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LASER ELLIPSOMETRIC METHODS FOR THE OPTICAL ANALYSIS OF THIN FILMS

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Abstract. Modern laser ellipsometric methods for analysing various multilayer systems are described in this paper. The basic features of the methods of multiple angle of incidence ellipsometry, combined reflection and transmission ellipsometry and combined immersion and multiple angle of incidence ellipsometry are specified in detail together with examples of their use in the complete optical analysis of chosen samples of different thin film systems.

Резюме. Описаны современные методы лазерной эллипсометрии для анализа многослойных систем. Подробно рассмотрены специфические особенности метода многоугловой эллипсометрии, метода отражательной эллипсометрии в комбинации с эллипсометрией пропускания и метода иммерсионной в комбинации с многоугловой эллипсометрией. Приведены примеры приложения этих методов для полного эллипсометрического анализа конкретных образцов разных многослойных систем.

1. Introduction

Methods of laser monochromatic ellipsometry are often employed for analysing various multilayer systems. In particular they are utilized for determining the values of the optical parameters characterizing both nonabsorbing or absorbing single and double layers placed on various substrates. The methods of the laser monochromatic ellipsometry can be used to evaluate the values of all optical parameters of the multilayer systems (e.g. the single and double layers); they are very useful from the practical point of view. These methods have been developed extensively during the last several years. It is significant that these methods for the complete optical analysis of the systems under consideration can be realized with relatively simple experimental arrangements as compared with those employed by methods of spectroscopic ellipsometry.

In this paper the most important methods of laser monochromatic ellipsometry will be presented separately.

2. Multiple angle of incidence ellipsometry

This method is based on the interpretation of the measured dependences of the system's ellipsometric parameters ψ (azimuth) and Δ (phase change) on the

angle of incidence φ_0 at a chosen wavelength λ of incident light in air (see e.g. ref. [1-3]). In this method it is necessary to carry out a minimization of the quantity

$$Q = \sum_{j=1}^N \left\{ \left(\frac{\psi_j - \psi'_j}{\delta_j} \right)^2 + \left(\frac{\Delta_j - \Delta'_j}{\delta_j} \right)^2 \right\} \quad (1)$$

in the space whose vectors have components identical with the optical parameters sought. In eqn. (1) the following substitutions are used:

$$\psi_j = \psi(\varphi_{0j}) \text{ and } \Delta_j = \Delta(\varphi_{0j}),$$

where φ_{0j} denotes the angle at which the experimental values of ψ and Δ are measured (N denotes the number of these angles), ψ'_j and Δ'_j are the experimental values corresponding to ψ_j and Δ_j , respectively, and δ_j and δ'_j are the standard deviations corresponding to ψ'_j and Δ'_j . The values of the optical parameters corresponding to the minimum of Q are considered to be the true ones. The minimization of the quantity Q can be performed by many least-squares procedures (e.g. using the procedure presented by Marquard [4]). It should be noted that in these least-squares procedures the standard deviations of the optical parameters found can be determined (see e.g. ref. [5, 6]). The theoretical values of the ellipsometric parameters, i.e. the values ψ_j and Δ_j , are calculated by means of the following equation:

$$\tan \psi e^{i\Delta} = \frac{\widehat{R}_p}{\widehat{R}_s}, \quad (2)$$

where \widehat{R}_p and/or \widehat{R}_s is the complex Fresnel coefficient of the system studied corresponding to p and/or s polarization of reflected light. For example, for an absorbing single layer located on an absorbing substrate the coefficients \widehat{R}_p and \widehat{R}_s are expressed as follows:

$$\widehat{R}_l = \frac{\widehat{r}_{1l} + \widehat{r}_{2l} e^{i\widehat{x}_l}}{1 + \widehat{r}_{1l} \widehat{r}_{2l} e^{i\widehat{x}_l}}, \quad (3)$$

where $l = p, s$, $\widehat{x}_l = \frac{4\pi}{\lambda} n_1 d_1 \cos \widehat{\varphi}_1$, $\cos \widehat{\varphi}_1 = \frac{1}{n_1} (\widehat{n}_1^2 - n_0^2 \sin^2 \varphi_0)^{1/2}$ and

$$\widehat{r}_{qp} = \frac{\widehat{n}_q \cos \widehat{\varphi}_{q-1} - \widehat{n}_{q-1} \cos \widehat{\varphi}_q}{\widehat{n}_q \cos \widehat{\varphi}_{q-1} + \widehat{n}_{q-1} \cos \widehat{\varphi}_q}, \quad (4)$$

$$\widehat{r}_{qs} = \frac{\widehat{n}_{q-1} \cos \widehat{\varphi}_{q-1} - \widehat{n}_q \cos \widehat{\varphi}_q}{\widehat{n}_{q-1} \cos \widehat{\varphi}_{q-1} + \widehat{n}_q \cos \widehat{\varphi}_q}, \quad (5)$$

$q = 1, 2$, $\widehat{n}_0 = n_0$, \widehat{n}_1 and $\widehat{n}_3 = \widehat{n}$ are the refractive index of the ambient ($n_0 = 1$ for air), the complex refractive index of the single layer and the complex refractive index of the substrate, respectively ($\widehat{n}_1 = n_1 + ik_1$, $\widehat{n} = n + ik$, n_1 and n and/or k_1 and k are the refractive and/or absorption indices of the single layer and substrate, respectively), d_1 denotes the thickness of the single layer, $\widehat{\varphi}_0 = \varphi_0$, $\widehat{\varphi}_2 = \widehat{\varphi}$ and

$$\cos \widehat{\varphi} = \frac{1}{n} (\widehat{n}^2 - n_0^2 \sin^2 \varphi_0)^{1/2}$$

If we know the values of n_0 , n and k we can evaluate, using MAI ellipsometry the values of n_1 , k_1 and d_1 , i. e. the values of the optical parameters of the film analysed. For example, we found the following values of n_1 , k_1 and d_1 , characterizing a chosen sample of the system formed by a glass plate and a thin gold film

$$n_1 = 0.3 \pm 0.1, k_1 = 3.2 \pm 0.1 \text{ and } d_1 = (35 \pm 4) \text{ nm.}$$

The value of the real refractive index n of the glass substrate was measured with a refractometer. It should be noted that the values of all the optical parameters in this paper correspond to the 632.8 nm wavelength. If the complete optical analysis of non-absorbing single layers is performed by MAI ellipsometry higher accuracy can be achieved in the determination of the optical parameters characterizing these layers. This statement can be illustrated through the results obtained by the MAI ellipsometric analysis of a chosen sample of the system consisting of anodic oxide film and substrate of GaAs single crystal.

$$n_1 = 1.817 \pm 0.005 \text{ and } d_1 = (107.2 \pm 0.9) \text{ nm.}$$

The values of the optical constants of the GaAs substrate were measured using an independent ellipsometric method [7].

In our laboratory multilayer systems were also successfully analyzed by MAI ellipsometry. In particular nonabsorbing double and triple layers can be characterized reliably with high accuracy. This statement is supported by the results obtained when analysing chosen samples of the double layers consisting of Si_3N_4 and SiO_2 films deposited onto the substrates of silicon single crystals, the double layer consisting of SiO_2 and TiO_2 films deposited on a glass plate substrates (see Table 1), the triple layers formed by Si_3N_4 , SiO_2 and MgF_2 films placed on the silicon single crystal substrates and the triple layers consisting of SiO_2 and TiO_2 films evaporated on the glass substrates (see Table 2). Table 1, row A lists the values of the optical parameters together with the values of their standard deviations characterizing the sample of the double layer $\text{Si}_3\text{N}_4/\text{SiO}_2$ (the Si_3N_4 film is the upper one). It should be noted that n_1 , d_1 and/or n_2 , d_2 denote the refractive index and the thickness of the upper and/or lower film, respectively. δn_1 , δd_1 , δn_2 and δd_2 denote the standard deviations of n_1 ,

Table 1. The values of the optical parameters and their standard deviations corresponding to chosen samples of the double layer (see text)

	n_1	δn_1	d_1 , nm	δd_1 , nm	n_2	δn_2	d_2 , nm	δd_2 , nm
A	2.012	0.004	112.8	0.3	1.459	0.003	205.0	0.4
B	1.47	0.02	99	6	2.05	0.03	195	5
C	2.25	0.01	124	1	1.451	0.008	143	2
D	1.46	0.01	160	3	2.27	0.02	81	2

Table 2. The values of the optical parameters and the standard deviations corresponding to chosen samples of the triple layer (see text)

	n_1	δn_1	d_1 , nm	δd_1 , nm	n_2	δn_2	d_2 , nm	δd_2 , nm	n_3	δn_3	d_3 , nm	δd_3 , nm
A	1.44	0.04	74.8	0.3	1.998	0.004	196.3	0.6	1.457	0.003	109.1	0.3
B	1.485	0.002	160	6	2.31	0.01	209	3	1.482	0.006	201	2

d_1 , n_2 and d_2 . In rows B—D of this Table the values of the same quantities are summarized for the chosen samples of the double layers $\text{SiO}_2/\text{Si}_3\text{N}_4$ (row B), $\text{TiO}_2/\text{SiO}_2$ (row C) and $\text{SiO}_2/\text{TiO}_2$ (row D). Table 2, row A and/or B list the values of the optical parameters together with their standard deviations for the chosen samples of the triple layers $\text{MgF}_2/\text{Si}_3\text{N}_4/\text{SiO}_2$ (the MgF_2 film is the upper one). The quantities n_1 , d_1 , δn_1 , δd_1 correspond to the upper film, etc. It should be noted that the values of the optical parameters introduced in both Tables 1 and 2 were also determined in an independent way by immersion spectroscopic reflectometry described in refs [8,9]. We found that the values of the corresponding optical parameters determined by both methods used agreed within the experimental accuracy. Moreover, it should be noted that MAI ellipsometry can be employed for analysing the double and triple layers studied (see Tables 1 and 2) successfully if the values of the optical parameters sought can be estimated with sufficient relative accuracy (about $\pm 20\%$) before the analysis (for details see ref. [10]). When the number N of thin films forming the system analysed increases ($N > 3$), certain difficulties arise in the complete analysis of this system by MAI ellipsometry. This fact is probably caused by correlation between the parameters sought. However, if nonabsorbing quarterwave stacks with arbitrary number of the films are analysed by MAI ellipsometry one can obtain reliable and precise values of the optical parameters characterizing these systems since the values of four optical parameters have to be found. This statement is supported by our results obtained by MAI ellipsometric analysis of a chosen sample of the seven-layer system $\text{H}(\text{LH})^3/\text{G}$ (H and/or L and G denote the layer with the high and/or low refractive index and the glass substrate, respectively), i.e.

$$n_{\text{H}} = 2.18 \pm 0.03, d_{\text{H}} = (77.8 \pm 7) \text{ nm}, n_{\text{L}} = 1.48 \pm 0.02, d_{\text{L}} = (115 \pm 1) \text{ nm},$$

where n_{H} and d_{H} and/or n_{L} and d_{L} denote the refractive index and the thickness of the TiO_2 and/or SiO_2 film, respectively.

It should be pointed out that the angular dependences of ψ and Δ of all the samples investigated were measured at angles of incidence in the 45° — 80° range.

3. Combined reflection and transmission ellipsometry

A method is known as combined reflection and transmission ellipsometry; it uses the simultaneous interpretation of the ellipsometric parameters for light reflected and transmitted on the air side and on the substrate side of layers. This method was used to perform a complete optical analysis of various single and double layers deposited on nonabsorbing substrates. This method is based on the minimization of the quantity Q defined as follows:

$$Q = \sum_{j=1}^N \left(\frac{F_j}{\delta_j} \right)^2, \quad (6)$$

where

$$F_j = \psi_{\text{RE}}(\varphi_{0i}) - \varphi'_{\text{RE}}(\varphi_{0i}) \quad j = i$$

$$F_j = \Delta_{\text{RE}}(\varphi_{0i}) - \Delta'_{\text{RE}}(\varphi_{0i}) \quad j = N_1 + i$$

$$F_j = \psi_{\text{RI}}(\varphi'_i) - \psi'_{\text{RI}}(\varphi'_i) \quad j = 2N_1 + i$$

$$F_j = \Delta_{\text{RI}}(\varphi'_i) - \Delta'_{\text{RI}}(\varphi'_i) \quad j = 3N_1 + i$$

$$F_j = \psi_{TE}(\varphi_{0i}) - \psi'_{TE}(\varphi_{0i}) \quad j = 4N_1 + i$$

$$F_j = \Delta_{TE}(\varphi_{0i}) - \Delta'_{TE}(\varphi_{0i}) \quad j = 5N_1 + i.$$

$i = 1, \dots, N_1$, $N = 6N_1$, N is integer, Δ' and ψ' denote experimental values corresponding to Δ and ψ , respectively and δ_j is the standard deviation corresponding to F_j . For example, $\psi_{RE}(\varphi_{01})$ is the experimental value of the theoretical quantity $\psi_{RE}(\varphi_{01})$ etc., and δ_1 is the standard deviation of $\psi_{RE}(\varphi_{01})$, etc. The symbols ψ_{RE} and Δ_{RE} , ψ_{RI} and Δ_{RI} , ψ_{TE} and Δ_{TE} represent the azimuth and phase change of the system studied for light reflected on the layers side, reflected on the substrate side and transmitted by the system when the light falls on this system on the layers side, respectively (for details see ref. [11–13]). Furthermore, (7) holds

$$\sin \varphi' = \frac{n_0}{n} \sin \bar{\varphi}_0 \tag{7}$$

where $\bar{\varphi}_0$ and/or φ' is the angle of incidence on the rear boundary of a nonabsorbing plate forming the substrate (the boundary without the layers) and/or the corresponding angle of incidence on the layers from the substrate side. By means of this method we analysed, for example, single gold layers deposited on the glass substrates. The results obtained for the chosen sample of thin gold film, the optical parameters of which are introduced above, are as follows:

$$n_1 = 0.21 \pm 0.01, \quad k_1 = 3.34 \pm 0.02, \quad d_1 = (30.6 \pm 0.3) \text{ nm}.$$

One can see that the values of n_1 , k_1 and d_1 are determined by combined ellipsometry more precisely than those evaluated by MAI ellipsometry. Moreover, the values of n_1 , k_1 and d_1 found by combined ellipsometry are probably more correct which is implied by the fact that the value of d_1 determined by Fizeau method is as follows: $d_1 = (30 \pm 2) \text{ nm}$ (for details see ref. [10]). Further, many nonabsorbing and absorbing double layers were studied by the combined ellipsometry. The results obtained for chosen samples of the MgF_2/Al , $\text{Al}_2\text{O}_3/\text{NiCr}$ and TiO/Au double layers are summarized in Table 3, rows A, B, C, respectively. One can see that the optical parameters of the MgF_2/Al and $\text{Al}_2\text{O}_3/\text{NiCr}$ are determined with high accuracy. This statement is also valid for the TiO/Au double layer with the exception of the value of the refractive index n_2 of the thin gold film. It should be noted that the accuracy of the optical parameters evaluated by combined ellipsometry is lower if the double layer studied is formed of two highly absorbing materials (e.g. by gold and nickel) (for details see ref. [12]).

For all samples analysed by combined ellipsometry, the angles of incidence φ_0 and $\bar{\varphi}$ were in the range $45^\circ - 80^\circ$, too.

Table 3. The values of the optical parameters and their standard deviations of the chosen samples of the double layer determined by the combined ellipsometry

	n_1	δn_1	k_1	δk_1	$d_1, \text{ nm}$	$\delta d_1, \text{ nm}$	n_2	δn_2	k_2	δk_2	$d_2, \text{ nm}$	$\delta d_2, \text{ nm}$
A	1.403	0.004	0	0	82.9	0.2	1.3	0.1	7.23	0.05	26	3
B	1.615	0.007	0	0	48.8	0.3	3.29	0.01	4.21	0.02	33.8	0.7
C	2.05	0.03	1.64	0.03	76	8	0.24	0.24	3.5	0.2	24	6

4. Combined immersion and multiple angle of incidence (IMAI) ellipsometry

This method is based on the simultaneous interpretation of the angular dependences of ψ and Δ of a system measured in air, and the values ψ and Δ of this system measured for one angle or more angles of incidence in several nonabsorbing ambients (e.g. in nonabsorbing liquids). Within the framework of IMAI ellipsometry the following quantity Q is minimized:

$$Q = \sum_{j=1}^N \left(\frac{f_j - f'_j}{\delta_j} \right)^2 + \left(\frac{g_j - g'_j}{\delta_j} \right)^2, \quad (8)$$

where

$$f_j = \psi(\varphi_{0j}) \quad j = 1, 2, \dots, K,$$

$$f_j = \bar{\psi}(n_{0i}) \quad j = K+i, \text{ where } i = 1, 2, \dots, N-K,$$

$$g_j = \Delta(\varphi_{0j}) \quad j = 1, 2, \dots, K,$$

$$g_j = \bar{\Delta}(n_{0i}) \quad j = K+i, \text{ where } i = 1, 2, \dots, N-K.$$

The symbols $\psi(\varphi_{0j})$ and $\Delta(\varphi_{0j})$ denote the azimuth and phase change of the system corresponding to the angle φ_{0j} in air respectively. The symbols $\bar{\psi}(n_{0i})$ and $\bar{\Delta}(n_{0i})$ denote the same quantities corresponding to the fixed angle of incidence when the sample is immersed in the chosen nonabsorbing liquid (we assume that one angle of incidence is used for measuring the parameters ψ and Δ in the liquids). f'_j and g'_j are the experimental values of f_j and g_j . δ_j and δ'_j represent the standard deviations of f'_j and g'_j , respectively. N and K are integers. K denotes the number of the angles of incidence for which the angles ψ and Δ are measured in air and $N-K$ denotes the number of immersion liquids in the immersion ellipsometry at the chosen angle of incidence.

By means of IMAI ellipsometry we analysed semiconductor surfaces covered by very thin native oxide films in particular. For example, for a chosen sample of silicon single crystal (a p -type wafer with (111) orientation and $\rho = 10 \Omega \cdot \text{cm}$) we obtained the following values of the optical parameters, characterizing the system formed by the silicon substrate and the native oxide film: $n = 3.879 \pm 0.001$, $n_1 = 1.52 \pm 0.02$ and $d_1 = (3.2 \pm 0.1) \text{ nm}$.

We assumed that the value of the absorption index k of the silicon substrate was known with the accuracy: $k = 0.02 \pm 0.01$ (for details, see ref. [14]). It is evident that the values of the optical parameters introduced are evaluated with relatively high accuracy (this statement is especially true for the refractive index n of the silicon substrate). It should be noted that the value of the refractive index n of silicon single crystal is in very good agreement with the values presented elsewhere (see e.g. ref. [15, 16]). If the existence of an interface layer between the silicon single crystal and its native oxide layer that was proved by several researchers ([15, 17, 18]) is respected we obtain the values of n_1 and d_1 as follows: $n_1 = 1.48 \pm 0.01$ and $d_1 = (3.0 \pm 0.2) \text{ nm}$ (the values of the optical constants of silicon and the interface layer together with the value of the thickness of this layer were assumed to be known [14]).

The measurements of ψ and Δ were performed in air at angles of incidence φ_0 in the 45° – 85° range. The angle of incidence on the sample placed in the cell containing the liquids was 70.03° . Water, acetone and toluene served as the nonabsorbing liquids.

5. Conclusion

In conclusion it should be noted that the ellipsometric parameters of all samples, whose optical parameters are presented here, were measured using a Gertner L119 ellipsometer. An He-Ne laser was used as light source ($\lambda = 632.8$ nm). ψ and Δ were measured with an accuracy of approximately 0.01° and 0.1° , respectively.

Of course, it is possible to use other laser as light source for ellipsometers utilized in practice. So far laser ellipsometry has been applied in visible and infrared regions mainly (e.g. CO_2 lasers are often utilized in the IR region).

Laser ellipsometry is more advantageous than classical monochromatic ellipsometry from the experimental point of view (thermic light sources such as mercury lamp are employed in classical ellipsometry).

The advantages of laser ellipsometry can be summarized as follows:

- 1) Laser beams are a powerful light flux so that by laser ellipsometry one can measure the ellipsometric parameters of thin film systems that are characterized by relatively low reflectance or transmittance.
- 2) Laser beams are highly collimated and monochromatic compared with those from thermic light sources. Thus, in laser ellipsometry, various systematic errors taking place in classical ellipsometry can be minimized (e.g. the errors connected with an adjustment of the angle of incidence).
- 3) Laser beams propagating through the ellipsometer can be very narrow. This fact allows to apply the method of combined reflection and transmission ellipsometry without any difficulties.

Of course, there are some disadvantages of the methods of laser ellipsometry. However, these disadvantages are not important. For example, speckle effects can complicate detection of a light flux (this effect can be minimized using a suitable geometrical arrangement of the ellipsometer).

Thus, one can see that the methods of laser ellipsometry are very helpful in many branches of applied research.

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