# Multilayers analysis using the phenomenon of surface plasmon resonance

Rosmiza Mokhtar[a], Zainul Abidin Hassan[b], W. Mahmood Mat Yunus[b] and Zainal Abidin Talib[b]
*[a]Department of Engineering Sciences and Mathematics, College of Engineering, Universiti Tenaga Nasional, Km 7, Jalan Kajang-Puchong, 43009 Kajang, Selangor, Malaysia*
*[b]Department of Physics, Faculty of Science, Universiti Putra Malaysia, 43400 UPM, Serdang, Selangor, Malaysia*

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In the present work, the Surface Plasmon Resonance (SPR) technique was used to fabricate a sensitive optical sensor as well as characterizing metal thin film. To achieve this, a computer program was developed to carry out an accurate curve fitting of theory to reflectivity data. In the program, the Fresnel's equation was used to compute the reflectance and transmittance of light incident on multilayers (series of N layers and N+1 interface) between semi-infinite ambient and substrate media. Then the effect of multiple reflection and transmission were calculated by using 2×2 scattering matrix techniques. The Visual Basic 6.0 standard edition was used in the present work whereby a window-based program with graphic user interface (GUI) was developed for the simulation of reflection and transmission. The simulations were presented to motivate an effort to understand the shape of the resonances when the sensor head (gold thin film) exposed to different environments. Furthermore, the effect of increasing thickness and the modification of effective permittivity ( $\varepsilon_r$ and $\varepsilon_i$ ) were also studied.

## I. INTRODUCTION

A surface plasmon is a coupled, localized, transverse magnetic (TM) electromagnetic field or charge density oscillation, which may propagate along an interface between two media with dielectric constants of opposite sign, such as a metal and a dielectric. A surface plasmon may be observed involving the reflectance and transmittance of light from a metal thin film through prism coupling method. Prism coupling can be constructed by depositing the metal thin film onto the prism surface. Optical excitation of plasmons is not possible by direct impact of light on a metallic surface, so a prism coupling arrangement is needed [1]. One possibility is to use Kretschmann configuration in which a p-polarized, collimated light beam passing through a glass prism undergoes total internal reflection (TIR) at glass thin metal film-dielectric interface [1,2]. The surface plasmon resonance condition is given as [3]:

$$k_{spr} = \frac{\omega}{c}\sqrt{\left(\frac{\varepsilon_1\varepsilon_2}{\varepsilon_1+\varepsilon_2}\right)} \tag{1}$$

where c is the speed of light in vacuum, $\omega$ is the light frequency, and $\varepsilon_1$, $\varepsilon_2$ are the dielectric constants of the metal and the medium in contact with the metal, respectively. The dielectric constant of metal given by $\varepsilon = \varepsilon_r - i\varepsilon_i$, is related to optical constants by expression $\varepsilon_r = n^2 - k^2$ and $\varepsilon_i = -2nk$. Here n and k are the index of refraction and extinction coefficient of the medium.

When a monochromatic, P-polarized plane wave is incident at an angle $\theta$ on a metal thin film through a prism, multiple reflection and transmission will be created. By varying the value of incident angle, the minimum in reflectance will be obtained. At certain angles of incidence, surface plasmon is excited and resonated along the interface between the metal and dielectric. At this point, the component of incident wave vector parallel to the interface, $k_x = \left(\frac{\omega}{c}\right)\sqrt{\varepsilon_1}\sin\theta$, must be equal to the surface plasmon wave vector, i.e. $k_x = k_{spr}$. This phase matching occurs when the incident angle in medium 1 (prism) is greater than the critical angle, $\theta_c = \sin^{-1}\left(\sqrt{\varepsilon_3}/\sqrt{\varepsilon_1}\right)$ [4]. In most surface plasmon resonance (SPR) experiments, gold and silver are widely used as a thin film on the prism surface.

The shape of the whole resonance curve depends primarily on the thickness and optical properties of the metal layer [5,6]. The shape of the surface plasmon resonance (SPR) curve can be quantitatively described by Fresnel's equation for the p-polarized light reflectivity of a multilayered system [1,7-9].

## II. METHODOLOGY

The Microsoft Visual Basic 6.0 is used as the programming language. The program was developed to calculate the reflectance and transmittance as a function of incidence angle. In the program, we have to input real and imaginary part of dielectric constants ( $\varepsilon$ ) of mediums, the incident light wavelength ($\lambda$), the incident angle of light ($\theta$) and the thickness of layers (d). To carry out an accurate fitting of experimental data to the theory, the value of dielectric constants ($\varepsilon$) and the thickness of layers (d) are adjusted until the lowest sum of square error was obtained.

Consider a multilayer system as shown in Fig. 1(a). Computation of the reflectance and transmittance for a system of multilayers films are based on application of 2×2 scattering matrix, where the electric field component in the media can be related by equation [7]:

$$\begin{bmatrix} E_0^+ \\ E_0^- \end{bmatrix} = \begin{bmatrix} S_{11} & S_{12} \\ S_{21} & S_{22} \end{bmatrix} \begin{bmatrix} E_n^+ \\ 0 \end{bmatrix} \quad (2)$$

where S is the scattering matrix of the system and is composed of the product of alternating interface and layer matrices $I_{ab}$ and $L_b$ respectively, for the system. For interface between two media a/b the interface matrix is defined as

$$I_{ab} = \frac{1}{t_{ab}} \begin{bmatrix} 1 & r_{ab} \\ r_{ab} & 1 \end{bmatrix} \quad (3)$$

and the layer matrix for layer b is given by

$$L_b = \begin{bmatrix} e^{i\beta} & 0 \\ 0 & e^{-i\beta} \end{bmatrix}. \quad (4)$$

The phase shift $\beta$ describes in each layer of thickness $d_b$ with index of refraction $N_b$ and angle of refraction $\theta_b$ is given by

$$\beta = kN_b d_b \cos\theta_b \quad (5)$$

where

$$k = \frac{2\pi}{\lambda}. \quad (6)$$

The matrix components, $r_{ab}$ and $t_{ab}$ are simply the Fresnel reflection and transmission coefficients for an interface a/b respectively.

Consider a ray of light in medium 0 with index of refraction $N_o$ incident on interface with medium 1 with index of refraction $N_1$ as given in Fig. 1(b). The Fresnel reflection coefficients at the 0-1 interface is given by

$$r_{01p} = \frac{N_1\cos\theta_0 - N_0\cos\theta_1}{N_1\cos\theta_0 + N_0\cos\theta_1} \quad (7)$$

$$r_{01s} = \frac{N_0\cos\theta_0 - N_1\cos\theta_1}{N_0\cos\theta_0 + N_1\cos\theta_1} \quad (8)$$

while the Fresnel transmission coefficients are given by

$$t_{01p} = \frac{2N_0\cos\theta_0}{N_1\cos\theta_0 + N_0\cos\theta_1} \quad (9)$$

$$t_{01s} = \frac{2N_0\cos\theta_0}{N_0\cos\theta_0 + N_1\cos\theta_1} \quad (10)$$

where $r_p$ and $t_p$ respectively are reflection and transmission coefficients for the wave with polarization parallel to the plane of incidence while $r_s$ and $t_s$ are reflection and transmission coefficients for the wave with polarization perpendicular to the plane of incidence.

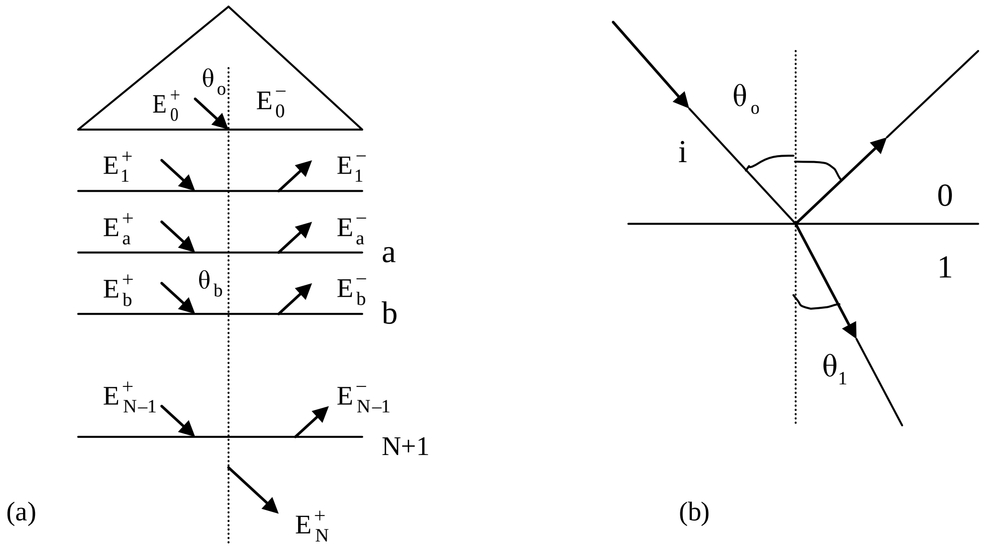


**FIG. 1.** (a) Propagation of EM fields in multilayer thin film system with N-1 layers, and (b) an incident ray I hits interface 0-1 at an angle $\theta_o$ and refracted into medium 1 at angle $\theta_1$ and reflected in medium 0 at angle $\theta_o$.

The scattering matrix represents the overall optical properties, (i.e. reflectance and transmittance of the system), expressed as a product of the interface matrices and layer matrices of equations (4) and (5) respectively. The scattering matrix for N-layered system is given by

$$S = I_{01}L_1I_{12}L_2.........L_N I_{N,N+1} \qquad (11)$$

$$S = \left(\frac{1}{t_{01}t_{12}t_{23}...t_{N,N+1}}\right)\begin{pmatrix} 1 & r_{01} \\ r_{01} & 1 \end{pmatrix}\begin{pmatrix} e^{i\beta_1} & 0 \\ 0 & e^{-i\beta_1} \end{pmatrix}\begin{pmatrix} 1 & r_{12} \\ r_{12} & 1 \end{pmatrix}$$
$$\times\begin{pmatrix} e^{i\beta_2} & 0 \\ 0 & e^{-i\beta_2} \end{pmatrix}...\begin{pmatrix} e^{i\beta_N} & 0 \\ 0 & e^{-i\beta_N} \end{pmatrix}\begin{pmatrix} 1 & r_{N,N+1} \\ r_{N,N+1} & 1 \end{pmatrix}.$$

Having calculated S for the multilayer system, the reflectance and transmittance are simply obtained from Eq. (12). The reflectance for the two-interface system is given by

$$R = \left[\frac{E_0^-}{E_0^+}\right]^2 = \left[\frac{S_{21}}{S_{11}}\right]^2. \qquad (12)$$

The transmittance for two-layer system is given by

$$T = \left[\frac{E_n^+}{E_0^+}\right]^2 = \left[\frac{1}{S_{11}}\right]^2. \qquad (13)$$

For reflectance of transmittance versus incidence angle of light the angle can be varied from $\theta_{min}$ to $\theta_{max}$, with step size 0.01$^o$. So a set of data for reflectance and transmittance as a function of incidence angle is obtained.

## III. RESULTS AND DISCUSSION

We presented the simulations to evaluate whether the results from the developed program represent the real situation.. When a p-polarized light wave is incident on the interface between two transparent media the reflected wave entirely disappears at a particular angle of incidence called the Brewster angle, $\theta_B$ and the incident wave is totally refracted into the second medium. Figs. 2 and 3 show the variation with angle of incidence of the reflectance for p and s polarized light when incident on the air/glass and air/silicon interface. The refractive index of air and glass are 1.0002 and 1.5 respectively. The complex refractive index of silicon is 4.05 − i0.028. In this simulation, we varied the incidence angle from 0 to 90 degrees with step 0.1$^o$ and the wavelength for the incident light is 546.1 nm. From the plot, the reflectance reached a minimum value at the Brewster angle of 56.30 and 76.10 respectively. We achieved a satisfactory agreement compared with the experimental results of Azzam and Bashara [7].

If a p-polarized optical plane wave is incident on a two-transparent-media interface from the side of high index of refraction, (e.g. prism/air interface), total internal reflection takes place at angle of incidence larger than a critical angle $\theta_C$. Fig. 4 shows the variation with angle of incidence of the reflectances $R_s$ and $R_p$, when light is incident on a glass/air interface. The value of refractive index of air and glass are similar with the one explained above. Once again, we varied the incidence angle from 0 to 90 degrees with step 0.1$^o$, and the wavelength of the incidence light is 546.1 nm. From the plot, the value of Brewster angle is 33.7 and the critical angle has a value of 41.9, which agreed well with the one reported by Azzam and Bashara [7].

For surface plasmon excitation simulation, we used gold metal with permittivity and thickness of –10.92 + i1.49 and 50 nm respectively. The dielectric constant of the prism was 2.6131 and the wavelength of the incidence light was 632.8 nm. We varied the incident angle from 34 to 47 degrees with step size of 0.1$^o$, which included the critical angle, $\theta_c$ and resonance angle, $\theta_{spr}$. The plot of reflectance as a function of incident angle for this simulation was illustrated in Fig. 5.

From the plot, we could determine three important parameters, i.e. the critical angle $\theta_c$, the minimum of reflectance, and the resonance angle, $\theta_{spr}$. The reflectance reached the minimum at the resonance angle of 40.65$^o$. The similar shape of the reflectivity curve was reported by Sadowski *et al.* [1].

The simulation of the developed program also included the investigation of the effect of a variation in thickness of layer, permittivity and incidence wavelength. In demonstrating the effect of a variation of layer thickness, the thickness of gold thin film was varied from 30 nm to 80 nm. The permittivity of gold thin film, prism and air are similar to the one explained above. We varied the incidence angle from 34 up to 47 degrees with step 0.1$^o$, while the incidence wavelength was 632.8 nm. Fig. 6 shows that when the thickness increased from 30 nm to 50 nm, the resonance angle shifted to the left and the reflectance (minimum) decreased. Furthermore, when the thickness increased from 50 nm up to 80 nm, the reflectance (minimum) increased but the resonance angle remained at the same value.

The relationship between the resonant angle, $\theta_{spr}$ with the thickness of layer is shown in Fig. 7. The results showed that $\theta_{spr}$ decreased drastically with increasing thickness from 20 nm to 70 nm, and then remained at the same value with the thickness ranging from 70 nm up to 120 nm.

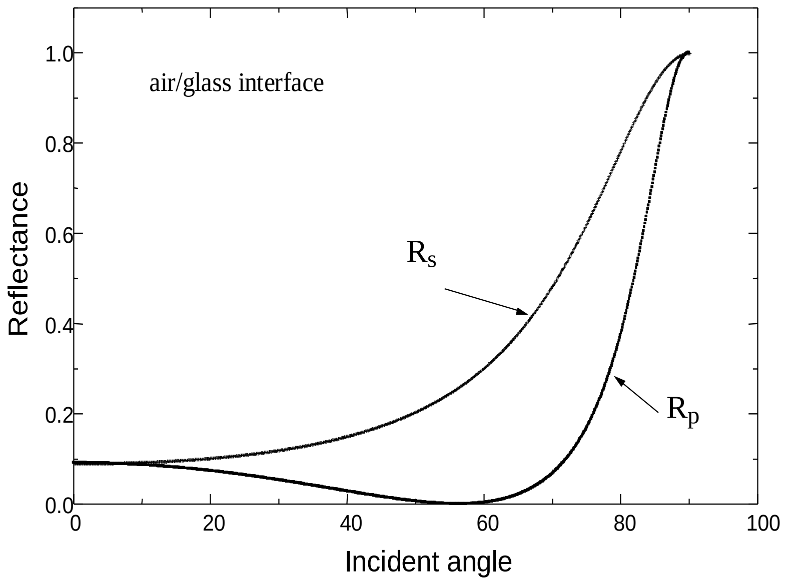


**FIG. 2.** The reflectance for the P and S polarization as a function of incidence angles for air/glass interface.

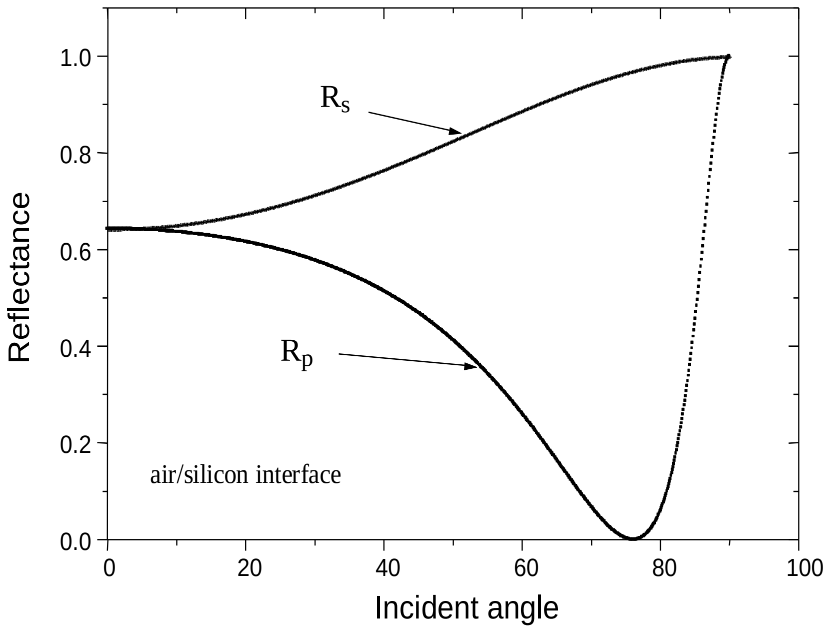


**FIG. 3.** The reflectance for the P and S polarization as a function of incidence angles for air/silicon interface.

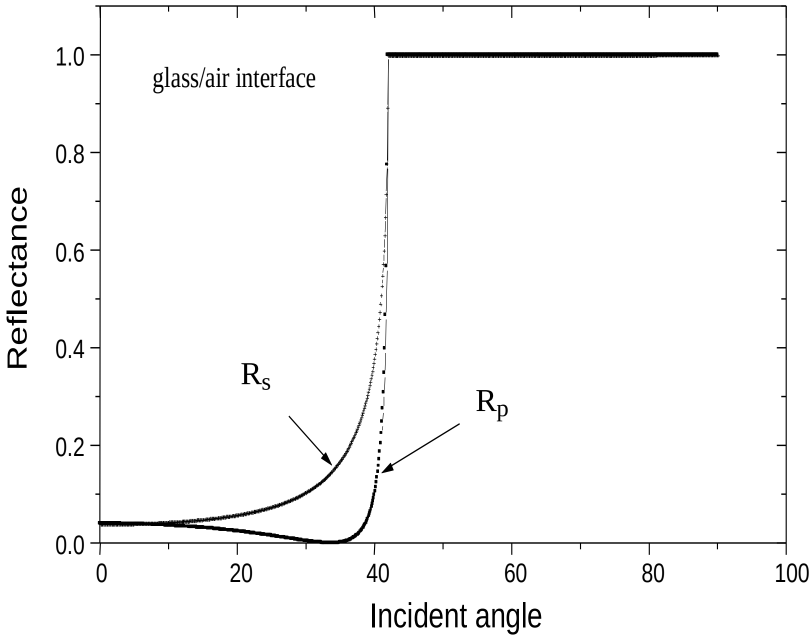


**FIG. 4.** The reflectance for the P and S polarization as a function of incidence angles for glass/air interface.

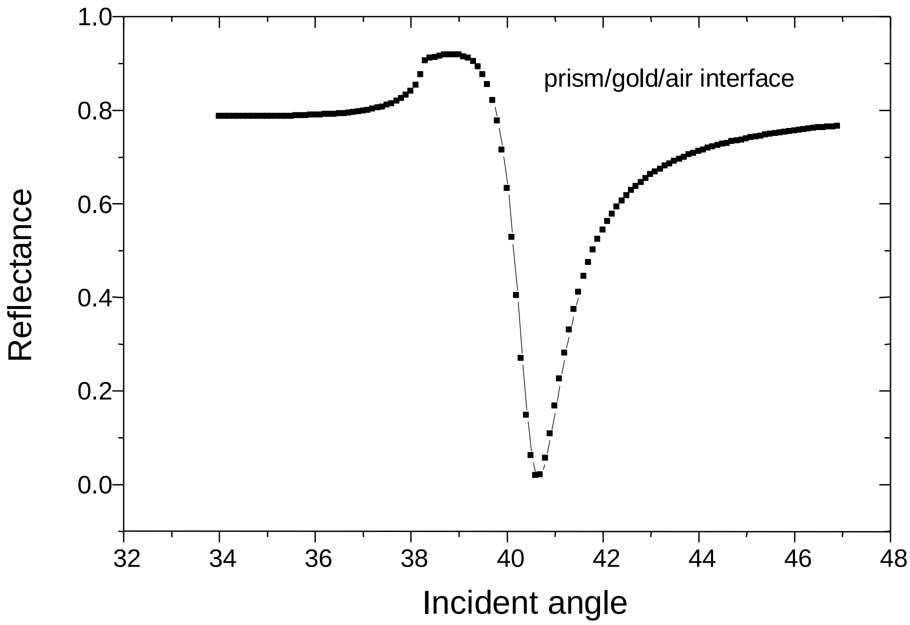


**FIG. 5.** The reflectance for the p-polarized light as a function of incidence angles for prism/gold/air interface.

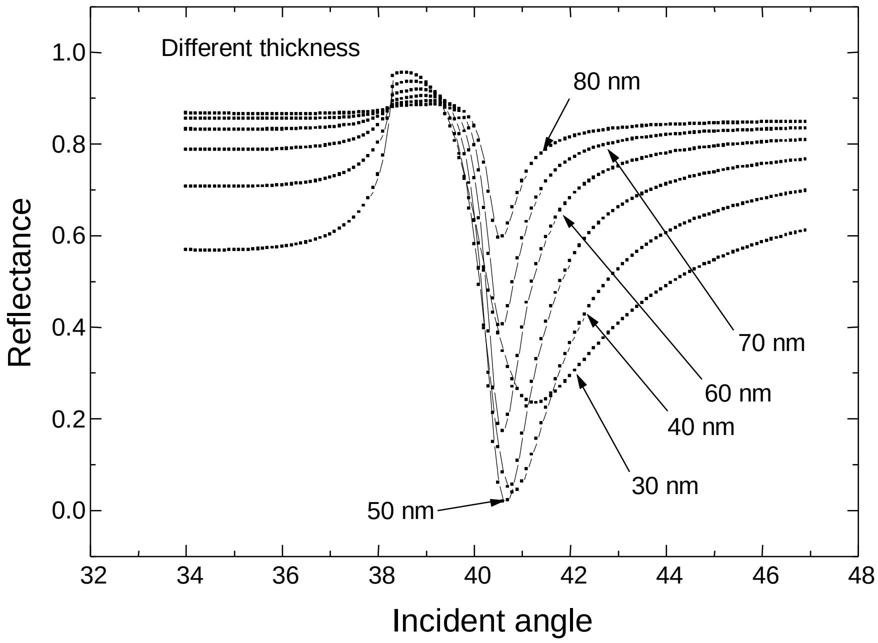


**FIG. 6.** The plot of reflectance as a function of incidence angle for different thickness.

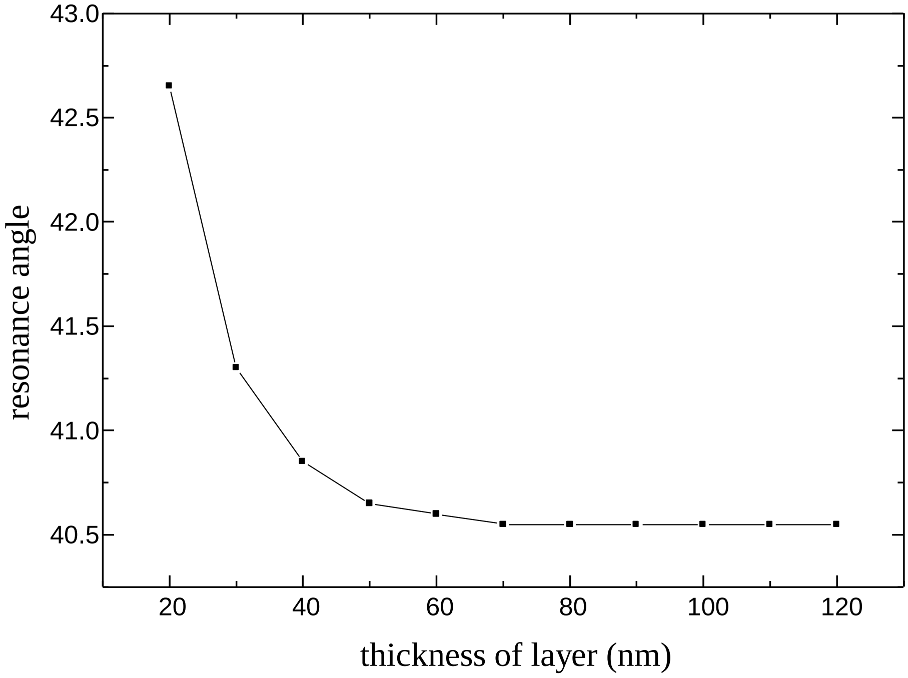


**FIG. 7.** The relationship between the resonant angle, $\theta_{spr}$ with the thickness of layer.

From Fig. 8, it is clearly seen that the reflectivity curve reached the minimum value at the layer thickness of 50 nm. The rate of decreasing of reflectivity (minimum) with thickness ranging from 20 to 50 nm is equal to the increasing of reflectivity (minimum) with thickness ranging from 50 nm to 90 nm. The increasing of reflectivity (minimum) becomes slower when the thickness increased from 90 nm to 120 nm.

The condition of the material of interest contributes to the effective values of optical constants at the interface, which can be determined by fitting the experimental data to the theory. By using the similar system as explained above, we varied the real and imaginary value of permittivity of gold thin film and the effect was shown in Figs. 9 and 10. From Fig. 9, the plot of reflectance as a function of incidence angle shifted to the right when the real permittivity is increased. The reflectivity curve also becomes wider with the increasing of real permittivity. With decreasing of imaginary permittivity from 4.0 to 1.49, as shown in Fig. 10, the minimum value of reflectivity curve decreased, but then increased when the value decreased to 0.5. The reflectivity curve becomes narrower with the decreasing of imaginary permittivity, but the resonance angle remains at the same value for imaginary value ranging from 0.5 to 2.0. From the observation above, it is clearly understood that the changing in real value of permittivity does not alter the depth of the curve, but gives effect to the value of resonant angle, $\theta_{spr}$. We also can conclude that by varying the value of imaginary permittivity, the depth of the reflectivity curve is clearly affected, but the $\theta_{spr}$ is slightly altered.

Fig. 11 shows the plot of reflectance as a function of incident angle for one-layer system and two-layer system. We used gold with the permittivity of –10.92 + i1.49 for the first layer and gold sulphide with the permittivity of –8.9 + i1.54 for the second layer. The thickness for the first and second layer was 50 nm and 22.2 nm, respectively. The formation of gold sulphide on the gold layer was done by exposing the hydrogen sulphide gas to the gold surface for a required duration of time. Then the exposed surface was used for the SPR measurement and the reflectivity curve was fitted to the theory to determine the optical constant of the new compound generated on the gold surface.

The theoretical curve of the two-layer system in Fig. 11 was determined from the curve fitting technique. When the number of layer increased, the plot shifted to the right without altering its broadness. The latter effect is due to the slight difference in imaginary value of permittivity. The different value of real permittivity caused the shifting of the plot where the resonant angle changed from $40.65^{o}$ to $41.05^{o}$.

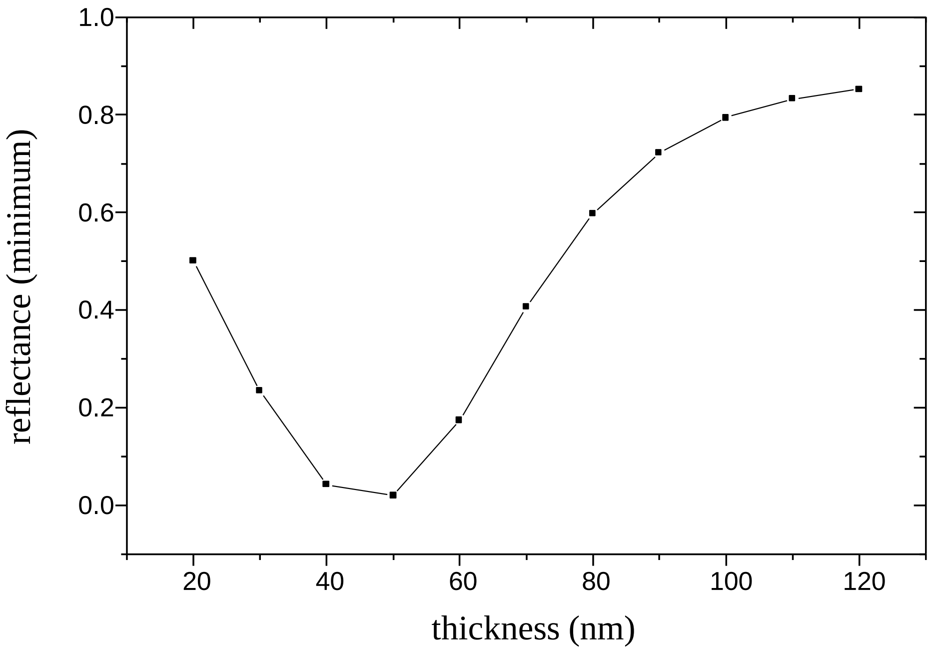


**FIG. 8.** The relationship between the minimum reflectivity with the thickness of layer.

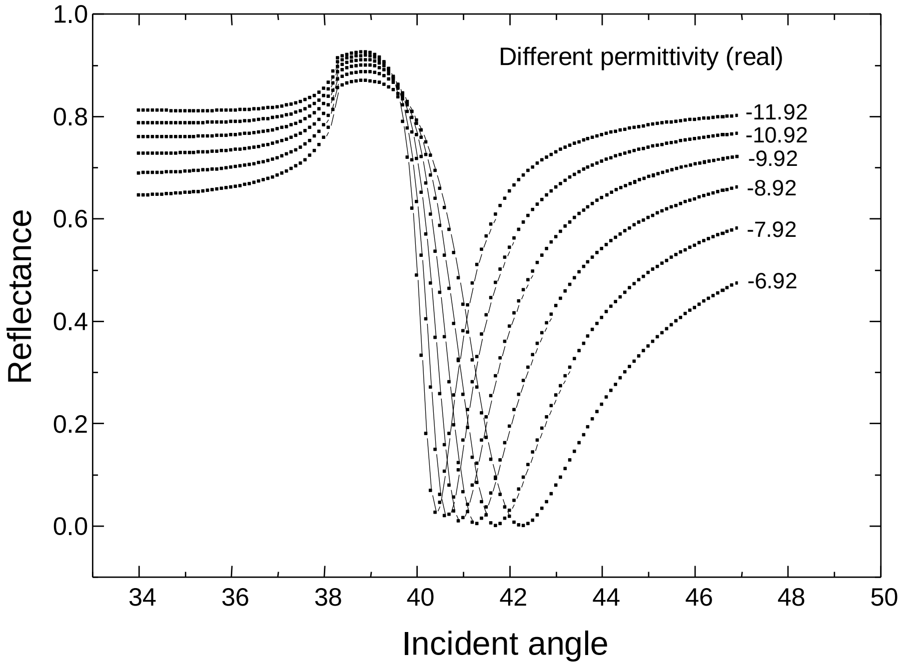


**FIG. 9.** The plot of reflectance as a function of incidence angle for different permittivity (real).

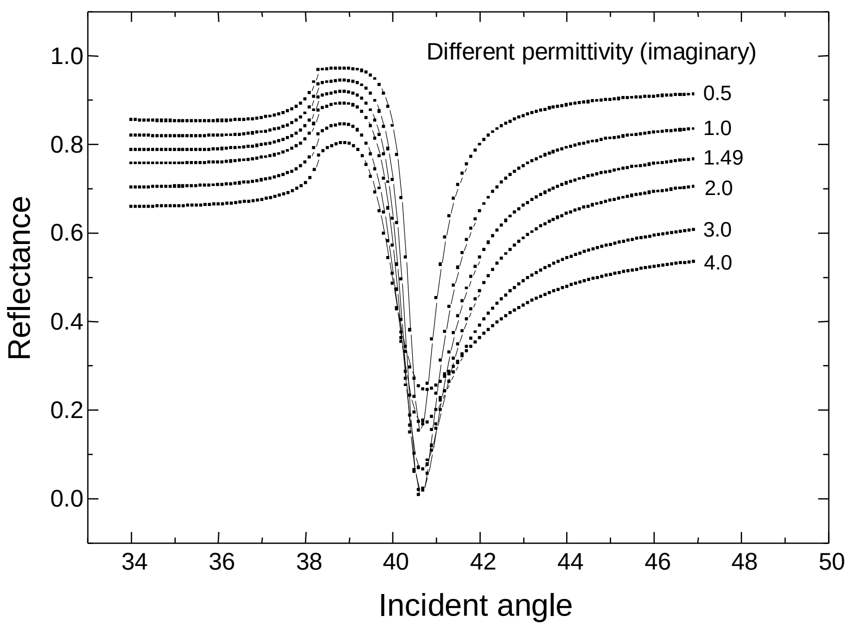


**FIG. 10.** The plot of reflectance as a function of incidence angle for different permittivity (imaginary).

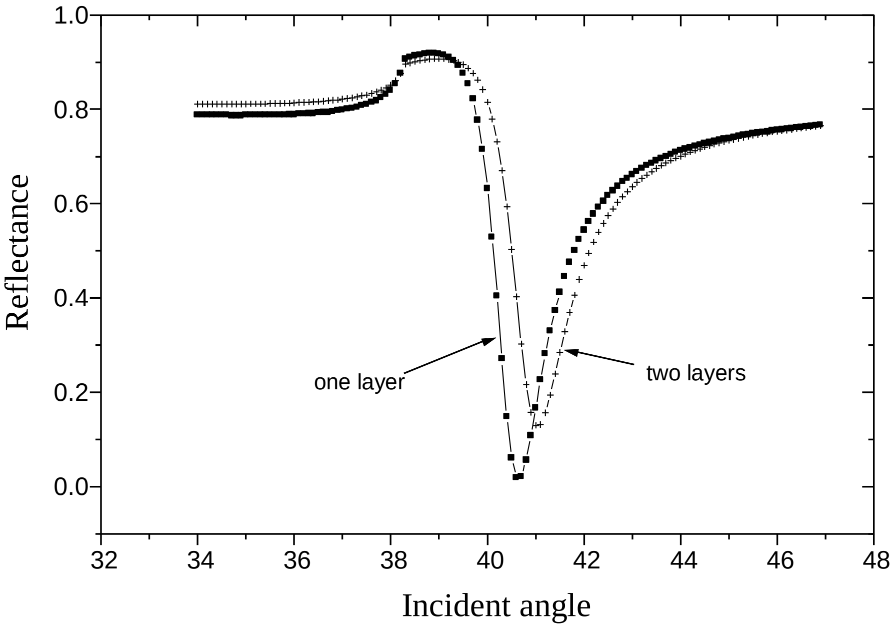


**FIG. 11.** Reflectivity curves for one layer and two layer of gold thin film as a function of incident angle.

## IV. CONCLUSION

We have demonstrated that the program developed could be used to carry out the required calculation to plot the reflectance for P and S polarized incident light, for single and multilayer analysis. We also achieved an excellent understanding of the shape of reflectivity curve when the optical constants of layers were varied.

## ACKNOWLEDGEMENT

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