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# ELLIPSOMETRY OF THIN FILM WITH CONTINUOUS DISTRIBUTION OF INDEX OF REFRACTION

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**ABSTRACT--Mathematical formulations are given for treatment of ellipsometric data of thin films with continuous distribution of index of refraction. A quadratic model and a Gaussian model, together with the computational procedures, are presented.**

Ellipsometry, which may be characterized as reflection polarimetry or polarimetric spectroscopy, is the measurement of the effect of reflection on the state of polarization of light. Such measurements may be interpreted to yield the optical constants of the reflecting material or, when the reflecting material is a film-covered substrate, the thickness and optical constants of the film. In ellipsometric measurement of surface films, the index of refraction and the extinction coefficient of the film are also obtained.

Due to the difficulties in experimental arrangement and the complexity in mathematical treatment, the surface films are usually treated as a single-layer film with homogeneous distribution of the index of refraction throughout its depth. Such a single-layer model is not a very satisfactory representation of ion implanted surface films (Whichard et al., 1988). It is the goal of the present paper to develop a multi-layer formulation for ellipsometric measurement of thin films with continuous distribution of index of refraction throughout its depth.

## SINGLE-LAYER ELLIPSOMETRY

The principle of ellipsometric measurement of single-layer films is available elsewhere (e.g., Heavens, 1955; Archer, 1968). A brief account of basic ellipsometry is given here.

Referring to Fig. 1, an incident light beam is reflected from the interface of two optical media having indices of refraction $n_1$ and $n_2$, respectively. The angle of incidence and the angle of refraction are $\phi_1$ and $\phi_2$, respectively. The amplitude of the electric vector component of the incident beam in the plane of incidence is denoted by $E_{12}^{ip}$, while $E_{12}^{is}$ denotes the component perpendicular to the plane of incidence. The reflected components are denoted by $E_{12}^{rp}$ and $E_{12}^{rs}$, and $E_{12}^{tp}$ and $E_{12}^{ts}$ denote the transmitted components. The subscript 12 indicates that the amplitude is directly related to the incident beam from the medium 1 striking at the interface between the media 1 and 2. By solving the Maxwell's equations under the boundary conditions at the reflecting interface, the following Fresnel reflection and transmission coefficients for the two planes of polarizations can be obtained:

$$r_{12}^{p} = \frac{E_{12}^{rp}}{E_{12}^{ip}} = \frac{n_1 \cos\phi_2 - n_2 \cos\phi_1}{n_1 \cos\phi_2 + n_2 \cos\phi_1} \tag{1}$$

$$r_{12}^{s} = \frac{E_{12}^{rs}}{E_{12}^{is}} = \frac{n_1 \cos\phi_1 - n_2 \cos\phi_2}{n_1 \cos\phi_1 + n_2 \cos\phi_2} \tag{2}$$

$$t_{12}^{p} = \frac{E_{12}^{tp}}{E_{12}^{ip}} = \frac{2\, n_1 \cos\phi_1}{n_1 \cos\phi_2 + n_2 \cos\phi_1} \tag{3}$$

$$t_{12}^{s} = \frac{E_{12}^{ts}}{E_{12}^{is}} = \frac{2\, n_1 \cos\phi_1}{n_1 \cos\phi_1 + n_2 \cos\phi_2} \tag{4}$$

For an optically absorbing medium, the complex index of refraction of the substrate,

$$n' = n - ik \tag{5}$$

is substituted for $n_2$ in equations (1) through (4). The extinction coefficient, k, is related to the absorption coefficient $\alpha$ (in units of $cm^{-1}$) by

$$k = \alpha\lambda/4\pi \tag{6}$$

where $\lambda$ is the vacuum wavelength.

Figure 2 shows the reflection and transmission of a light beam by a single-layer thin film having a thickness $d_1$. Because there are two interfaces, the beam will be reflected many times between the two interfaces. In the following discussion, the superscripts p and s are omitted unless otherwise stated, because the formulation is valid for both polarizations. Assuming an unity amplitude and a zero phase angle for the incident beam, the resultant amplitude is given by (Heavens, 1955; Archer, 1968)

$$\rho_{02} = R_{02}\ \beta = \frac{r_{01} + r_{12}\,\alpha^2}{1 + r_{01}r_{12}\alpha^2} \tag{7}$$

$$\tau_{02} = T_{02}\ \gamma = \frac{t_{01}t_{12}\alpha}{1 + r_{01}r_{12}\alpha^2} \tag{8}$$

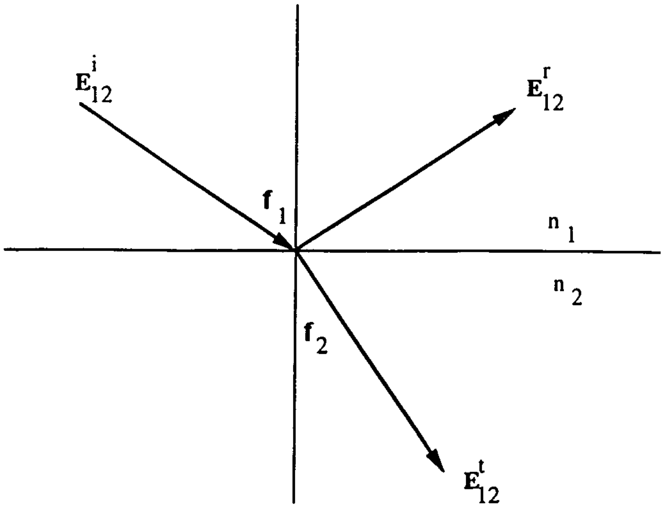


FIG. 1. Reflection, refraction, and transmission of a light beam at the interface of two media with the indices of refraction $n_1$ and $n_2$.

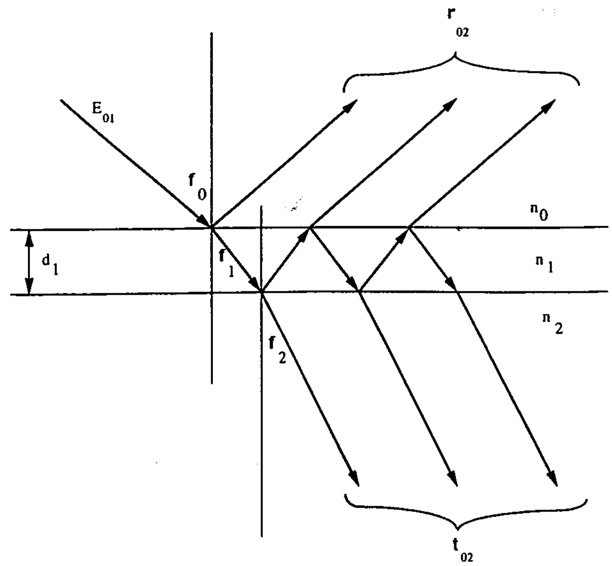


FIG. 2. Multiple reflection, refraction, and transmission of a light beam by a single-layer thin film.

where

$$\alpha = \exp[-(k_1 + in_1)\delta_1]. \tag{9}$$

The absorption coefficient is $k_1$, $n_1$ is the index of refraction,

$$\delta_1 = (2\pi d_1)/(\lambda \cos \phi_1), \tag{10}$$

$d_1$ is the thickness of the thin film, and $\lambda$ is the light wavelength. The resultant reflected and transmitted amplitudes are $R_{02}$ and $T_{02}$, respectively; $\beta$ and $\gamma$ are the complex phase angles. These two amplitudes and angles are measurable. The Fresnel coefficients ($r_{01}$, $r_{12}$, $t_{01}$, and $t_{12}$) are determined according to equations (1) through (4). The Fresnel coefficients are related to the experimentally measured parameters $\Psi$ and $\Delta$:

$$\Psi = \arctan \frac{R^pE^s}{R^sE^p} \tag{11}$$

$$\Delta = \beta^p - \beta^s \tag{12}$$

$$\frac{\rho_{02}^{\ p}}{\rho_{02}^{\ s}} = \tan\Psi \, \mathrm{Exp}[i\Delta] \tag{13}$$

where the superscript p denotes the parameters associated with the electric vector component in the plane of incidence, while the superscript s denotes that associated with the component perpendicular to the plane of incidence.

## MULTI-LAYER ELLIPSOMETRY

The reflection and refraction of a light beam by a multi-layer thin film is illustrated in Fig. 3. The boldface arrows represent the resultant light beams of multiple reflection and refraction. For example, $R_{0i}$ represents the superposition of all the light beams being reflected and refracted by the top i layers and finally coming out of the top surface. The recursion formulae for multi-layer thin film can be obtained in much the same way as equations (5) and (6) are derived:

$$\rho_{0,i+1} = \rho_{0,i} + \frac{\tau_{0,i}\,\tau_{i,0}\,r_{i,i+1}\,\alpha^2_{i+1}}{1 - \rho_{i,0}\,r_{i,i+1}\,\alpha^2_{i+1}} \tag{14}$$

$$\tau_{0,i+1} = \frac{\tau_{0,i}\,t_{i,i+1}\,\alpha^2_{i+1}}{1 - \rho_{i,0}\,r_{i,i+1}\,\alpha^2_{i+1}} \tag{15}$$

$$\alpha_{i+1} = \exp[-i\delta_{i+1}] \tag{16}$$

$$\delta_{i+1} = (360/\lambda)d_{i+1}(n_{i+1}^{\ 2} - \sin^2\phi_i)^{1/2} \text{ degrees} \tag{17}$$

where $\rho_{0,i}$ is the resultant reflection of the incident beam by the first i layers of the thin film, while $\rho_{i,0}$ is the resultant reflection of the beam entering from the i + 1th layer back into the first i layers. The subscripts of the rest of the symbols are likewise understood. The thickness and the index of refraction of the i + 1th layer are denoted by $d_{i+1}$ and $n_{i+1}$, and $\phi_i$ is the angle of incidence in the ith layer. If the absorption is not negligible, all $\delta_i$ are complex numbers. Accordingly, the Fresnel coefficients $\rho_{02}$ in equation (13) should be replaced with $\rho_{0,i+1}$ for multi-layer films.

In principle, if the indices of refraction and the coefficients of absorption are given for all the layers, the resultant Fresnel coefficients can be computed according to the recursion formulae (14) through (17), in which all $\phi_i$ are determined by Snell's Law:

$$\frac{\sin\phi_i}{\sin\phi_0} = \frac{n_0}{n_i} \tag{18}$$

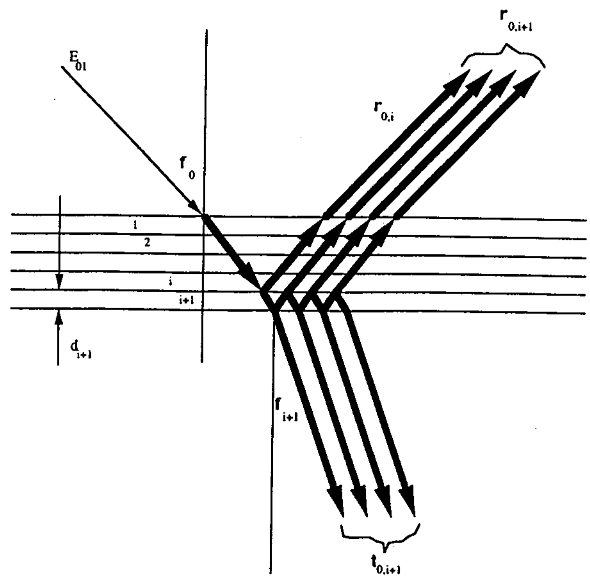


FIG. 3. Multiple reflection, refraction, and transmission of a light beam by a multi-layer thin film. Each of the thick arrow lines represents a resultant beam due to multi-reflection and refraction.

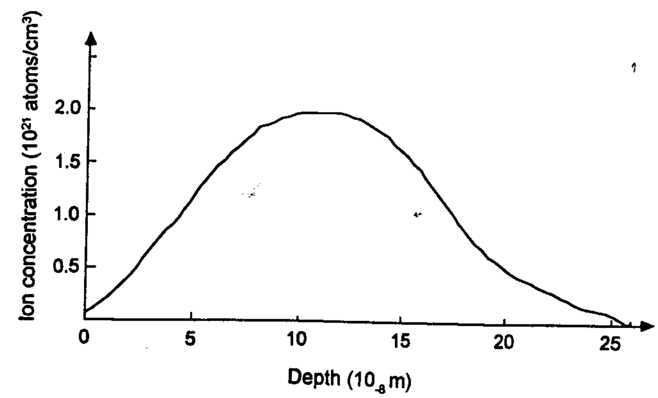


FIG. 4. Typical distribution of ion concentration in the surface region of a silica sample modified by ion implantation (from Whichard et al., 1988).

These resultant Fresnel coefficients are used to determine the values of $\Delta$ and $\Psi$ according to equations (11) through (13). The calculated $\Delta$ and $\Psi$ are then compared to the experimentally measured values. Regression procedures can be devised to determine the indices of refraction of each layer so that the calculated $\Delta$ and $\Psi$ best fit the measured values.

However, there are practical difficulties with this method. The indices (and possibly the coefficients of absorption) of each layer and the thickness of the thin film are not known. There are n + 1 (2n + 1 if the absorption is not negligible) unknowns. It will demand at least $(n + 1)^2$ sets of independent measurements of $\Delta$ and $\Psi$ to determine these unknowns with reasonable confidence. For a four-layer model, there are at least five unknowns, and at least 25 independent sets of $\Delta$ and $\Psi$ are needed. The independent values of $\Delta$ and $\Psi$ can be obtained by varying the incidence angle and the wavelength of the light (and hence changing all $\delta_i$). There is limited freedom for varying the incidence angle as it is usually set near the Brewster's angle for better sensitivity and accuracy. The number of compact He-Ne lasers of different frequencies is also limited. It may be managed to obtain some 20 data sets by making different combinations of the incidence angles and the laser frequencies. Any model involving more than four layers will demand continuously tunable lasers which are much more complex in operation and maintenance, much more expensive and unwieldy. The quadratic model and the Gaussian model presented in this paper enable a yield of continuous distribution of the index of refraction of a thin film, while requiring about 20 data points of $\Delta$ and $\Psi$.

## QUADRATIC DISTRIBUTION MODEL

In many applications, the thin films are usually formed by adding an impurity ingredient into the substrate by ion implantation. The density of the impurity is a function of the depth from the surface and so is the change of index of refraction. Figure 4 shows a typical distribution of concentration of implanted ions (Whichard et al., 1988). The index of refraction in the ion-implanted region increases by <6%. The well-behaved distribution function and the small change in the index of refraction justify a quadratic approximation of the index of refraction:

$$n(x) = n_0 + n_1 - a(x - x_0)^2 \qquad (|x - x_0| < \xi) \qquad (19)$$

$$n(x) = n_0 \qquad \text{otherwise} \qquad (20)$$

$$\xi = \mathrm{sqrt}(n_1/a) \qquad (21)$$

where $n_0$ is the known index of refraction of the substrate and $x_0$ is the average depth of ion implantation. The half-width of the implanted thin film is $\xi$. The constants a, $n_1$, and $x_0$ are to be determined by fitting equation (19) into the experimental data of $\Delta$ and $\Psi$ as outlined in for multi-layer ellipsometry. The three unknowns can be reasonably well determined by about 10 data points. Often times, $\xi$ and $x_0$ can be determined by other independent methods such as ion-backscattering-depth-profile technique. In that case, $n_1$ can be expressed in terms of a:

$$n_1 = a\xi^2. \qquad (22)$$

We then have only one free parameter, a, to be determined.

When the surface is heavily implanted so that the index of refraction changed significantly, equation (19) may be modified to include a fourth-order term:

$$n(x) = n_0 + n_1 - a(x - x_0)^2 + b(x - x_0)^4 \qquad (|x - x_0| < \zeta) \qquad (23)$$

$$n(x) = n_0 \qquad \text{otherwise} \qquad (24)$$

$$\zeta = \mathrm{sqrt}[a/(2b) - \mathrm{sqrt}(a^2 - 4n_1 b)/(2b)]. \qquad (25)$$

The contribution of the fourth-order term is expected to be much smaller than that of the second-order term. Attention has to be paid in numerical calculations so that the argument of the square-root function in equation (25) is always positive.

If the density distribution of implanted ions is not a symmetric function about $x_0$, equation (19) may be modified by including a linear term and a third-power term:

$$n(x) = n_0 + n_1 + b(x - x_0) - a(x - x_0)^2 + c(x - x_0)^3 \qquad (\zeta_1 < |x - x_0| < \zeta_2) \qquad (26)$$

where the two limits $\zeta_1$ and $\zeta_2$ are the positions where the index of refraction n(x) reduces to $n_0$ and can be determined numerically by a computer program.

## GAUSSIAN DISTRIBUTION MODEL

Due to statistical nature of the motion of the ions, the implanted ions usually distribute about the average depth according to Gaussian distribution (Whichard et al., 1988). In that case, the index of refraction can be expressed as a Gaussian function of the depth:

$$n(x) = n_0 + n_1 \exp[-(x - x_0)^2/\sigma^2] \tag{27}$$

where $n_1$ and $\sigma$ are the parameters to be determined by fitting the ellipsometric data. If $\sigma$ is determined independently from ion-backscattering-depth-profile, then only a single parameter, $n_1$, needs to be determined. The modification of index of refraction should be well determined with about 12 data points if the Gaussian model does apply.

In case the absorption coefficient is not negligible, the extinction coefficient, k, needs to be taken consideration. It will involve more free parameters and, therefore, demand more data points. However, the computational procedure should be essentially the same. The modification of the absorption coefficient is usually a minor factor, and a quadratic model should be sufficient to represent modification of the absorption coefficient.

## COMPUTATIONAL PROCEDURE

To determine the unknown parameters involved in the models presented, a regretion procedure is deviced and described. Adoption of either the quadratic or the Gaussian model is assumed. It is assumed that the values of $n_0$ are known. The procedure is: 1) divide the film into a number of layers with equal thickness $\Delta x$; the depth of the ith layer is $x_i$; 2) set the best-guessed initial values for the parameters $n_1$, a, and $x_0$ in the quadratic model, or $n_1$, $\sigma$, and $x_0$ in the Gaussian model; calculate the limit $\xi$ accordingly; 3) calculate the indices of refraction for each layer according to equation (19) or (27); 4) for each set of angle of incidence, $\phi_{j0}$ and $\lambda_j$ at which $\Delta_j$ and $\Psi_j$ are measured, calculate the angles, $\phi_{ji}$, at each layer according to Snell's Law, equation (18); the subscript j denotes the jth measurement of $\Delta_j$ and $\Psi_j$ at the incidence angle $\phi_{j0}$ and the wavelength $\lambda_j$; 5) calculate $\delta_{ji}$ and $\alpha_{ji}$ for each layer according to equations (16) and (17); 6) calculate $\rho_{0,i}$, $\tau_{0,i}$ through $\rho_{0,i+1}$, and $\tau_{0,i+1}$ by making use of the recursion formulae equations (14) and (15) for the jth measurement; 7) calculate $\Delta_j$ and $\Psi_j$ according to equations (11) through (13), replacing $\rho_{02}$ in equation (13) with $\rho_{0,i+1}$; 8) these theoretically calculated $\Delta_j$ and $\Psi_j$ are to be compared to the experimentally measured values $\Delta_{je}$ and $\Psi_{je}$, and an error function $\Gamma$ is constructed as the criterium of data fitting:

$$\Gamma = \frac{1}{m}\sum_{j=1}^{m}\left(\frac{\Delta\Psi_j}{\Psi_{je}}\right)^2 + \frac{1}{m}\sum_{j=1}^{m}\left(\frac{\Delta\Delta_j}{\Delta_{je}}\right)^2 \tag{28}$$

with $\quad \Delta\Psi_j = \Psi_j - \Psi_{je}$ (29)

and $\quad \Delta\Delta_j = \Delta_j - \Delta_{je}$ ; (30)

in equation (28), m is the number of data points; 9) slightly change the parameters of $n_1$, $x_0$, and a (or $\sigma$) in step 2 and repeat steps (3) through (8) in a manner of trial-and-error, to reduce $\Gamma$; and 10) repeat the steps 1 through 9 until $\Gamma$ reaches the minimum.

## CONCLUSION

A multi-layer model is necessary to represent a thin film with finite thickness and varying index of refraction along its depth. An independent multi-layer model, however, requires a large number of independant data points that practically limit the number of layers manageable. This difficulty can be circumvented by adopting continuous distribution models that do not involve as many fitting parameters, and, therefore, do not require as many data points. The quadratic and Gaussian distribution models presented in this paper are adequate for many thin films with continuous distribution of index of refraction.

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