Refractive index dispersion measurement of absorbing layers from transmittance spectra

Celia Sánchez-Pérez and Augusto García-Valenzuela

Centro de Ciencias Aplicadas y Desarrollo Tecnológico, Universidad Nacional Autónoma de México, Ciudad Universitaria, Apartado Postal 70-186, Distrito Federal 04510, México

Abstract. We describe a fast and accurate method for the measurement of refractive index spectra of absorbing layers from transmittance spectra at two angles of incidence. The method is less sensitive to surface conditions than other photometric techniques. © 2006 Society of Photo-Optical Instrumentation Engineers.

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Photometric techniques¹⁻⁴ used to measure the optical constants of materials are easily adapted to obtain spectra of the optical constants and are suitable to be used with thin films. The accuracy is typically on the second or third significant digit. With adequate instrumentation the measurement of the reflectance R and transmittance T of a layered sample can be measured with relatively high precision (commonly of about 0.1%). Also, robust inversion algorithms have been devised to obtain the optical constants and thickness of thin absorbing films from a few optical transmittance measurements.⁵ However, in all reported photometric techniques, the inversion of the experimental data relies on the use of Fresnel reflection coefficients; and surface roughness, scratches, or small particles on the surface may introduce large errors. In fact, in many cases the surfaces of a sample material do not have optical quality.

In this letter, we describe a simple and accurate method to determine the refractive index spectra of an absorbing layer that is less sensitive to the surface conditions than other techniques.

Consider a slab with a complex refractive index $n + i\kappa$ and an optical beam incident at an angle θ_i . If the slab absorbs most of the incident light in a single pass, we can ignore multiple reflections within the slab. It is not difficult to show that in this case the intensity of the transmitted beam is given by

$$I_t = I_0 T_1 T_2 \exp(-2k_0 \tau d), \quad (1)$$

where I_0 is the intensity of the incident beam, d is the thickness of the slab, k_0 is the wave number in a vacuum, and T_1 and T_2 are the transmittance coefficients of the interfaces of the slab,

$$\tau = 2^{-1/2}[-a + (a^2 + b^2)^{1/2}]^{1/2}, \quad (2)$$

where $a = n^2 - \kappa^2 - \sin^2 \theta_i$, $b = 2n\kappa$, θ_i is the angle of incidence, and we have assumed that the refractive index of the incidence medium is one. If we suppose $\kappa \ll n$, and expand Eq. (2) in a power series of κ and neglect terms of order κ^2 we have $\tau \approx \kappa / \cos \theta_m$ where $\cos \theta_m = (1 - \sin^2 \theta_i / n^2)^{1/2}$. Thus, θ_m is given by the usual Snell's law with the real part of the refractive index of the slab.

The method proposed in this letter consists of measuring the exponential decay factor of the transmittance, $\exp(-2k_0 \kappa d / \cos \theta_m)$, at two angles of incidence, θ_1 and θ_2 , and then solving for the real part of the refractive index of the slab as

$$n = \left(\frac{\sin^2 \theta_1 \ln^2 e_1 - \sin^2 \theta_2 \ln^2 e_2}{\ln^2 e_1 - \ln^2 e_2} \right)^{1/2}, \quad (3)$$

where $e_j = \exp[2k_0 \kappa d / (1 - \sin^2 \theta_j / n^2)^{1/2}]$. Note that the determination of n from Eq. (3) does not require knowledge of the film thickness, nor does it depend on the validity of Fresnel relations at the interfaces of the layer system. Now, to measure only the exponential factor we must normalize the transmittance measurements given by Eq. (1) by the factor $I_0 T_1 T_2$. This factor must be estimated by additional measurements at angles of incidence, θ_1 and θ_2 .

The error in determining n will come from the errors in estimating the factors $I_0 T_1 T_2$. A thorough analysis of the errors will be presented elsewhere. Nevertheless, to give an idea of the attainable accuracy of the method, we plot in Fig. 1 the relative error in obtaining n as a function of the absorbance $A = 2k_0 \kappa d$ of the layer for a few values of the relative error on measuring the exponential decay factor e .

We can see that if the relative errors $\Delta e_j / e_j$ for both $j = 1$ and 2 are similar and less than 2%, and if the absorbance is larger than 0.6, that is, a transmittance on normal incidence of less than 54%, then the error $\Delta n / n$ is less than 1%.

In order to demonstrate the viability of the method in practice, we measured the refractive index of water in the wavelength range of 500 to 900 nm. We used a thin rectangular cell made of glass slabs filled with distilled water and fixed on top of a goniometer. The thickness of the cell was about 1 mm. However, we did not measure it precisely since the method does not require knowing the layer thick-

Fig. 1 Relative error on the refractive index, $\Delta n / n$, versus the absorbance of the sample assuming $n = 1.33$ and for a relative error in the values of e_1 and e_2 of: $\Delta e_1 / e_1 = \Delta e_2 / e_2 = -2\%$ (solid line), $\Delta e_1 / e_1 = 0$ and $\Delta e_2 / e_2 = -2\%$ (dotted line), $\Delta e_1 / e_1 = -2\%$ and $\Delta e_2 / e_2 = 0$ (dashed line).

ness. We added a small amount of black ink to the water to reduce the transmittance at normal incidence to less than 50% for all wavelengths in the specified range.

To measure the transmittance we used a white light beam incident at a fixed angle with respect to the normal to the entrance facet of the cell. The light beam was generated with a white light source (BPS100 BWTEK Inc.) coupled to a pigtailed optical fiber. The beam was transmitted through the cell and collected by a 600- μm -diameter optical fiber coupled to a miniature spectrophotometer (HR2000 Ocean Optics). Transmitted spectra were measured at two angles of incidence for the absorbing and pure water. These latter spectra are equal to the factors $I_0T_1T_2$ and were used to normalize the transmittance spectra of the absorbing water solution at the corresponding angle of incidence. It is worth mentioning that the glass cell was not of optical quality nor was it cleaned by any special procedure. As a matter of fact we measured the factor T_1T_2 for the empty cell used in our experiment and found that it deviated appreciably (around 5% at normal incidence) from the predictions of the usual formulas based on the Fresnel reflection and transmission coefficients. The reason is that the interfaces of the cell were not of optical quality.

In Fig. 2 we plot the refractive index calculated using Eq. (3) for wavelengths within the range $\lambda = 500$ to 900 nm. In the inset of this figure we show the normalized transmittance spectra at $\theta_1 = 0^\circ$ and $\theta_2 = 52^\circ$. Finally, in Fig. 3 we plot the errors of the calculated refractive indices. These were obtained by comparing our results with reference data for the refractive index of water at 25°C . We can see that the relative error using an adjacent averaging smoothing over 25 nm in wavelength oscillates around 0.3% from 550 to 900 nm and only reaches 1.4% at the lowest wavelength. Since the present

Fig. 2 Refractive index versus wavelength from measurements (solid line) and reference data at 25°C (symbols). The normalized transmittance spectra of the sample are shown in the inset.

Fig. 3 Relative error on the refractive index for raw data (dotted line) and using an adjacent averaging smoothing over 23 nm in wavelength (solid line).

method is a photometric technique, this error is actually very low and is already useful to characterize absorbing films used in optical devices.

The application of this method to solid films will require a different technique to estimate accurately the normalization factors $I_0T_1T_2$ at each angle of incidence. Using TM polarized light and angles of incidence near the angle of minimum reflectance (due to the Brewster effects) will probably be convenient.

The accuracy obtained by our method in the absence of high-quality surfaces is comparable to that of other photometric techniques when they are applied to samples with optical quality surfaces. However, our method is applicable to only highly absorbing layers.

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