Optical interference method to obtain thickness and refractive indices of a uniaxial medium

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Optical interference fringe measurements of the thickness of weakly absorbing media can be rapid, accurate, and nondestructive. When the refractive index n of the sample is known, it will give us the layer thickness d. If, however, n is unknown, at least two independent spectrophotometric measurements are needed to obtain both n and d. A statistically based scheme is proposed to analyze the interference pattern in order to determine the refractive index and the thickness of the sample. The absolute interference order is also determined with the proposed technique. The major approximation inherent in the method is that the layer must be weakly absorbing and nondispersive over the wavelength region of interest. The method is applied to determine the optical constants of a uniaxial medium with the optical axis parallel to the faces.

I. INTRODUCTION

Optical interference fringe measurements of the thickness of transparent and weakly absorbing media can be obtained in a fast and accurate way.[1–8] The experimentally measured quantity is the optical reflectance or transmittance of the sample as a function of the wavelength at a fixed angle of incidence (constant angle reflectance/transmittance interference spectroscopy, CARIS and CATIS, respectively) or at a fixed wavelength and a variable angle of incidence (variable angle reflectance/transmittance interference spectroscopy, VARIS and VATIS, respectively).

In the most common mode of usage, the interference fringe pattern obtained from these methods is combined with a previous knowledge or measurement of the refractive index of the sample, in order to determine its thickness.[9–15] When the refractive index of the sample material is unknown, at least two measurements at different incidence angles, using CARIS and CATIS methods or two measurements at different wavelengths using VARIS and VATIS methods, are needed.

In the present study, the object was to develop a technique for determining the refractive indices and the thickness of a weakly absorbing and nondispersive uniaxial sample from two or more CATIS measurements. This method is based on the prediction of the *optimum* value of the absolute interference order (N_{opt}) of the wavelength of the maxima experimentally obtained, defined as the integer value that minimizes a quadratic error function (see Sec. II A). The value of N_{opt} is not taken into account in other studies[2,3] that implicitly assume no errors in the measurement of the wavelengths at which the maxima are obtained. Another problem arises from the fact that the real part of the refraction index is considered to be independent of the wavelength: this is a good approximation when the spectral range used is narrow, but fails in the case of wider spectral ranges (see, for example, Refs. 3 and 13). This effect was observed and measured by Čelustka *et al.*[9] The method can be applied, without loss of generality, to the minima of the interference pattern. It could also be applied, with appropriate modifications, to CARIS, VARIS, or VATIS measurements and for isotropic media. The sample we are interested in is a plastic film commonly used as a physical substrate for other composite films, such as the thermoplastic photoconductor commonly used in holography.[16]

The importance of optical characterization of these films is an increasing trend in modern industry: the optical constants of plastic materials are of very great importance from an industrial viewpoint and have been correlated with manufacturing parameters such as dry pressure and final thickness obtained.[17] In Sec. II the theory of the technique of analysis is briefly presented. The experimental results are presented and discussed in Sec. III.

II. THEORY

Consider the experimental arrangement shown in Fig. 1. The sample to be characterized is flat and of thickness d and refractive index $\tilde{n}$. The incoming beam is assumed to be monochromatic and linearly polarized, with the electric vec-

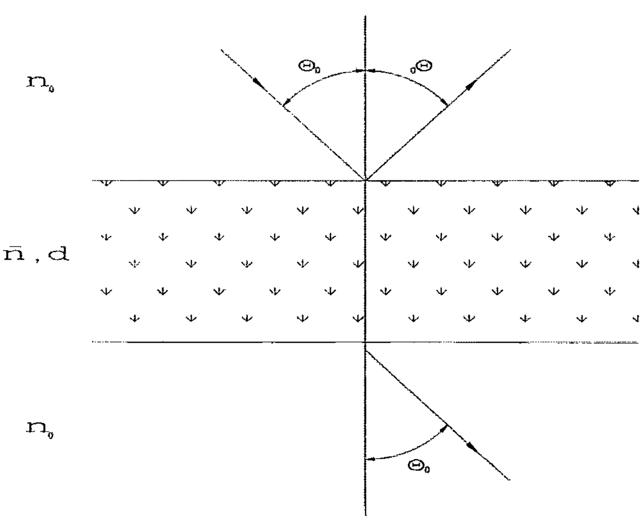

FIG. 1. Reflection and transmission of a plane wave in the sample of plastic considered as a Fabry–Perot interferometer. $n_0=1$ (air).

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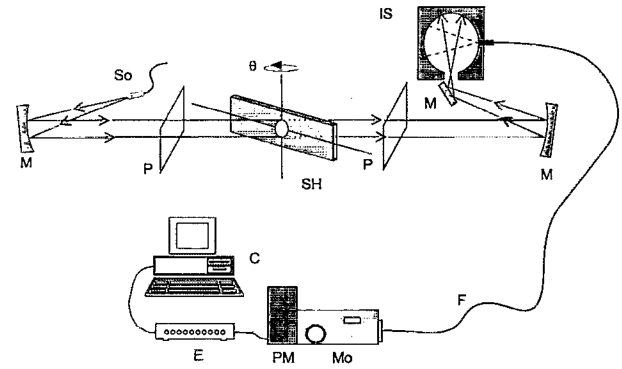

FIG. 2. Spectrogoniometer scheme (for a description, see the text).

tor either parallel (p) or perpendicular (s) to the plane of incidence. The sample may be considered weakly absorbing and nondispersive over the wavelength range of interest. It may be considered a Fabry–Perot interferometer. It is well known that in the multiple-beam fringes obtained, each surface is described by transmission and reflection coefficients, which are given in terms of the refractive indices and the angles of incidence and refraction by the Fresnel formulas.[18]

Let us consider in Fig. 1 an incident monochromatic plane wave which forms an angle θ_0 with the first interface, and let us consider the refraction index of the sample as a complex number given by $\bar{n}=n-i\kappa$. The amplitude transmittance may be written as

$$t=\frac{(1-r^2)\exp(-i\beta)}{1-r^2\exp(-i2\beta)}, \tag{2.1}$$

where β is given by

$$\beta=\frac{2\pi}{\lambda}d\bar{n}\cos\theta_1, \tag{2.2}$$

and r stands for the Fresnel reflection coefficient in the first interface. It should be pointed out that θ_1 is a complex number as derived from the Snell law of refraction when considering an absorbing medium. In order to obtain the overall transmission irradiance, $T(\lambda)$, we shall define the following auxiliary variables u and v as

$$u=\frac{2\pi}{\lambda}d\,\mathrm{Re}(\bar{n}\cos\theta_1),\qquad v=\frac{2\pi}{\lambda}d\,\mathrm{Im}(\bar{n}\cos\theta_1), \tag{2.3}$$

where Re and Im stand for the real and imaginary parts, respectively. Explicit expressions for both u and v are obtained after a little algebra

$$u=\frac{2\pi}{\lambda}d\rho_1\cos(\phi_1/2),\qquad v=\frac{2\pi}{\lambda}d\rho_1\sin(\phi_1/2), \tag{2.4}$$

where ρ_1 and ϕ_1 are the modulus and argument of the complex number $\bar{n}\cos\theta_1$ and are given in terms of the incidence angle and the refraction index of the sample as

$$\rho_1=\sqrt{(n^2-\kappa^2-\sin^2\theta_0)^2+(2n\kappa)^2},\qquad \phi_1=\arctan\left(\frac{-2n\kappa}{n^2-\kappa^2-\sin^2\theta_0}\right). \tag{2.5}$$

Then $T(\lambda)$ may be written as

$$T(\lambda)=\frac{|1-r^2|^2\exp(2v)}{[1-|r|^2\exp(2v)]^2+4|r|^2\exp(2v)\sin^2[2(\delta-u)]}, \tag{2.6}$$

where δ is the phase of the reflection coefficient.

Equation (2.6) may be recast as

$$T(\lambda)=T_0(\lambda)\,\frac{1}{1+m(\lambda)\sin^2[2(\delta-u)]}, \tag{2.7}$$

where $T_0(\lambda)$ is given by

$$T_0(\lambda)=\frac{|1-r^2|^2\exp(2v)}{[1-|r|^2\exp(2v)]^2} \tag{2.8}$$

and $m(\lambda)$ stands for

$$m(\lambda)=\frac{4|r|^2\exp(2v)}{[1-|r|^2\exp(2v)]^2}. \tag{2.9}$$

When the imaginary part of the refraction index is high, the value of $m(\lambda)$ tends to zero and thus there is no observable modulation of the transmitted irradiance. In our case we have considered low values for κ; thus as a first approximation, the phase shift for two subsequent waves may be obtained neglecting the effects of κ in Eq. (2.4) and producing

$$u\approx\frac{2\pi}{\lambda}d\sqrt{n^2-\sin^2\theta_0}. \tag{2.10}$$

Equation (2.10) expresses the fact that for weakly absorbing media, the phase shift is quasi-independent of the imaginary part of the refraction index. This will be tested for the measurements presented in Sec. III. In this case Eq. (2.7) may be expanded in powers of $m(\lambda)$, and retaining the first terms, we obtain

$$T(\lambda)\cong T_0(\lambda)\{1-m(\lambda)\sin^2[2(\delta-u)]\}, \tag{2.11}$$

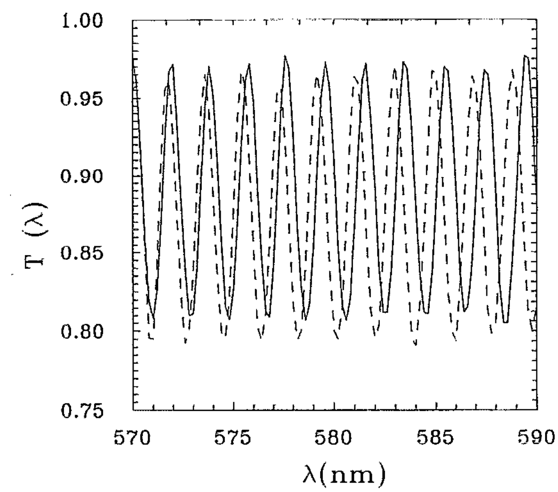

FIG. 3. Interference pattern for p (continuous line) and s (dashed line) polarization obtained for the sample of polystyrene considered.

TABLE I. Consistency of the values of the indices and thickness predicted for the polystyrene sample considered for each pair of angles of incidence (θ_{01} and θ_{02}) (s polarization). stde stands for the standard error of the parameters inside the brackets.

θ_{01} and θ_{02}	n_0	stde(n_0)	d (μm)	stde(d) (μm)
$\theta_{01}=0°$, $\theta_{02}=30°$	1.712	0.002	51.58	0.06
$\theta_{01}=0°$, $\theta_{02}=40°$	1.7127	0.0009	51.56	0.03
$\theta_{01}=0°$, $\theta_{02}=50°$	1.7144	0.0004	51.52	0.01

which is the common type of equation for an interference pattern in two-beam interferometers.

Since $T_0(\lambda)$ is a slowly varying function of wavelength, this function will not modify the position of the maxima of Eq. (2.11), which are given by the conditions

$$\begin{aligned} &2(\delta-u)=2N\pi, \quad \text{maxima},\\ &2(\delta-u)=(2N+1)\pi, \quad \text{minima}, \end{aligned} \tag{2.12}$$

N being an integer number, known as the interference order.

A. Analysis of the spectrophotometric data

The spectrophotometric data are obtained with a spectrogoniometer designed by us and manufactured by SOPRA[19] working in transmittance mode. The characteristics and performance of this system are extensively described in Ref. 20. The light source (**So**) is a stabilized halogen lamp and after transmission at the sample (situated in **SH**), the light is collected by an integrating sphere (**IS**). The spatial uniform signal coming from **IS** is introduced into a monochromator (**Mo**) by means of an optical fiber (**F**). The output from **Mo** is detected by a photomultiplier and photocounting system (**PM**). An electronic rack (**E**) drives the detection system. The whole system is commanded by a host computer (**C**). Polarizers (**P**) are included in the system to select the appropriate orientation of the electromagnetic field. An angular resolution of 0.02° in the determination of the angle and a spectral resolution of 0.7 nm are achieved with this system. The mirrors (**M**) both have a numerical aperture of 0.0834 and the dynamic range of the detection system in linear response is 10^4 (see Fig. 2).

An example of the interference pattern obtained is shown in Fig. 3 for an incidence angle of 0° for both p and s polarizations. This figure shows a shift of the wavelength at which the maxima are obtained between both polarizations. In order to obtain the optical thickness, and thus the refraction index (n) and the thickness (d) of the sample considered, a statistically based algorithm was developed. The steps of the algorithm are:

(1) The wavelength of the maxima is detected and sorted in descending order. Let k be the number of maxima obtained in the spectral range considered, N_1 the first interference order, and θ_0 the angle of incidence; thus, taking into account Eqs. (2.10) and (2.12), the following equation may be written:

$$\begin{aligned} &2d\sqrt{n^2-\sin^2\theta_0}=N_1\lambda_1,\\ &2d\sqrt{n^2-\sin^2\theta_0}=(N_1+1)\lambda_2,\\ &\quad\vdots\\ &2d\sqrt{n^2-\sin^2\theta_0}=(N_1+k)\lambda_k. \end{aligned} \tag{2.13}$$

It was assumed in Eq. (2.13) that the refraction index is independent of the wavelength in the spectral range considered (from λ_1 to λ_k). Since the material considered is weakly absorbing, the value of the phase of the Fresnel reflection coefficient δ takes a near zero value and is ignored in Eq. (2.13). This hypothesis will be tested later with the experimental results obtained.

(2) For each pair of consecutive maxima, λ_j and λ_{j+1}, the interference order $N_1(j)$ is estimated as follows:

$$N_1(j)=\frac{\lambda_{j+1}}{\lambda_j-\lambda_{j+1}}-j, \tag{2.14}$$

where j runs from 1 to $k-1$. The values of $N_1(j)$ must be integer numbers as is imposed by Eq. (2.12). Nevertheless, the values of $N_1(j)$ predicted by Eq. (2.14) by the $k-1$ pairs of consecutive maxima will be different among them and, generally, noninteger values. The problem is to determine which is the *optimum* value of the interference order. This optimum value is determined from a statistical basis.

(3) The average interference order, defined as

$$\bar{N}_1=\frac{\sum_{j=1}^{k-1}N_1(j)}{k-1}, \tag{2.15}$$

and its variance σ_{N_1}, are calculated. The sources of errors are assumed to be distributed according to a Gaussian distribution function, thus the optimum value of N_1 should be in the interval $I=(\bar{N}_1-3\sigma_{N_1}, \bar{N}_1+3\sigma_{N_1})$ with a 99% confidence.

(4) Let i be an integer value in the interval I; thus the average optical thickness for this integer value is calculated as $\langle\Delta\rangle_i=\langle d\sqrt{n^2-\sin^2\theta_0}\rangle_i$.

(5) According to the value of $\langle\Delta\rangle_i$, the wavelengths of the maxima $[\lambda_j^p(i)]$ are recalculated, taking into account that j runs from 1 to k since there are j maxima.

(6) The residuals for each value of i are calculated as follows:

$$R(i)=\sum_{j=1}^{k}[\lambda_j^m-\lambda_j^p(i)]^2; \tag{2.16}$$

λ_j^m being the wavelength of the jth maximum measured and $\lambda_j^p(i)$ the wavelength of the jth maximum predicted by each value of the integer $i\in I$.

TABLE II. Values of the indices and thickness predicted for the polystyrene sample considered. stde stands for the standard error of the parameters inside the brackets.

n_0	stde(n_0)	n_e	stde(n_e)	d (μm)	stde(d) (μm)
1.7130	0.0012	1.6602	0.0013	51.55	0.03

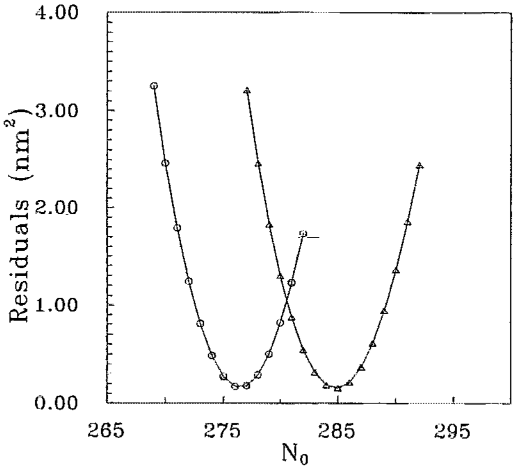

FIG. 4. Residuals for the determination of the optimum interference order for p(O) and s(Δ) polarization obtained for the sample of polystyrene considered in Fig. 2. The incidence angle considered is $\theta_0=0°$.

(7) The optimum interference order is selected to be the integer $i \in I$ with a minimum value of $R(i)$.
(8) Let N_{opt} be the optimum value of the interference order; then the optical thickness is calculated as the average of the optical thickness for the $k-1$ pairs of maxima, and its error as the standard error.

In order to check the validity of the aforementioned procedure, the values of the predicted wavelengths should be plotted versus the measured values, and a linear regression analysis between the measured and predicted wavelengths based on the maximum-likelihood criterion should be considered to determine its applicability. The residuals of the wavelengths, defined as

$$R_\lambda = \lambda_j^m - \lambda_j^p(N_{opt}), \tag{2.17}$$

should be distributed as a Gaussian distribution function (white noise) with zero mean and a certain variance σ_r. In

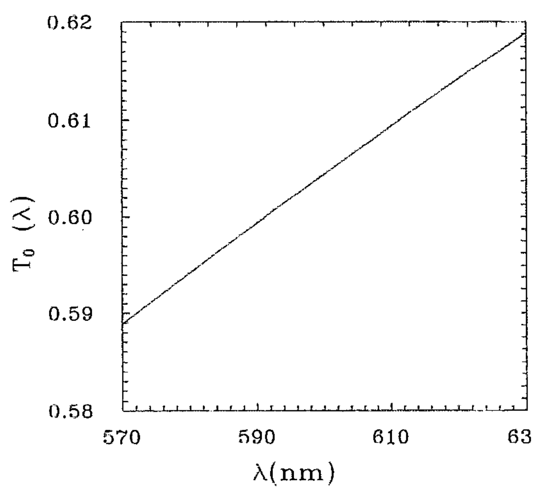

FIG. 5. Experimental shape of the term $T_0(\lambda)$ of Eq. (2.8) as a function of the wavelength in the spectral range employed. The incidence angle considered is $\theta_0=0°$.

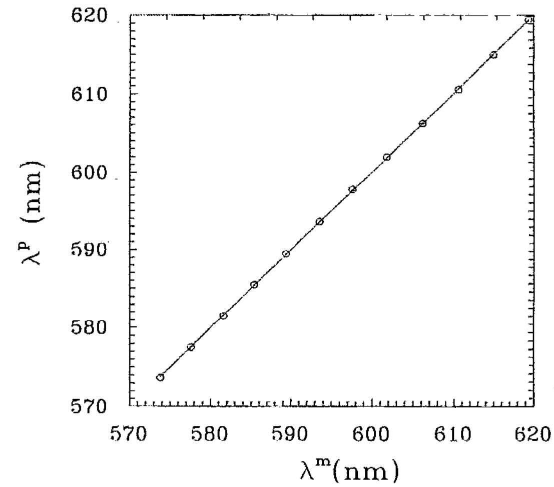

FIG. 6. Predicted wavelengths (λ_j^p) vs measured wavelengths (λ_j^m) of the maxima for p polarization.

Eq. (2.17), $\lambda_j^p(N_{opt})$ is the wavelength of the jth maximum predicted by the optimum interference order (N_{opt}).

III. EXPERIMENTAL RESULTS

The samples considered in this study were plastic uniaxial media manufactured by extrusion procedures: these procedures are responsible for the anisotropy of the material. The molecular chains are aligned, during the manufacturing, in the extrusion direction and nonuniformities of the refraction index should be expected, whereas in the perpendicular direction, the refraction index should be independent of the portion of the sample considered. The optical axis of the sample is considered to be parallel to the faces of the samples. Due to the anisotropy of the sample, the neutral axes of the sample were first determined. Measurements at incidence angles of $\theta_0=0°$, 40°, and 55° were conducted for s polarization and at an incidence angle of $\theta_0=0°$ for p polarization. The values of the refraction index and thickness predicted for s polarization at normal incidence and the other angles were all consistent among themselves and are listed in Table I.

The algorithm described in Sec. II A was applied to the the spectrophotometric data, obtaining the values of the thickness d and the refractive index in the ordinary axis n_0. With the data for p polarization, the value of d was used to predict the refractive index in the extraordinary axis n_e. The results are listed in Table II with the errors predicted by the statistical method. The residuals as a function of the interference order for both polarizations are shown in Fig. 4: there is a minimum value, for each polarization considered, at a cer-

TABLE III. Fitting of the predicted wavelengths (λ_j^p) vs measured wavelengths (λ_j^m) of the maxima for p polarization in Fig. 5. 21 degrees of freedom. Model proposed $\lambda_j^p = m\lambda_j^m$.

Polarization	m	stde(m)
s	1.000014	0.000038
p	1.000015	0.000035

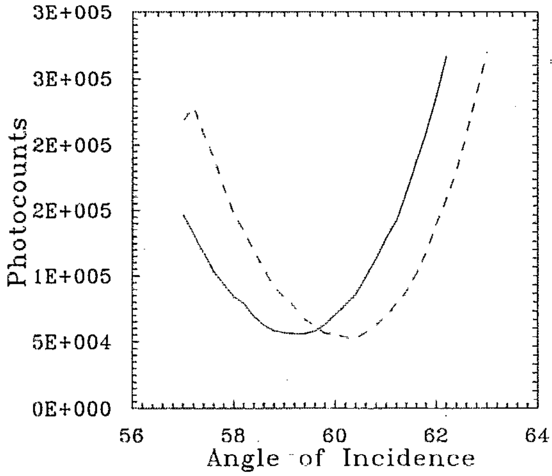

FIG. 7. Determination of the Brewster angle for both axes of the sample: ordinary axis (dashed line) and extraordinary axis (continuous line).

tain integer value that is taken as the N_{opt}. The value of N_{opt} is different for p and s polarization, thus indicating that the index of refraction for both axes is different, as was expected. It should be pointed out that the measurements were carried out in the same area of the sample using a mask to avoid the influence of local nonuniformities of the sample.

In Sec. II, it was pointed out that the term $T_0(\lambda)$ in Eq. (2.7) was a slowly varying function of the wavelength. Figure 5 shows the shape of this term, confirming the aforementioned hypothesis that the position of the maxima are unaltered by this term. The data listed in Table II were used for this calculation. The imaginary part of $\bar{n}$ was neglected when computing the variable u. We have computed $T(\lambda)$ with the data listed in Table II with the above-mentioned approximation and with the exact formula [Eq. (2.7)], showing that there is no appreciable difference between them. It is also clear from Table II that the sample considered shows a high degree of anisotropy (up to 10^{-2}).

With the values of the optimum interference order N_{opt} obtained for both polarizations, we are able to predict the values of the wavelengths of the maxima. A direct way to test the validity of the hypothesis of independence of the refraction index on the wavelength is to compute a linear regression analysis between the measured and the predicted wavelengths at which the maxima occur (λ_j^p vs λ_j^m, respectively). The value of 1 for the slope of this curve will confirm our hypothesis and the deviations from this value will give us information about the effect of spectral dispersion in the material considered. A plot for p polarization is shown in Fig. 6. The results of the fitting are listed in Table III. We conclude that our hypothesis may be considered significative from a statistical viewpoint, as the slope of the curve has a value close to unity. The residuals of the wavelengths (R_λ) [Eq. (2.17)] were showed to be distributed as white noise and the Kolmogorov–Smirnov criterion was applied, giving a significance level of 0.936 and 0.651 for s and p polarizations, respectively. An experimental measurement of the Brewster angle at $\lambda=632.8$ nm for both axes of the sample was carried out in order to check its anisotropy in an independent way. Figure 7 shows that the Brewster angle is slightly different for each orientation, thus confirming the anisotropy.

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