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Determining the refractive index of a $\lambda/4$ thin film on a thick substrate from a transmittance measurement

Robert Chow, Gary E. Loomis and Peter Bilot

Introduction

The refractive index is a key optical constant required by optical thin film designers. The coater-designer team must constantly verify the refractive indices of the non-absorbing optical thin films since the refractive index of a deposited material can vary by switching coating systems or operators as well as expected changes during the course of a coating run.

A transmittance measurement on a spectrophotometer is an easy and accurate (usually to within $\pm 0.5\%$ spread) method of determining the refractive index. In one technique, the refractive index is obtained from visually curve-fitting a calculated (using a thin film design program and selecting the refractive index) transmittance spectrum to the measured transmittance spectrum.

There are two other techniques which will be discussed in this report: A quick, approximate method vs. the exact derivation. What we have not been able to find in the literature is the exact transmittance dependence as a function of the refractive indices (layers) through which the light passes and which accounts for the substrate back reflections. There are undocumented approximation methods as well as one in the literature by Cheremukhin and Rozhnox.¹ Otherwise, most texts either derive the transmittances through optical multilayers² or just the effect of back reflections on the transmittance for thick substrates.^{2,3} Without correcting for substrate back reflections, the derived refractive indices from the measured transmittances are in error by as much as 10%.

One may follow the derivation by the numbered equations and note the physical requirements that underlie them. Equation [5] is the exact expression for calculating the refractive index of the film. The other mathematical expressions are given as guideposts for those needing reference to Maxwell's Equations in optical textbooks.^{2,4} Otherwise, we have tried to verbalize the importance of the non-numbered expressions. The approximate method turns out to be rather accurate and the formula is given in eqn. [8].

Exact Derivation

A typical transmission measurement is sketched in Figure 1. The transmittance, T_m , of a single layer deposited onto a substrate is measured with a spectrophotometer with an ambient air environment. The measured transmittance, expressed as the components of the transmittances through the interfaces 'a' and 'b' are T_a and T_b , respectively, is

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$$T_m = \frac{T_a T_b}{(1 - R_a R_b)}.$$

For non-absorbing media, we use the energy conservation relationships

$$T_p = 1 - R_p, \text{ where } p = \text{either interface a or b,}$$

to simplify the measured transmittance equation to

$$\frac{1}{T_m} = \frac{1}{T_a} + \frac{1}{T_b} - 1 \quad [1]$$

The transmittance at interface 'a' is made to account for the existence of the film by using the characteristic matrix approach. In its full details,

$$T_a = \frac{4 \eta_o \operatorname{Re}(\eta_s)}{(\eta_o B + C)(\eta_o B + C)^*}$$

where η_o is the modified (dependent on the incident angle) optical admittance of the ambient surroundings, η_s is the modified optical admittance of the substrate, Re is taking the real part of the complex modified refractive index, $*$ denotes the complex conjugate of the expression in the parenthesis, and B and C are calculated from the relationship

$$\begin{bmatrix} B \\ C \end{bmatrix} = \prod_{j=1}^q [M_j] \begin{bmatrix} 1 \\ \eta_s \end{bmatrix}.$$

The characteristic matrix $[M_j]$ is associated with the j th layer and has the form of

$$[M_j] = \begin{bmatrix} \cos \delta_j & i \sin \delta_j / \eta_j \\ i \eta_j \sin \delta_j & \cos \delta_j \end{bmatrix},$$

where $\delta_j = 2\pi N_j d_j \cos \theta / \lambda$ is the phase shift experienced by light of wavelength, λ , traveling through a film of geometric thickness d , incident angle θ and complex refractive index N . The i is the complex number $\sqrt{-1}$. At normal incidence (the typical orientation of transmittance measurements), a single layer of a $\lambda/4$ optical thickness and for non-absorbing media, the equation for T_a is greatly simplified to

$$T_a = 1 - \left(\frac{n_o n_s - n_s \bullet n}{n_o n_s + n_s \bullet n} \right)^2, \quad [2]$$

where n_o , n_s and n are the real parts of the refractive indices of air, substrate and film, respectively. The terms within the parenthesis are actually the reflectance of the thin film, R_a . There are a couple points to be made by going quickly through the characteristic matrix derivation. First, the formulation already sums all the waves propagating in the transmitting direction. In other words, there is no need to consider back reflections at the air/film and film/substrate interfaces. Secondly, if one has a multilayer composed of high and low refractive index materials that are $\lambda/4$ optically thick, one may use the quarter-wave rule to determine the refractive index of one of the layers if the refractive index of the 2nd layer is known.

The light reflected at interface 'b' is incoherent due to variations in parallelism and flatness between the front and back surfaces of a thick substrate. So the basic Fresnel transmittance equation is used at interface b,

$$T_b = \frac{4 \operatorname{Re}(\eta_s) \operatorname{Re}(\eta_o)}{(\eta_s - \eta_o)(\eta_s - \eta_o)^*}.$$

Fortunately, at normal incident angle and for non-absorbing media, the Fresnel transmittance equation reduces to a manageable form,

$$T_b = \frac{4 n_s \bullet n_o}{(n_s + n_o)^2}. \quad [3]$$

Inserting eqns [2] to [3] into eqn [1] and rearranging for the film refractive index leads to

$$n = + \sqrt{n_o \bullet n_s \frac{(1-K_2)}{(1+K_2)}}, \quad [4]$$

$$\text{where } K_2 = -\sqrt{1 - \frac{1}{K_1}} \quad \text{and} \quad [5]$$

$$K_1 = \frac{1}{T_m} - \frac{1}{T_b} + 1. \quad [6]$$

The subtlety in utilizing eqn. [1] is to carry through the negative and positive values from the two square root operations. The final values are compared to a real situation and the appropriate signs kept. The sign tracking process has been done and we give the appropriate signs in eqns. [4] and [5].

As an example, the calculation of the refractive index of a physical-vapor-deposited hafnia film follows. A transmittance curve of a $\lambda/4$ (where $\lambda = 532$ nm) hafnia layer on fused silica is given in Fig. 2. The transmittance at the $\lambda/4$ is 72.6%, $n_o = 1.0$, $n_s = 1.46$. Using eqns. [3], [5] and [6] gives $T_b = 0.889$, $K_1 = 1.25$ and $K_2 = -0.449$, respectively. Inserting the appropriate values into eqn. [4] gives the hafnia refractive index as 2.10.

The effect of the T_m measurement error on the film refractive index is determined from the derivative of n with respect to T_m (eqn. [4]).

$$\frac{dn}{dT_m} = \frac{\sqrt{n_o n_s}}{4 T_m^2 K_2 K_1^2} \left[\frac{1}{\sqrt{(1+K_2)(1-K_2)}} + \frac{\sqrt{1-K_2}}{(1+K_2)^{3/2}} \right]. \quad [7]$$

A plot of eqn [7] as a function of transmittance is shown in Fig. 3 for various refractive indices of the substrate (1.5, 1.75 and 2.0). Errors in the T_m measurement have a minimal effect on the calculated film refractive index at transmittance values between 50% and 70%. In order to decrease the effect of a measurement error, one would use a substrate with a lower refractive index. For a given substrate, the dn/n error would be larger for films that have a lower refractive index than those with a higher refractive index.

Continuing with the example above, the accuracy of measuring T_m is $dT_m = 0.001$, where $\pm 0.5\%$ converts into a 0.001 spread. At $T_m = 0.726$ and a substrate refractive index of ≈ 1.5 , Fig. 3 shows that $dn/dT_m \approx -2.2$. Solve for dn by multiplying dn/dT_m ; which is $0.001 \times (-2.2) = 0.0022$, the error spread for n . The refractive index of hafnia is 2.10 ± 0.001 .

At this point we compare eqn. [4] to the approximation method. One measures the transmit. We then subtract out the backside reflectance of the bare substrate/air interface to obtain the approximate film reflectance (obtain T_b from eqn [3])

$$\begin{aligned} R_a &= (1-T_m)-(1-T_b) \\ &= T_b-T_m. \end{aligned}$$

Solve for n_f from the reflectance formula for a single $\lambda/4$ layer at normal incidence.

$$R_a = \left[\frac{n^2 - n_o n_s}{n^2 + n_o n_s} \right]^2.$$

Rearranging gives

$$n = \sqrt{n_o n_s} \sqrt{\frac{1+K_3}{1-K_3}} \quad [8]$$

$$\text{where } K_3 = \sqrt{T_b - T_m}.$$

The effect of the error of the T_m measurement is determined from the derivative of eqn. [8] with respect to T_m :

$$\frac{dn}{dT_m} = \sqrt{n_o n_s} \left[\frac{1-K_3}{2 K_3 (1+K_3)^2} + \frac{1}{K_3 \sqrt{1-K_3^2}} \right] \quad [9]$$

Using the values in the hafnia example above, the refractive index is approximately 2.06. The error associated with this measurement is from Fig. 4, ± 0.0014 . The error is small and the refractive index is only 2% off from the exact derivation of the film refractive index. The refractive index ratio derived from the exact and approximate methods is plotted as a function of transmittance in Figure 5. The curve shows no significantly divergence of the approximate method until the measured transmittance values are below 20%. Other assumptions (i.e., low absorption material) would have failed before reaching this transmittance value. The approximate method is adequate for calculating the refractive index of single layers.

Both the exact and approximate methods of determining the refractive index of thin optical coatings are within the measurement errors of commercially available spectrophotometers.

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¹ G.S. Cheremukhin and V.P. Rozhnox, "Calculation of the thickness and index of refraction of a thin transparent film for the example of zirconium and titanium oxide", *Sov. J. Opt. Technol.* 45(Jan. 1978)1.

² A.H. Macleod, "Thin film optical filters", Macmillan Publ. Comp., N.Y., 2nd edition, 1986.

³ P.W. Baumeister, R. Hahn and D. Harrison, "The radiant transmittance of tandem arrays of filters", *Opt. Acta* 19(1972)853.

⁴ E. Hecht, "Optics", Addison-Wesley Publ. Comp., 2nd edition, 1989.

Figure 1. Sketch of the transmittance measurement.

Figure 2. Transmittance response of a hafnia quarter wave thick coating on fused silica.

Figure 3: The refractive index error associated with different substrates and transmittances

**Figure 4: Error in the refractive index
for different substrates and
transmittance values**

Figure 5: The deviation of the approximate values from the exact refractive index values

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