The dependence of ellipsometric parameters $\Delta$ and $\Psi$ on refractive index of superficial film

A D Berdie$^1$, A A Berdie$^2$ and S Jitian$^1$

$^1$Politehnica University of Timisoara, Department of Electrical Engineering and Industrial Informatics, 5 Revolutie Str., Hunedoara, 331128, Romania
$^2$"Lucian Blaga" University, Faculty of Letters and Arts, 5-7 Bd. Victoriei, Sibiu, 550024, Romania

E-mail: jitian_s@yahoo.com

Abstract. The ellipsometrical analysis of external specular reflection of light on non-absorbing superficial films allows us to know factors which influence the ellipsometric measurement of the analyzed system. For optical non-absorbing superficial films the curves $\Delta = f(\Psi)$ are closed, the curves $\Delta = f(d_f)$ and $\Psi = f(d_f)$ are periodical, while the curves $\Delta = f(n_f)$ and $\Psi = f(n_f)$ are pseudo-periodic. Observations on the dependence of the ellipsometric parameters $\Delta$ and $\Psi$ on $n_f$ are presented.

1. Introduction
A change in the light polarization state occurs in the case of external reflection ($n_f > n_o$) of a monochromatic light radiation on a homogeneous and isotropic, optical non-absorbing, superficial film deposited on a solid reflector support.

The model of external reflection of a monochromatic radiation on a non-absorbing optical superficial film is schematically represented in Figure 1.

![Figure 1](image)

Figure 1. The model of the external reflection of a plan-polarized monochromatic radiation on the substrate-superficial film system

The change in the polarization state of the reflected radiation can be measured by ellipsometry [1]. The measurable ellipsometric parameters are $\Delta$ and $\Psi$. The first parameter $\Delta$ expresses the phase shift of the electrical component of the light radiation parallel to the plane of incidence relative to the perpendicular component on the incidence plane, after reflection and $\tan \Psi$ represents the relative attenuation of the two components after reflection [2-6].
\[ \tilde{\rho} = \tan \Psi \cdot \exp(i\Delta) = \frac{\tilde{E}_p^{(i)}}{\tilde{E}_s^{(i)}} = \frac{\tan \psi^{(i)} \cdot \exp(i\delta^{(i)})}{\tan \psi^{(i)} \cdot \exp(i\delta^{(i)})} = \frac{E_p^{(i)}}{E_s^{(i)}} \cdot \exp(i(\delta_p' - \delta_s' - \delta_i' + \delta_i')) \] (1)

The complex size \( \tilde{\rho} \) can be calculated according to the ellipsometric parameters \( \Delta \) and \( \Psi \) by the relation:

\[ \tilde{\rho} = \tan \Psi \cdot \exp(i\Delta) = \tan \Psi \cos \Delta + i \cdot \tan \Psi \sin \Delta = \text{Re} \tilde{\rho} + i \cdot \text{Im} \tilde{\rho} = \frac{\tilde{R}_p}{\tilde{R}_s} \] (2)

taking into account the reflections and therefore the attenuations and phase shifts thus produced on all interfaces.

The complex parameters \( \Delta \) and \( \Psi \) are expressed according to the Fresnel complex reflection coefficients corresponding to the reflections on the two interfaces \( \Sigma_{12} \) and \( \Sigma_{23} \):

\[ \tilde{R}_p = \tilde{r}_p^{12} + \tilde{r}_p^{23} \exp \tilde{D} \] and \( \tilde{R}_s = \tilde{r}_s^{12} + \tilde{r}_s^{23} \exp \tilde{D} \]

when:

\[ \tilde{D} = -\frac{4\pi id_i}{\lambda} \sqrt{n_i^2 - n_0^2 \sin^2 \varphi_0} \] (3)

The expression \( \exp \tilde{D} \) is a periodic function which depends on: the refractive index of the superficial film \( n_i \), its thickness \( d_i \), the incidence angle \( \varphi_0 \) and the wavelength \( \lambda \) of the incident radiation [3].

The complex parameters: \( \tilde{R}_p \), \( \tilde{R}_s \) and \( \tilde{\rho} \) should be repeated after particular values of the film thickness \( d_i \) or the refractive index \( n_i \) for which \( \tilde{D} \) is a multiple of 2\( \pi \).

\[ \tilde{D} = -\frac{4\pi id_i}{\lambda} \sqrt{n_i^2 - n_0^2 \sin^2 \varphi_0} = m \cdot 2\pi i \] for \( m = 1,2,3,... \) (5)

The value of film thickness for which the exponential function \( \exp \tilde{D} \) is repeated is \( d_{\text{min}} \).

\[ d_i = m \frac{\lambda}{2\sqrt{n_i^2 - n_0^2 \sin^2 \varphi_0}} \text{ and } d_{\text{min}} = \frac{\lambda}{2\sqrt{n_i^2 - n_0^2 \sin^2 \varphi_0}} \] for \( m = 1,2,3,... \) (6)

For thicker films, the thickness shall be expressed by the relationship:

\[ d_i = d_0 + m \cdot d_{\text{min}} \text{ for } m = 1,2,3,... \] (7)

where \( d_0 \) is the thickness, determined by ellipsometrical measurement (which is called standard solution) and \( m \) is an integer [9].

For example, if \( \Delta = 186.37^0 \) and \( \Psi = 51.317^0 \), the film thickness with \( n_i = 1.4 \) can be 120nm or \( d_i = (120 + m \cdot 256.4) \text{ nm} \) for \( m = 1,2,3,... \) [8].

2. The dependence of ellipsometric parameters \( \Delta \) and \( \Psi \) on refractive index of superficial film

The ellipsometric parameters \( \Delta \) and \( \Psi \) can be expressed with the relations:

\[ \Delta = \arctg \frac{\text{Im} \tilde{\rho}}{\text{Re} \tilde{\rho}} \quad ; \quad \Psi = \arctg |\tilde{\rho}| \] (8)
Both parameters are periodic functions that depend on the thickness of the surface film and its refractive index, as shown in Figure 2.

![Figure 2](image2.png)

**Figure 2.** The dependence of $\Delta$ and $\Psi$ on the refractive index and superficial film thickness

$\tilde{n}_s = 2.67(1-1.18i); \varphi_0 = 60^0; \lambda = 562.5$ nm

The ellipsometric parameters $\Delta$ and $\Psi$ are periodic functions toward the refractive index of the surface film, with the same period. Therefore, the curves $\Delta = f(\Psi)$ shown in Figure 3 are closed curves [10], [11].

![Figure 3](image3.png)

**Figure 3.** The plots $\Delta = f(\Psi)$ for superficial films with $n_f = 1.2 \div 2.5$, deposited on a metal substrate.

$\tilde{n}_s = 2.67(1-1.18i); \varphi_0 = 60^0; \lambda = 562.5$ nm; $\Delta_0 = 147.118^0$ and $\Psi_0 = 33.116^0$

$\Delta_0$ and $\Psi_0$ are the ellipsometric parameters measured for the film-free substrate.

The curves are more scattered as the film is thinner, so that $n_f$ can be determined with better precision.

The complex parameters: $\Delta$ and $\Psi$ are repeated after a certain value of the refractive index $n_f$ of the film for which $\tilde{D}$ is an integer multiple of $2\pi i$, that is, when the refractive index of the superficial film fulfills the condition:

$$n_f = \sqrt{(n_0 \sin \varphi_0)^2 + \left(m \frac{\lambda}{2d_0}\right)^2}, \text{ for } m = 1,2,3,\ldots$$  

(9)
The curves $\Delta = f(n_f)$ and $\Psi = f(n_f)$ are pseudo-periodic for this reason, as can be seen from Figures 4 and 5 [9].

![Figure 4](image1.png)

**Figure 4.** The dependence of $\Delta$ on the superficial film refractive index $n_f$

![Figure 5](image2.png)

**Figure 5.** The dependence of $\Psi$ on the superficial film refractive index $n_f$

All curves $\Delta = f(n_f)$ or $\Psi = f(n_f)$ pass through the same point $(\Delta_0 = 147.12^0; \Psi_0 = 33.116^0; n_{f0} = 2.942)$ for the superficial film thickness an integer multiple of 100 nm ($d_f = m \cdot 100$ nm; where $m = 1, 2, 3, \ldots$; $d_0 = 100$ nm) as seen in Figures 4b and 5b. This point corresponds to $n_{f0} = 2.9425$. Also, all $\Delta = f(n_f)$ or $\Psi = f(n_f)$ curves pass through the same point $(\Delta_0 = 147.12^0; \Psi_0 = 33.116^0; n_{f0} = 1.65153)$ for the superficial film thickness an integer multiple of 200 nm ($d_f = m \cdot 200$ nm; where $m = 1, 2, 3, \ldots$; $d_0 = 200$ nm). This point corresponds to $n_{f0} = 1.6515$. If in relation (9) the condition is that $m = 1$ then we obtain:

$$d_0 = \frac{\lambda}{2 \sqrt{n_f^2 - n_{f0}^2 \sin^2 \phi_0}}$$

(10)

Values of $n_{f0}$ for different values of $d_0$ are shown in Table 1.

| $\phi_0$ (deg) | 60 |
|----------------|----|
| $\lambda$ (nm) | 562.5 |
| $d_0$ (nm)     | 350 300 250 200 150 100 |
| $n_{f0}$       | 1.18 1.28 1.48 1.65 2.06 2.94 |
If the thickness of the surface film is small compared to the wavelength of the light radiation (tens of nanometers), then equation (9) can be approximated as:

\[ n_f \approx m \frac{\lambda}{2d_f} = m \cdot n_{f\min}; \quad m = 1, 2, 3, \ldots \]  

(11)

and the curves \( \Delta = f(n_f) \) and \( \Psi = f(n_f) \) are periodic. The value of \( n_{f\min} \) from equation (11) is all the greater as \( d_f \) is smaller, as shown in Figures 4a and 5a. For nano-sized film, the value of \( n_{f\min} \) is in the order of hundreds.

The curves \( \Delta = f(n_f) \) and \( \Psi = f(n_f) \) are pseudo-periodic for films with thicknesses comparable to wavelengths.

Figures 4 and 5 show that there may be a great uncertainty in determining \( d_f \). For example all films with multiple thicknesses of 200 nm and \( n_f = 1.65 \) have the same \( \Delta \) and \( \Psi \).

3. The choosing experimental conditions in an ellipsometric measurement in order to obtain minimal errors in determining the refractive indices of superficial films

The ellipsometric parameters \( \Delta \) and \( \Psi \) may have greater or smaller variations with the change of the refractive index \( n_f \) of the superficial film depending on: the thickness \( d_f \) of the film, the angle of incidence \( \varphi_0 \) during the ellipsometric measurement or the type of the substrate on which it is located (expressed by the value of the refractive index \( n_s \) of the substrate). The analysis of how \( \Delta \) and \( \Psi \) vary when changing these parameters allows their optimal choice to obtain minimal errors in determining the refractive index of the superficial film [12].

This paper presents an evaluation of errors in the determination of the refractive index \( n_f \) of superficial films due to experimental errors of measurement of \( \Delta \) and \( \Psi \).

3.1. Films with thicknesses in the order of magnitude of the wavelengths

The precision in determining the refractive index \( n_f \) of the superficial film is even better as the thickness of the film is larger, as can be seen from Figures 4 and 5. The variation of \( \Delta \) and \( \Psi \) versus the refractive index \( n_f \) of the superficial film is higher. Thus, for films with a thickness of 100 nm the period of variation of \( n_f \) is 2.8 and for films with a thickness of 1000 nm the period of variation of \( n_f \) is 0.26. For films with thicknesses of 300 nm or 500 nm the variations of \( \Delta \) and \( \Psi \) versus \( n_f \) are quite large for superficial films with \( n_f = 1.55 \div 1.65 \) as shown in Figure 6. At 200 nm and 400 nm the curves \( \Delta = f(n_f) \) show a minimum around \( n_f = 1.65 \). The variation of \( \Psi \) with \( n_f \) is very small so that errors in the measurement of \( \Psi \) cause great errors in the determination of \( n_f \).

In order to see how much is influenced the determination of \( n_f \) by the experimental errors in the measurement of \( \Delta \) and \( \Psi \) the derivatives of the refractive index \( n_f \) of the superficial film according to the ellipsometric parameters \( \Delta \) and \( \Psi \) are analyzed. For large variations of \( \Delta \) and \( \Psi \) versus the refractive index \( n_f \) of the superficial film, small errors are obtained in the determination of the refractive index.

The derivatives of refractive index \( n_f \) of the superficial film according to \( \Delta \) and \( \Psi \) are shown in Figure 7.

In determining the refractive index \( n_f \) of the superficial film, for some film thicknesses and for certain refractive index values, large errors are obtained. If errors in determining the refractive index \( n_f \) of the superficial film are too high, some experimental conditions, such as incidence angle or incident wavelength, may be changed in order to obtain lower errors.
The variation of Δ and Ψ values for the refractive index $n_f$ of the superficial film in the range $n_f = 1.3 \div 1.7$; ($n_i = 2.67(1-1.18i)$; $\phi_0 = 60^0$; $\lambda = 562.5$ nm)

Figure 6. The first derivative of refractive index $n_f$ of the superficial film according to Δ and Ψ; $n_i = 2.67(1-1.18i)$; $\phi_0 = 60^0$; $\lambda = 562.5$ nm

Figure 7. The first derivative of refractive index $n_f$ of the superficial film versus Δ or Ψ; $n_i = 2.67(1-1.18i)$; $\phi_0 = 60^0$; $\lambda = 562.5$ nm

3.2. Thin film thicknesses of the nanometer size order

In the range of the refractive index values of usual materials $n_f = 1.3 \div 1.7$, the variation of Δ and Ψ with the refractive index is small so that the experimental errors of the Δ and Ψ produce quite high errors in the appreciation of the $n_f$ or $d_f$ values.

Variations of Δ and Ψ versus the refractive index of the superficial film, for thin films are shown in Figure 8.

The first derivatives of the refractive index $n_f$ of the superficial film versus Δ or Ψ for films with thickness of the order of magnitude of nanometers or tens of nanometers are shown in Figure 9.

For thin films with thicknesses in the order of nanometer tens, there is an error of 0.01 in the determination of $n_f$ when the error in the measurement of Δ is 0.2 degrees. Large errors can occur due to the experimental measurement of the ellipsometric parameter Ψ. In this case, some experimental conditions such as incidence angle or wavelength of incidence radiation should be modified.

For nanometer-sized film thicknesses, the same experimental measurement errors of Δ and Ψ will produce errors in the determination of $n_f$ approximately 10 times higher, since the variation of the ellipsometric parameters Δ and Ψ with the refractive index of the superficial film is smaller.
Figure 8. The variation of Δ and Ψ values for the refractive index $n_f$ of the superficial film in the range $n_f = 1.3 \div 1.7$; ($\tilde{n}_s = 2.67(1-1.18i); \varphi_0 = 60^\circ; \lambda = 562.5$ nm)

Figure 9. The first derivative of refractive index $n_f$ of the superficial film according to Δ and Ψ; $\tilde{n}_s = 2.67(1-1.18i); \varphi_0 = 60^\circ; \lambda = 562.5$ nm
3.3. The influence of the incidence angle on the shape of the curves $\Delta = f(n_f)$ and $\Psi = f(n_f)$

The value of the incidence angle in the ellipsometric measurement may be changed to avoid large errors in determining the refractive index $n_f$ of the superficial film due to a small variation of the ellipsometric parameters $\Delta$ and $\Psi$ versus the refractive index of the film. The change in the appearance of the curves $\Delta = f(n_i)$ and $\Psi = f(n_i)$ with the change of incidence angle is shown in figure 10. If for a particular superficial film the error in the determination of the refractive index $n_i$ is too high, the slope of the curves $\Delta = f(n_i)$ and $\Psi = f(n_i)$ can be increased by changing the incidence angle. Thus, areas with large errors can be avoided.

![Figure 10](image1.jpg)

**Figure 10.** The variation of $\Delta$ and $\Psi$ depending on the refractive index $n_i$ of the superficial film for different incidence angle; $n_s = 2.67(1-1.18i)$; $d_f = 500\text{nm}$; $\lambda = 562.5\text{ nm}$

3.4. The influence of the substrate refractive index $n_s$ on the shape of the curves $\Delta = f(n_f)$ and $\Psi = f(n_f)$

The change in the appearance of the curves $\Delta = f(n_i)$ and $\Psi = f(n_i)$ with the change in the nature of the substrate on which the superficial film lies for two different thicknesses of the superficial film is shown in Figure 11. Changing the nature of the substrate can change the range in which the error in determination of the refractive index $n_i$ of the superficial film is high.

![Figure 11](image2.jpg)

**Figure 11.** The variation of $\Delta$ and $\Psi$ depending on the refractive index $n_i$ of the superficial film for different substrates with different refractive indices ($d_f = 400 \text{ nm and 500 nm}$; $\phi_0 = 60^\circ$; $\lambda = 562.5 \text{ nm}$)

All calculations were performed based on the Fortran's McCrackin program, modified accordingly and the graphics were processed in Matlab.

4. Conclusions

The ellipsometric analysis of the specular external reflection of light radiation on superficial films optical non-absorbing allows to know the experimental factors on which the analyzed system depends and which influence the error of determining the refractive index of superficial film. For these films the curves $\Delta = f(n_i)$ and $\Psi = f(n_i)$ are pseudo-periodic and curves $\Delta = f(\Psi)$ are closed.
The analysis of the periodicity of these curves allows accurate determination of the refractive index of superficial films. Conclusions on areas where experimental measurement errors of the ellipsometric parameters $\Delta$ and $\Psi$ produce small errors in the determination of the refractive index $n_f$ of the superficial film can be obtained from the curve shape.

The experimental parameters: incidence angle $\varphi_0$, wavelength $\lambda$, and refraction index of incidence medium $n_0$ can be properly modified to obtain minimal errors in determining the refractive index of superficial film.

It is important to perform an evaluation of errors in the determination of the refractive index $n_f$ of superficial films due to experimental errors of measurement of $\Delta$ and $\Psi$.

Information from the shape of the curves in the areas where experimental errors in the measurement of ellipsometric parameters $\Delta$ and $\Psi$ produce small errors in determining the refractive index $n_f$ of a superficial film was obtained.

Changing experimental conditions in an ellipsometric measurement such as: the incidence angle $\varphi_0$ or the nature of the substrate on which the superficial film lies (expressed by the value of the refractive index $n_s$ of the substrate) can avoid large errors in determining the refractive index $n_f$ of superficial film.

References

[1] Azzam R M A and Bashara N M 1977 Ellipsometry and Polarized Light, North-Holland publ. Comp., Amsterdam - New York - Oxford
[2] Jitian S 1994 Modification de l’état de polarisation de la lumière à la réflexion sur les surfaces solides, Bul. St. Univ. Politehnică Timișoara 39(53) 101-106
[3] Moisil G and Moisil D 1973 Teoria si practica elipsometriei, Ed. Tehnica, Bucuresti
[4] Jitian S and Chifu E 1986 The Ellipsometric Study of Polimer Films on Metals, Studia Univ. Babeș-Bolyai. Chemia 31(2) 69-75
[5] Chen K H, Hsu C C and Su D C 2003 A method for measuring the complex refractive index and thickness of a thin metal film, Appl. Phys. B – Lasers and Optics 77 839-842
[6] Azzam R M A and Khan M E R 1983 Complex reflection coefficients for the parallel and perpendicular polarizations of a film–substrate system, Applied Optics 22(2) 253-264
[7] Jitian S 2013 The ellipsometrical study of adsorption-desorption of the corrosion inhibitors on metallic surfaces, Romanian Reports in Physics 65(1) 204-212
[8] Jitian S 2015 The Ellipsometrical Analysis of External Reflection of Light on Superficial Films on Solid Substrates, American Journal of Optics and Photonics 3(4) 48-53
[9] Jung T H J, Bork J, Holmgaard T and Kortbek N A 2004 Ellipsometry. Detection of nanostructures, Technical report, Aalborg University, Institute of Physics and Nanotechnology, Aalborg
[10] Pristinski D, Kozlovskaya V, and Sukhishvili S A 2006 Determination of film thickness and refractive index in one measurement of phase-modulated ellipsometry, J. Opt. Soc. Am. A 23(10) 2639-2644
Manallah A and Bouafia M 2016 Determination of Optical Constants of Semiconductor Thin Films by Ellipsometry, International Science Index, Materials and Metallurgical Engineering 10(7) 927-930
[11] Chandler-Horowitz D, Nguyen N V and Ehrstein R J 2003 Assessment of Ultra-Thin SiO₂ Film Thickness, Measurement Precision by Ellipsometry, CP683, Characterization and Metrology for VLSI Technology. 2003 International Conference, edited by D. G. Seiler, A. C. Diebold, T. J. Shaffner, R. McDonald, S. Zollner, R. P. Khosla, and E. M. Secula, American Institute of Physics 0-7354-0152-7/03, pp 326-330
[12] McCrackin F L 1969 A Fortran Program for Analysis of Ellipsometer Measurement, Natl. Bur. Std., Technical Note 479, Washington, USA