THE PUBLISHING HOUSE
OF THE ROMANIAN ACADEMY

PROCEEDINGS OF THE ROMANIAN ACADEMY, Series A,
Volume 9, Number 2/2008, pp. 000-000

ELLIPSOMETRIC ARRANGEMENT FOR THIN FILMS CONTROL

A. BELDICEANU, A. RIZEA

S.C. Pro Optica S.A., Bucharest, Romania
Corresponding author: ancabel1@yahoo.com

An ellipsometric experimental set up of PSA type with an extinction factor E.F.= 10^{-5}, inspired from the Gaertner's L119 and a powerful BASIC software for the theoretical calculus of the *ellipsometric parameters* ψ and Δ*, including original practical* solutions, were developed. Encouraging results were obtained in applying the simple ellipsometric method of azimuths to determine the optical constants: refractive index n, absorption index k and geometrical thickness d, for the polished film on BK7 and quartz glass substrates, with this new optoelectronic device.

Key words : Ellipsometry, Polishing film, Optical constants, Thin films

1. INTRODUCTION

In the seventies the azimuthal ellipsometry (AE) was developed and applied only for metallic surfaces, with the following advantages over the classical null ellipsometry (NE) [1, 2]: an efficient method for the alignment of the components, reduced sources of optical and mechanical errors, the possibility to adapt the azimuthal range with the nature of the sample, and the possibility of a simple modulation of measurements

In optoelectronics the optical constants of the polished films have to be known first of all in order to deposit thin films on dielectrics. The ellipsometry gives the possibility of determining n and d from the changes produced by reflection or transmission in a plane monochromatic linearly polarized light.

The changes in the polarization ellipse of light are described by the *ellipsometric parameters* ψ and Δ.. The fundamental ellipsometric equation gives the relation between ψ and Δ and the complex reflection or transmission Fresnel coefficients R_p and R_s/T_p and T_s, for a system of thin homogeneous isotropic layers of refractive index n_f, and geometrical thickness, d, on a plane surface of known index of refraction n_s.

We proceeded to the transformation of an old Askania goniometer in an ellipsometer inspired from the Gaertner's L119 and the development of a powerful software. For the validation of our laboratory arrangement and *method of calculation* for n and d in the case of a single layer on a transparent substrate, a number of tests and determinations on a BK7 disc and on a fused quartz fresh polished plate were done. In comparison with any kind of optical glass, flint or crown, [3], the quartz has the best chemical stability and many results are reported on *polished films* [1, 3, 4]. The aging process is also slower for quartz than for any crown or flint glass.

The polished film or layer on an optical glass surface, quartz glass included, was studied by many authors, especially by japanese scientists [3, 7]. In the domain of transparent optical materials, fused silica and BK7 are the most important ones.

We intend to show that a simple ellipsometric method for determining optical constants n and d of thin films matches laboratory conditions and measuring demands, if some experimental and calculation solutions are taken into account.

1. ELLIPSOMETRY

2.1 Theory

At the extinction of the polarized reflected or transmitted light, i.e. for the intensity of light $I = 0$, one can write the simple mathematical relations, (1), (2) between the measured azimuths θ and γ of the polarizing prisms and the ellipsometric parameters ψ and Δ of the sample [1, 2].

$$tg\,\psi = \sqrt{tg\,\vartheta_1 \cdot tg\,\theta_2} \cdot \sqrt{\frac{tg\,\theta_2 \cdot tg\,2\gamma_2 - tg\,2\gamma_1 \cdot tg\,\theta_1}{tg\,\theta_1 \cdot tg\,2\gamma_2 - tg\,2\gamma_1 \cdot tg\,\theta_2}} \tag{1}$$

$$\cos\Delta = tg\,2\theta_1 \cdot \frac{tg^2\psi - tg^2\theta_1}{2tg\,\psi \cdot tg\,\theta_1} \tag{2}$$

In turn, ψ and Δ depend on the optical constants (the refractive index **n,** the absorbtion index **k** and geometrical thickness **d** of the thin film and on the optical constants of the bulk material, by transcendental relations: the fundamental equation of ellipsometry, written with the p and s Fresnel complex coefficients of reflection and transmission, [5], with long exponential terms, is given here in its simple form, (3) and (4).

$$tg\,\psi_r \cdot e^{i\Delta} = \frac{R_p}{R_s} \tag{3}$$

$$tg\,\psi_t \cdot e^{i\Delta} = \frac{T_p}{T_s} \tag{4}$$

2.2 Modeling

The model adopted for the polished layer formed on optical materials during different processes like aging, chemical reactions, technological operations, etc., *is the ideal transparent homogeneous and isotropic thin film on a plane surface*.

The exact formulae for the Fresnel coefficients for p and s polarized components of light that meets one or more thin films on a glass substrate, in reflection or transmission, are developed with Matrix calculus, and exact Ψ and Δ values are readily obtained.

A Visual Basic software for the calculus of Ψ and Δ values, is used to determine n, k and d in the case of one or more thin films on optical support, by *graphical interpolation* from theoretical curves $\Psi = \Psi$ (n,d) with the measured Ψ and Δ values.

Theoretical curves Ψ (n_1), Ψ (n_2), etc., with d_1, d_2, d_3 as a parameter are obtained with different values choosen for the pair (n, d) in the domain of magnitude that we are waiting for, based on earlier reported experiments [4], or not, for the type of optical glass that is under ellipsometric control (Fig.1b).

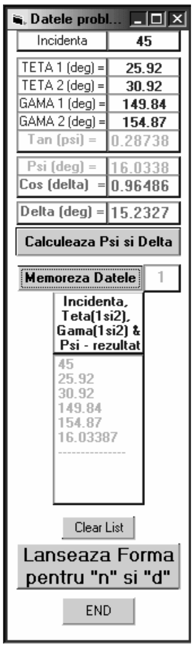
The experimental values for Ψ and Δ are calculated with another program from equations (2.1), (2.2) with the measured azimuths of the polarizer θ_1 and θ_2, and the corresponding azimuths of the analyzer, γ_1 and γ_2, at the minimum of light intensity (Fig.1a).

2.3 Experimental set up

The experimental laboratory arrangement of PSA type is given below in Fig. 2.

Our experimental arrangement was developed with two Glan Thompson prisms from C.Z.Jena, mounted in very precise large diameter rotating circles, a sensitive detection system of Pritchard type, from Kollmorgen Corporation, and other opto-mechanical high quality accessories, to solve the problems of stray light which arises from scattering and induced anisotropy in the optical path of light.

The new apparatus has no longer the $\lambda/4$ compensator, which introduces, due to its intrinsic sensitivity, many experimental errors.



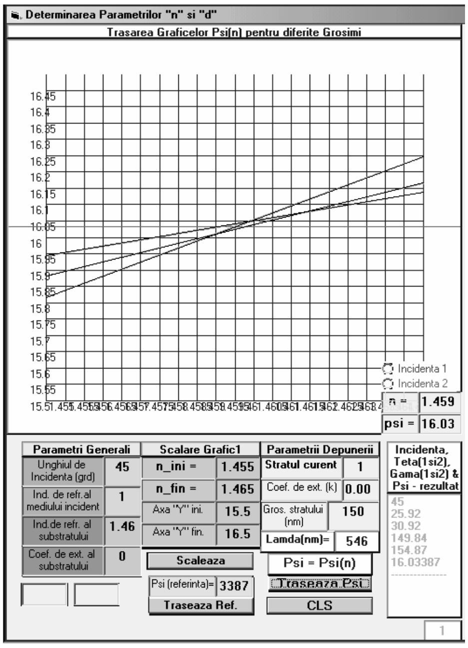
Incidenta = 45

TETA 1 (deg) = 25.92
TETA 2 (deg) = 30.92
GAMA 1 (deg) = 149.84
GAMA 2 (deg) = 154.87
Tan (psi) = 0.28738
Psi (deg) = 16.0338
Cos (delta) = 0.96486
Delta (deg) = 15.2327

Memoreza Datele: 1

Incidenta, Teta(1si2), Gama(1si2) & Psi - rezultat
45
25.92
30.92
149.84
154.87
16.03387

Figure 1a



n = 1.459
psi = 16.03

Parametri Generali	
Unghiul de Incidenta (grd)	45
Ind. de refr.al mediului incident	1
Ind.de refr. al substratului	1.46
Coef. de ext. al substratului	0

Scalare Grafic1	
n_ini =	1.455
n_fin =	1.465
Axa "Y" ini.	15.5
Axa "Y" fin.	16.5

Psi (referinta)= 3387

Parametrii Depunerii	
Stratul curent	1
Coef. de ext. (k)	0.00
Gros. stratului (nm)	150
Lamda(nm)=	546

Incidenta, Teta(1si2), Gama(1si2) & Psi - rezultat
45
25.92
30.92
149.84
154.87
16.03387

1

Figure 1b

The equipment consists of the basic apparatus and the accessories noted bellow:

-a precision Askania goniometer - spectrometer with quality telescope and collimator objectives aberrations corrected and mechanically precise rotation movements.

-two polarisation high quality prisms (ADP Glan-Thompson prisms), mounted in rotating divided 120 mm diameter metal circle assemblies.

-an OSRAM spectral lamp with vapors of Hg and Cd, 1A, with an interferential filter for λ= 546.1 nm; both of them are attached in the focal plane of the 500 mm focal length collimator.

-a Spectra Pritchard type photometer ("aperture photometer") with 12 decades of amplification that permits measurements at the very low level of illumination where the extinction of light (crossed polarizers) is reached; the original optical fiber of this apparatus has another important feature: it is characterized by lacking of polarization effects and so we can collect all light in the exit pupil of the apparatus, to ensure sensitivity, without uncertainty.

For testing the correctness of our set up we two plane parallel faces plates: one of BK7 and the other of amorphous quartz of SQ type from C.Z. Jena, recognized in the practice of ellipsometry for its very thin polishing film; the second face is not polished, in order to avoid a second reflection to be also measured .

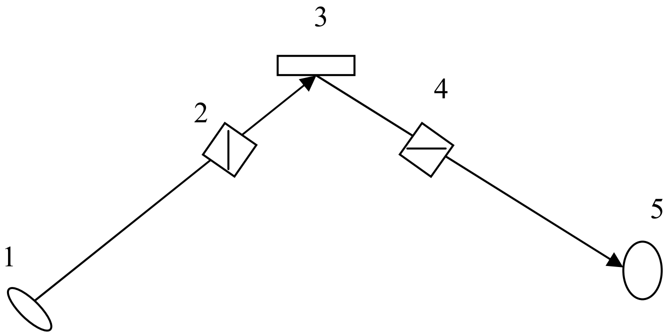


Figure 2

The PSA type scheme contains the components 1-5 specified below.

1- monochromatic and collimated light source;
2- Glan - Thompson polarizing prism (P);
3- rotating table with reflecting sample (S);
4- Glan - Thompson analysing prism (A);
5- Spectra Pritchard photometer

3. RESULTS

3.1. Method

Generally, from two measured quantities, Ψ and Δ, one determines the unknown n and d of a thin film by interpolation of theoretical curves, $\Psi=\Psi(n)$ and $\Delta=\Delta(n)$, d as a parameter; for n and d one gives the values in an expected range for the glass under test.

The measured Ψ and Δ for two selected values of the azimuth of the polarizer Θ_1, Θ_2 and for the two corresponding determined values of the azimuth of the analyzer, γ_1, γ_2, at the point where the extinction of light is detected, are calculated; Θ_1 and Θ_2 are taken in the range of maximum sensitivity of the method, differing with the nature of the substrate and of the film.

Instead of the measured value of Δ, unstable in not very clean environments, another parameter, ψ measured at a new angle of incidence of light, is introduced. Two experimental parameters are in this way available in order to determine two unknown constants: n and d for the polishing film. The bulk values for the refractive index of quartz, ns, are taken from the literature and are also measured with a Pulfrich refractometer, for $\lambda = 546.1$ nm.

The alignment procedure for our ellipsometer follows Steel, [4].

3.2. Measurements

a) The E. F. extinction factor measured on the whole set up has a good value, situated between 5×10^{-6} and 1×10^{-5}.

b) A set of readings and the final results for the polished film on amorphous quartz plate.
For two values of the incidence angle $\alpha_i^1=45°$ and $\alpha_i^2=50°$ the corresponding values obtained for the ellipsometric parameter ψ are :

$\psi^1=16.03°$ and $\psi^2=12.61°$

The pairs of experimental parameters, fixed (Θ) and varied (γ), in order to reach the extinction of light, from which we have calculated the above ellipsometric parameters, are:

$\Theta_1=25.92°$ $\gamma_1 = 149.84°$
$\Theta_2=30.92°$ $\gamma_2 = 154.87°$
$\Theta_1=25.92°$ $\gamma_1 = 156{,}25°$
$\Theta_2=30.92°$ $\gamma_2 = 160.48°$

If we suppose the values for the optical constants of bulk glass given in the catalog C.Z. Jena to be true, at the wavelength $\lambda = 546.1$ nm

$n_s=1.46000$ and we assume $k_s=0.001$

we obtained the following measured optical constants for the thin polishing film:

$n_f=1.459$, $k_f=0.0001$, $d_f=100$ nm (1000A)

c) A set of readings and the final results for the polishing film on BK7 glass plate

At an angle of incidence $\alpha_i = 56°$ and with the index of refraction $n_s = 1.5183$ the experimental readings were:

$\Theta_1= 42.3°$ $\gamma_1 = 2.3°$
$\Theta_2= 67.3°$ $\gamma_2 = 11.3°$

The corresponding ellipsometric parameters are:

$\psi = 1.32°$ $\Delta = 3.7°$

The optical constants obtained for the thin polishing film are:

$n_f=1.525$ $d_f=89.9$ nm

The precision in the determination of the angle of incidence of light, was 20″.

The accuracy for the azimuths P and A was of one minute of arc for 0°- 360°.

4. DISCUSSION

To evaluate the performances of any polarimeter or ellipsomter one has to measure the quality factor known as the extinction factor, E.F., of the arrangement, defined as the ratio I_{min}/I_{max}, where I_{min} and I_{max} are the values of the intensity of light for polarizers in cross and in parallel positions. The E.F.=10^{-5} includes these experimental arrangement in the first class of polarimeters; we mention that for two polarizing prisms the E.F. is 10^{-6} and for medium polarimeters is 10^{-4}.

First of all we found some simple direct ways to verify the correctness of the experimental set up.

a) In the calculus, with two different sets of measured azimuths, at the same angle of incidence, named 1 and 2, the two members of the equality (4.1), derived from (2.1) and (2.2), M_1 and M_2, are not allowed to differ:

$$\sigma_2(ctg\theta_1 - tg\theta_2)\cdot tg2\gamma_1 = \sigma_2(ctg\theta_2 - tg\theta_2)\cdot tg2\gamma_2 \tag{5}$$

where $\sigma_2 = tg^2\psi$

The discrepancies between the two members of the equality (5), resulted from the measured θ and γ are always less than the fifth decimal, respectively: M_1=0.5547145 and M_2=0.5546617.

b) An encouraging immediate result was that the measured value of Ψ at the angle of incidence of 45° is 17.06 ° compared with the reported value in [2], which is 17° 2'=17.03°; in both cases the probe under test was fresh polished quartz glass.

For testing the method of calculus we made determinations on fused silica and BK7.

a) The not aging optical material known as quartz glass has a polishing layer with n situated between 1.461 and 1.467; d has reported values between 350 Å and 650 Å [3]. Different sets of readings, for differently washed surfaces gave us also for n greater value than the bulk one, respectively 1.465 ; for d we obtained less than 1000 Å , with a supposed absorptivity , k= 0.015.

b) From earlier experiments on a NE ellipsometer, [4], on a BK7 optical glass plate with very clean polished surfaces designed for laser cavity mirrors, and also from [6], we know that the ellipsometric parameter Δ has a high sensitivity to the method for cleaning the probe.

We compared our present results with those obtained on a BK7 disc determined by NE from the measured data with a PCSA ellipsometer, in the model of a single or double film for the polishing layer [4, 8] and with the results reported by H. Sakata [3]. The discrepancies in n may be the result of the age of the surface, of the technique of polishing and is for sure the result of the method of cleaning the surface. And of course the uncertainty in n is important. For BK7 n_f is generally smaller that the n_s and this is caused by the uncertainty in the determination of Δ.

The final estimation for n and k is made with two exact decimals for BK7 and with 5×10^{-3} for fused silica; the sensitivity for the thickness d is about 5-10 nm.

The desired reproducibility (of the ellipsometric measurements for the angle of incidence and for the azimuths) is ensured by the mechanical complex movements on the basic Askania goniometer and from the rotating large polarization circles. The mechanical precisions are, compared to those of the Gaertner ellipsometer: 1′/ 0.6′ for the azimuths, and identical, respectively 20″, for the angle of incidence of light.

5. CONCLUSIONS

Encouraging results were obtained in applying the simple ellipsometric method of azimuths, developed by [1, 2] for metallic surfaces, in order to determine the optical constants, refractive index n, absorbtion index k and geometrical thickness d, for the polishing film on BK7 and quartz glass dielectric substrates.

An ellipsometric experimental set up of PSA type with an extinction factor E.F.= 10^{-5} and a powerful BASIC software for different applications, with any number of thin films, in reflected or transmitted light, were developed in our optical measurements and vacuum laboratories.

For fresh polished quartz, the optical constants, determined at two angles of incidence, are in good agreement with the reported ones, [3, 7].

For old glass, polished and cleaned with the methods of IOR – Pro Optica, from measurements at one angle of incidence, repeated on two different ellipsometers [8], the values for n_f are a bit greater than the values for $n_{s,}$ not respecting the reported ones, [3, 4, 7]. Refining the technique of cleaning the optical surfaces and also the measuring method, will give us the possibility to become sure that by polishing BK7, the value for n_f becomes greater than that of n_s .

A difficult problem in the development of any ellipsometric method is a theoretical one, namely the elaboration of a program for determining by matrix calculus, in an efficient and elegant way, the ellipsometric parameters ψ and Δ, and finally the optical constants, n, d, k of thin surface films.

Also for an ellipsometrist, a difficult experimental task is to apply the method for the study of dielectric with weak reflection probes, we mean a method with good results for strongly reflecting metallic surfaces [1, 2].

Both described problems, the weak light signal from a glass surface and the dedicated software were resolved. We can also say that the contribution of stray unpolarized light is diminished in our measurements, as long as we eliminated the delta sensitive parameter.

REFERENCES

1. SOM S.C., CHOWDHURY C., Journal of Optical Society of America, 61, 665A, 1971.
2. MONIN J., BOUTRY G. A., Nouvelle Revue d'Optique, 3, Tome 4, 1973.
3. SAKATA Hironobu, Jap. Journ. of Appl. Phys., 2, 12, 1973
4. BRATESCU G., ZAMFIR E., Z. Mares, BELDICEANU A., DINU B., BUHAI A., Analele Univ. Buc., 1977.
5. VASICEK A., Optics of thin films, London, 1970, second edition.
6. MOISIL G., MOISIL A., Teoria si practica elipsometriei (Theory and practice of the ellipsometry, roum.), Ed. Tehnica,1969.
7. KINOSITA Koreo, NISHIBORI Minao, YAMAMOTO Masaki, Optica Acta, Vol.17,No. 2, 116-112, 1970
8. GHITA L., BELDICEANU A., Research Report, in Romanian, unpublished, Bucharest, 1982.

Received February 26, 2008