial-based surface plasmon resonance biosensor for near infrared

SARIKA PAL¹, Y.K. PRAJAPATI², J.P. SAINI³, VIVEK SINGH⁴

¹Department of Electronics and Communication Engineering, K.I.E.T Group of Institutions, Ghaziabad-201206 (U.P.), India; e-mail: narensarru@gmail.com

²Department of Electronics and Communication Engineering, Motilal Nehru National Institute of Technology, Allahabad-211004 (U.P.), India; e-mail: yogendrapra@gmail.com

³Department of Electronics and Communication Engineering, B.I.E.T., Jhansi-284128 (U.P.), India; e-mail: jps_uptu@rediffmail.com

⁴Department of Physics, Faculty of Sciences, Banaras Hindu University, Varanasi, (U.P.), India; e-mail: viveks@bhu.ac.in

The present study investigates the angular response and sensitivity of a surface plasmon resonance biosensor with metamaterial, by taking the advantage of the remarkable property of metamaterials. The proposed biosensor numerically shows that silver with a metamaterial layer enhances the sensitivity. The thickness of metamaterial and silver layer has been optimized. On comparing these results with a conventional surface plasmon resonance biosensor, it is observed that the sensitivity of the proposed biosensor is improved by introducing the metamaterial. The proposed biosensor has a sensitivity 6.3124 times higher than that of the conventional surface plasmon resonance sensor.

Keywords: metamaterials, sensor sensitivity, reflectance, evanescent field.

1. Introduction

Surface plasmon resonance (SPR) biosensor was first demonstrated for bio-sensing in 1983 by LIEDBERG *et al.* [1]. The principle, development and applications of SPR biosensors have been well described in several excellent papers [2–5]. The presence of free electrons at the interface of two materials is essential for the generation of surface plasmons (SPs). In practice, it always implies that one of these materials should be metal where free conduction electrons are abundant. When the incident light illuminates the interface between the metal and dielectric layer, an electron density oscillation occurs and as a result, SPs are excited and the evanescent wave is generated, which is exponentially decayed along the axis. This phenomenon is known as surface

plasmon resonance and it is only possible by coincidence of the wave vectors of the incident light and the SPs. These wave vectors are function of refractive indices of the dielectric, metal and analyte, *i.e.*, sensing medium. In sensing medium, a change in concentration of analyte will produce a local change in the refractive index (RI) near the metal surface. The change in RI will lead to a change in the propagation constant of SPs and in the angle of incidence, in order to satisfy the resonance condition. The resonance condition depends on the incident angle, wavelength of the light and the dielectric constants of both the metal and the silica-based prism [6]. Thus, the resonance condition determines the performance of the biosensor in term of sensitivity.

In most SPR sensor applications the plasmon supporting material is gold or silver, as they readily support plasmon modes at the range of visible frequencies. But, gold is usually preferred as it is resistant to oxidation and corrosion in different environments [7, 8]. But, the main drawback is the fact that biomolecules adsorb poorly in gold, which limits the sensitivity of the conventional SPR biosensor. Thus, silver would be better choice to improve the performance of the SPR sensors only if the surface of silver is made chemically inert [9].

First, theoretically [10, 11] and experimentally [12] demonstrated, metamaterials have been widely investigated in the field of left-handed materials, photonic crystals and magnetic metamaterials [13–15]. DE-KUI QING and GANG CHEN [16] found that metamaterials, which are artificially constructed materials, could enhance the intensity of the evanescent wave in the cladding without altering the propagation constant of the waveguide for both transverse electric (TE) and transverse magnetic (TM) mode. The idea for metamaterial-based sensing is to utilize the existence of evanescent electromagnetic waves at the interface between metamaterial and the dielectric analyte [17].

Recently, researchers have focused to their extraordinary absorptive properties in order to gain an absorptive element, which is one of the fundamental materials for microbolometer [18], optical attenuators [19] and sensing applications [20].

In order to improve the performance of a biosensor, researchers have proposed and fabricated waveguide sensor coated with metamaterial layers [21–24]. Very recently, UPADHYAY *et al.* also reported that sensitivity of the waveguide sensor can be improved by using metamaterials [25–27].

In this paper, we proposed a design of a SPR biosensor using a metamaterial layer. Our approach is to protect the silver layer against oxidation by using metamaterial layer which provides better adsorption of biomolecules, resulting in greater change in refractive index at sensor surface. In Section 2, modeling section and related equations are described. In Section 3, simulations for our proposed optimized SPR sensor are compared with a conventional SPR sensor and finally our paper is concluded in Section 4 with some significant remarks.

2. Theoretical background

Schematic diagram of the proposed SPR biosensor is shown in Fig. 1a. The k -th layer (k is from 2 to $N - 1$) has the thickness d_k and the refractive index $n(\lambda)$. Light source

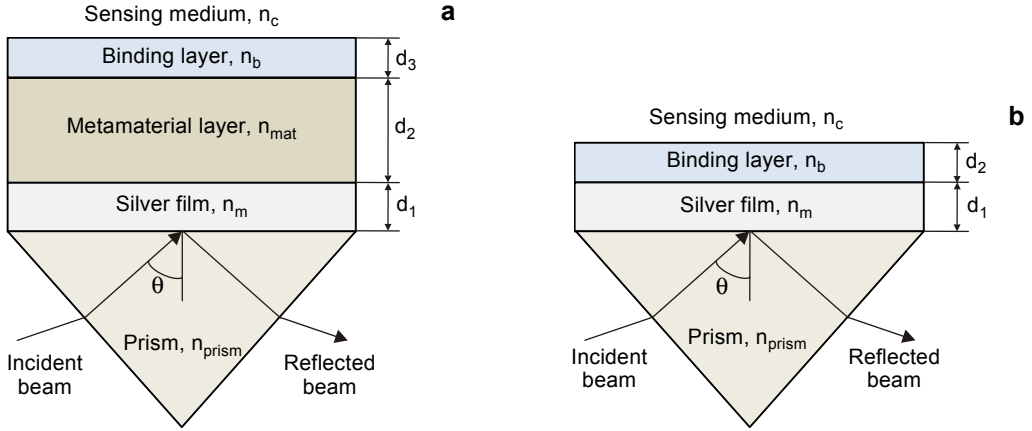


Fig. 1. Schematic diagram of proposed SPR biosensors configuration with their thickness and refractive indices (a), and conventional SPR biosensors configuration with their thickness and refractive indices (b).

of 1000 nm wavelength is used for the excitation. Here, the first layer is the prism and on the top of that is a silver layer, having the refractive indices $n_s = n_{\text{prism}} = 1.515$. The second layer is a silver layer which is deposited on the base of the prism. The wavelength dependence refractive index of the silver metal layer using the Drude–Lorentz model is given by

$$n_m(\lambda) = \sqrt{1 - \frac{\lambda^2 \lambda_c}{\lambda_p^2 (\lambda_c + i\lambda)}} \quad (1)$$

where λ_c is the collision wavelength (1.7614×10^{-5} m) and λ_p – plasma wavelength (1.4541×10^{-7} m) [28] and the thickness d_1 of silver is optimized. The third layer is metamaterial which is added on the silver layer and the thickness d_3 of the metamaterial is optimized. It is assumed that the periodic layer of metallic rods plus circular split ring resonators work as a metamaterial layer, which produces simultaneously negative permittivity and negative permeability. The size and period of the metallic rods and split ring resonators are less than the operating wavelength [29, 30]. Permittivity and permeability of the metamaterial are frequency dependent and can be written as:

$$\varepsilon_{\text{mat}}(\omega) = 1 - \frac{\omega_{\text{pl}}'^2}{\omega''^2 + i\Psi\omega''} \quad (2)$$

$$\mu_{\text{mat}}(\omega) = 1 - \frac{l\omega''^2}{\omega''^2 - \omega_{\text{iso}}^2 + i\Psi\omega''} \quad (3)$$

where $\omega_{\text{pl}}'^2$ is the plasma frequency, Ψ is the electron scattering rate, ω_{iso}^2 is the resonance frequency and l is the fractional area of a unit cell occupied by the split ring [17].

The numerical value of electric permittivity and magnetic permeability of the metamaterial is chosen $\{\epsilon_{\text{mat}}, \mu_{\text{mat}}\} = \{-4 + 0.001i, -2.4 + 0.001i\}$ [23–27]. The refractive index of the metamaterial n_{mat} is given by $n_{\text{mat}} = -(\epsilon_{\text{mat}}\mu_{\text{mat}})^{1/2}$. The fourth layer is a biomolecular recognition element (BRE) or a binding layer which is grown on the metamaterial layer for the attachment of the ssDNA. The binding layer (*e.g.*, ssDNA) thickness d_4 is defined as a 50 nm dielectric layer which was placed in the water and its refractive index n_b varied from 1.462 to 1.468 to produce a local increase in the refractive index. The initial value of 1.462 corresponds to the binding layer [31]. The fifth layer which is also the last layer of this biosensor is a sensing layer n_c . The refractive index of the sensing layer n_c is considered to vary from 1.33 (pure water, before absorption of biomolecules) to 1.35 (impure water, after absorption of biomolecules) in this paper. Conventional SPR biosensor configuration is also investigated here to do the comparison with the proposed sensor, which is shown in Fig. 1b. Parameters for both configurations are same except the metamaterial layer, which is an additional layer in proposed SPR biosensor.

Incident light at the interface of metal and prism, having particular wavelength ($\lambda = 1000$ nm), generates an evanescent wave. This evanescent wave penetrates through the silver layer and propagates along the x direction (parallel to the interface of the layers) with the propagation constant $k_x = n_{\text{prism}}(2\pi/\lambda)\sin(\theta)$, to generate the SPs at the silver-dielectric interface. Because of these SPs, surface plasmon wave (SPW) started to propagate along the silver-dielectric interface. The angle of incidence θ is controlled to adjust the propagation constant k_x in order to match with the propagation constant of SPW. The evanescent wave of SPW penetrates deeply in sensing medium and decreases exponentially. The incorporation of the metamaterial layer maximizes the evanescent field of SPW into the binding layer. Thereby the sensing layer refractive index modifies the propagating conditions for the evanescent field which in turn produces a significant change in the propagation wave of SPs, gives rise to a large resonance angle shift in compared to conventional SPR sensor [32]. In this manner, the complete theoretical formalism and experimental methods and protocols developed for conventional plasmon sensors are directly applicable in metamaterial-based sensors. Fresnel's multilayer reflection theory is used in the attenuated total reflection method to show the interaction between the light wave and the surface plasmon for p -wave (transverse magnetic wave) [33]. The plot of the total reflected intensity R versus the angle of incidence θ is called the SPR curve.

In this proposed multilayer structure (prism, metal, metamaterial and binding layer), the reflection coefficient for p -polarized incident light is obtained using a transfer matrix method. For a transfer matrix, consider a generalized N -layer model as shown in Fig. 1. The tangential field at the first boundary is related to that at the final boundary by,

$$\begin{bmatrix} U_1 \\ V_1 \end{bmatrix} = M_2 M_3 M_4 \dots M_{N-1} \begin{bmatrix} U_{N-1} \\ V_{N-1} \end{bmatrix} = M \begin{bmatrix} U_{N-1} \\ V_{N-1} \end{bmatrix} \quad (4)$$

For p -wave at boundary k ,

$$U_k = H_y^T + H_y^R \quad (5a)$$

$$V_k = E_y^T + E_y^R \quad (5b)$$

and

$$M_k = \begin{bmatrix} \cos(\beta_k) & -i \sin(\beta_k/q_k) \\ -iq_k \sin(\beta_k) & \cos(\beta_k) \end{bmatrix} \quad (6)$$

where $q_k = (\mu_k/\varepsilon_k)^{1/2} \cos(\theta_k)$ and $\beta_k = (2\pi/\lambda)n_k d_k \cos(\theta_k)$

From Eq. (6), after some mathematical calculations, the reflection coefficient for p -wave incident light is given by

$$r_p = \frac{(M_{11} + M_{12} q_N)q_1 - (M_{21} + M_{22} q_N)}{(M_{11} + M_{12} q_N)q_1 + (M_{21} + M_{22} q_N)} \quad (7)$$

$$M_{ij} = \left(\prod_{k=2}^{N-1} M_k \right)_{ij}, \quad i, j = 1, 2, \dots \quad (8)$$

Finally, the reflectivity R for the given multilayer structure is given by $R = |r_p|^2$. Performance characterization parameters for SPR sensors are sensitivity, detection accuracy and the quality parameter. Sensitivity of the SPR biosensor is measured as the small change in the refractive index of the cover region δn_c with the change in resonance condition $\delta \theta_{\text{res}}$ in the reflectance curve; therefore the sensitivity is given by $S = \delta \theta_{\text{res}}/\delta n_c$ [6]. Also, the sensitivity can be obtained as the derivative of the SPR response with respect to analyte concentration or refractive index of the analyte. Thus, the sensitivity can be obtained by the slope (change reflectance/change refractive index) $S = (\text{change reflectance})/\delta n_c$ [34].

3. Results and discussion

Figure 1a shows the schematic diagram of the proposed SPR biosensor configuration. The reflectivity is calculated and plotted for both the configurations. Excitation of a surface electromagnetic wave is recognized as the angle at which minimum reflected intensity is occurred and this angle is taken as the resonance angle. Figure 2 represents the reflectance curve of the proposed SPR sensor with a metamaterial layer and the conventional SPR sensor without a metamaterial layer using p -wave polarization. From Fig. 2, it is seen that the resonance angle and the reflectance obtained for the

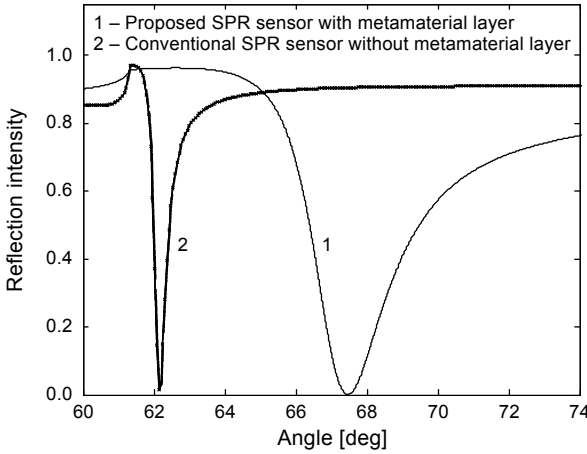


Fig. 2. Reflectance curve for proposed SPR biosensor (1) and conventional SPR biosensor (2). The refractive indices and thickness used are: $n_{\text{prism}} = 1.515$, $n_m = 0.1969 + 6.7957i$, $\epsilon_{\text{mat}} = -4 + 0.001i$, $\mu_{\text{mat}} = -2.4 + 0.001i$, $n_{\text{mat}} = -(\epsilon_{\text{mat}}\mu_{\text{mat}})^{1/2}$, $n_b = 1.462$ and $n_c = 1.33$, $d_1 = 42$ nm (optimized silver layer thickness), $d_2 = 349$ nm (optimized metamaterial layer thickness) and $d_3 = 50$ nm.

proposed biosensor are 67.46693 deg and 0.00001, respectively, which is higher than the angle of total internal reflection that occurs between the prism and water (61.38860 deg). The reflectance curve is also obtained for a conventional SPR biosensor without a metamaterial layer, and the observed resonance angle and reflectance are 62.15733 deg and 0.0050, respectively. Thus, the deepest plasmonic dip (*i.e.*, reflectance value) of the proposed sensor is at the resonance angle which corresponds to the surface electromagnetic wave propagating at the metamaterial/binding layer interface. It is depicted that SPR sensor with metamaterial gives the deeper plasmonic dip in comparison to the SPR without metamaterial, due to strong excitation of SPs. Strong excitation leads to the highest Ohmic loss in the silver metal of the proposed SPR sensor as shown in Fig. 2.

The attachment of binding surface is an important phenomenon in the proposed biosensor. When the binding layer is attached on the metamaterial layer (surface), the refractive index of sensing medium n_c changes, and this change in the refractive index depends on the hybridization event and binding surface density or concentration. It is seen from Fig. 3 that as the refractive index of sensing medium changes from $n_c = 1.330$ to $n_c = 1.345$ the dip of reflectance shifts towards a higher value of the incident angle θ . In Fig. 3, at resonance condition, the angle is 67.46578, 68.78358, 70.21598 and 71.99215 deg at the binding layer refractive index 1.330, 1.335, 1.340 and 1.345, respectively. In Fig. 3, it can be observed that the resonance angle shift is increasing with binding layer refractive indices due to binding surface concentration.

The thickness of a metamaterial and silver metal layer has been optimized for the optimum performance of the proposed SPR biosensor. Figure 4 shows the variation

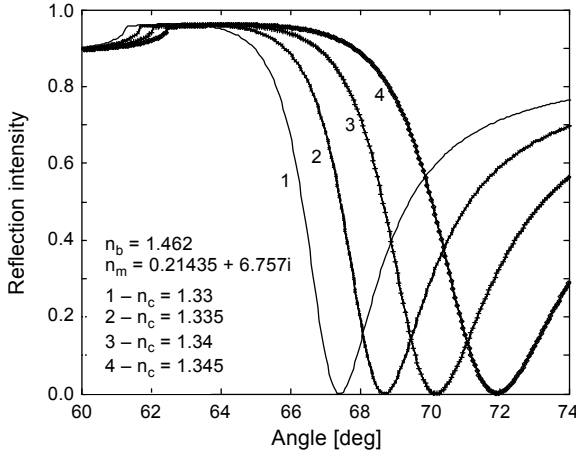


Fig. 3. Reflectance curves for proposed SPR biosensor. The refractive indices and thickness used are: $n_{\text{prism}} = 1.515$, $n_m = 0.1969 + 6.7957i$, $\epsilon_{\text{mat}} = -4 + 0.001i$, $\mu_{\text{mat}} = -2.4 + 0.001i$, $n_{\text{mat}} = -(\epsilon_{\text{mat}}\mu_{\text{mat}})^{1/2}$, $n_b = 1.462$, $d_1 = 42$ nm, $d_2 = 349$ nm and $d_3 = 50$ nm.

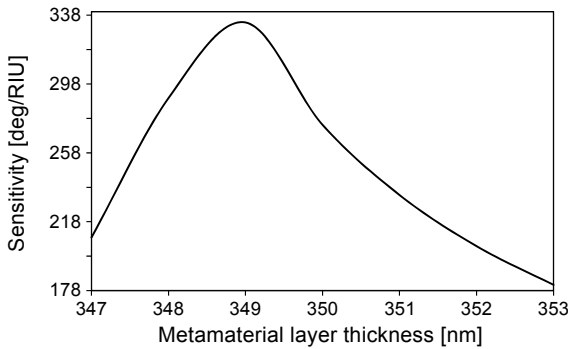


Fig. 4. Variation in sensitivity with metamaterial thickness of proposed SPR sensor at $n_c = 1.33$ and $n_b = 1.462$.

in sensitivity with metamaterial thickness for a constant silver thickness, *i.e.*, 42 nm. From Fig. 4, it is seen that the maximum sensitivity is 333.747915 deg/RIU (RIU – refractive index unit) at 349 nm optimized metamaterial layer thickness for the binding layer refractive index 1.462, which corresponds to 6.534011 deg resonance angle shift. Figure 5 shows the variation in the minimum reflectance of the proposed SPR sensor with silver film thickness for an optimized metamaterial layer thickness, *i.e.*, 42 nm. It is seen that for the silver film thickness of 42 nm, there is a minimum reflectance which is necessary for complete coupling of incident power to SPs at the binding layer with the refractive index 1.462.

Figure 6 shows the reflectance change as a function of a refractive index or concentration of the binding layer. From Fig. 6, it is seen that for a conventional SPR biosensor

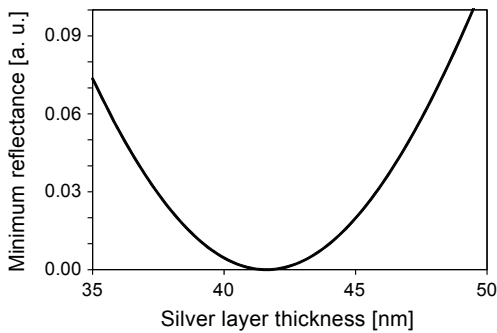


Fig. 5. Variation in reflectance with the silver layer of proposed SPR sensor at $n_c = 1.33$ and $n_b = 1.462$.

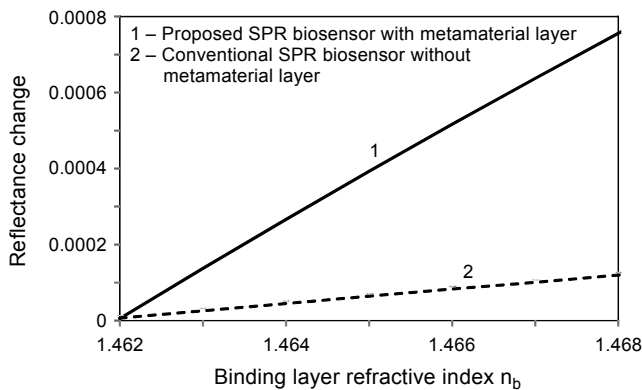


Fig. 6. Analysis between the reflectance and the refractive index of binding surface for proposed SPR biosensor (1) and conventional SPR biosensor (2).

without a metamaterial layer, the maximum reflectance increases from 0 to 0.000119711, while for the proposed metamaterial layer based SPR biosensor, the variation is from 0 to 0.000755659. Hence, the reflectance shift is completely linear over a range of the binding layer. In other words, the proposed SPR biosensor has the sensitivity 6.3124 ($0.000755659/0.000119711$) times higher than that of a conventional SPR biosensor without a metamaterial layer.

In order to study the dependence of sensitivity for the proposed SPR sensor at the optimized value of a metamaterial layer and silver film thickness, Fig. 7 shows the variation in sensitivity as a function of the sensing layer refractive index n_c for without metamaterial and with metamaterial at the fixed binding layer refractive index $n_b = 1.462$. It is observed from Fig. 7 that the sensitivity enhances by the inclusion of a metamaterial layer from 341.1555 deg/RIU at $n_c = 1.33$ to 489.8768 deg/RIU at $n_c = 1.35$ and SPR biosensor without a metamaterial layer increases the sensitivity from 93.8833 deg/RIU at $n_c = 1.33$ to 97.4033 deg/RIU at $n_c = 1.35$. It is observed

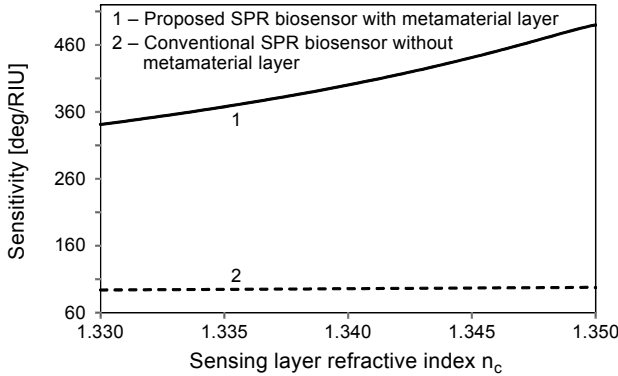


Fig. 7. Variation in sensitivity as a function of sensing layer refractive index for proposed SPR biosensor (1) and conventional SPR biosensor without metamaterial (2), $n_b = 1.462$.

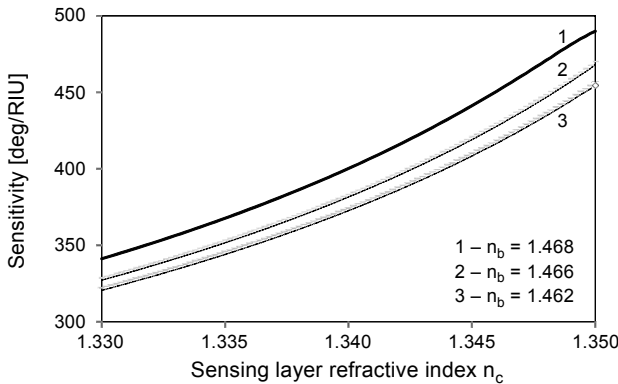


Fig. 8. Variation in sensitivity as a function of sensing layer refractive index for proposed SPR biosensor at $n_b = 1.462$, 1.466 and 1.468 .

from Fig. 7 that the introduction of a metamaterial layer increases the sensitivity due to the maximization of the evanescent field into the binding layer. Thereby the sensing layer refractive index modifies the propagating conditions for the evanescent field which in turn produces a significant change in propagation waves of SPs leading to a larger resonance angle shift as compared to the sensor without metamaterial, *i.e.*, a conventional SPR sensor [35]. Thus, the sensitivity increases with the incorporation of a metamaterial layer.

In order to study the binding surface dependent sensitivity, for a proposed biosensor, Fig. 8 shows the variation in sensitivity with metamaterial of different binding surface refractive indices $n_b = 1.462$, 1.466 and 1.468 at the fixed wavelength for the sensing medium refractive index $n_s = 1.33$ and 1.35 , respectively. At $n_b = 1.462$ sensitivity increases linearly from 320.5564 to 454.3583 deg/RIU. However, at $n_b = 1.466$

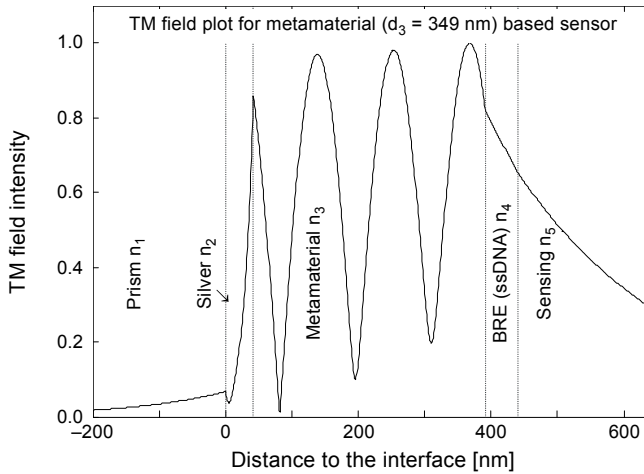


Fig. 9. Transverse magnetic (TM) field intensity as a function of distance normal from interface.

sensitivity increases linearly from 327.1863 to 468.1098 deg/RIU and at $n_b = 1.468$, the increase in sensitivity is from 341.1555 to 489.8768 deg/RIU. It can be observed that sensitivity increases at a higher binding surface refractive index. This indicates the dependence on the culture concentration of the analyte and its mobility.

Although the addition of a metamaterial layer increases the sensitivity, at the same time it increases the broadness of a SPR reflectance curve due to plasmon damping, as shown in Fig. 2. Damping occurs due to a wave vector shift into a metamaterial layer with a propagation velocity of electromagnetic waves smaller than in the sensing layer, which in turn leads to a decrease in the propagation velocity of a surface plasmon wave, and results in broadening of a SPR reflectance curve. Similar behavior has already been shown by POCKRAND [36]. Due to damping, the reflectance curve becomes broader and shallower in comparison with the reflectance curve of a SPR sensor without a metamaterial layer. The remedies for the broadening of a SPR reflectance curve using metamaterial will be shown in future communication.

Finally, the transverse magnetic (TM) field of p -polarized light is plotted in Fig. 9 for the verification of field distribution in different layers of the proposed metamaterial-based SPR sensor. A substantial decrease in the reflection intensity is observed at the resonance angle when the surface plasmon is excited. At the resonance condition, the magnetic field attains its maximum value, whereas the reflection intensity attains its minimum value [37, 38]. The absorption of the incident light takes place in the silver layer as a result of which the intensity of the TM field is increased in the silver layer and a peak is observed at the silver-metamaterial interface. The field intensity suddenly falls in the metamaterial layer and several modes are observed due to the large thickness of the metamaterial layer, as shown in Fig. 9. The field intensity starts to decrease at an approximately constant rate and deeper in the sensing layer through the binding layer (*i.e.*, ssDNA). By increasing the depth of the evanescent field in the binding layer, the

interaction volume of an evanescent field with the biomolecules is increased, which in turn maximizes the sensitivity [26].

4. Conclusion

The angle interrogation method for setting up the resonance condition is used to analyze the proposed SPR sensor. The reflected light is measured as a function of the angle of incidence with the fixed value of wavelength and thickness of constituent material layers, and then a sharp dip of reflected light is observed at the resonance angle due to sufficient excitation of SPs. The sensitivity of the proposed SPR sensor is studied and compared with that of a conventional SPR sensor without a metamaterial layer. It is observed that the proposed SPR sensor improved the sensitivity in comparison with a conventional SPR sensor without a metamaterial layer due to the adsorption of binding analyte which tunes the electromagnetic properties of metamaterial near the sensor surface.

Acknowledgements – The present work is partially supported by the Department of Science and Technology (DST), New Delhi, India, under the fast track young scientist scheme no. SB/FTP/ETA-0478/2012.

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*Received July 2, 2015
in revised form September 27, 2015*