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Determination of thin film optical properties by the photoacoustic OPC technique

C García-Segundo†, M Villagrán-Muniz† and S Muhl‡

† Centro de Instrumentos, UNAM, Circuito Exterior, Ciudad Universitaria, Apdo Postal 70-186, CP 04510, México, DF
‡ Intituto de Investigación en Materiales, UNAM, Circuito Exterio, Ciudad Universitaria, Apdo Postal 70-360 y 70-284, México, DF

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Abstract. Through the use of maxima and minima of interference fringes in transmittance spectra registered by the open photoacoustic cell (OPC) detection technique, we have determined the refraction index n and thickness d of aluminium nitride thin films which had previously been deposited on opaque and transparent substrates. The results for the transparent substrate are in agreement with those obtained from the transmission spectra. To determine n as a function of λ we developed a novel procedure for the analysis of the OPC spectra. The n values so obtained were introduced in an appropriate Airy transmittance function, and their numerical evaluation permits the reproduction of the experimental transmittance OPC data and optical curves. This scheme was used to establish the uniqueness of the calculated data.

1. Introduction

Photoacoustic (PA) detection techniques applied to the study of dielectric, semiconductor or metallic thin films are usually focused to investigate their thermoelastic properties or to determine their optical absorption spectra [1–3]. The determination of the optical parameters is normally carried out by other optical methods, such as elipsometry, transmission and interferometry [4, 5]. However, many of these methods are specific with respect to the type of film that can be treated. Various related reviews are available, such as those by Heavens, Macleod, Palik, and Abelès [6–9], where the more popular techniques are described together with the associated problems. Specifically, the determination of the optical constants by numerical or graphical means can be ambiguous because of the existence of multiple nonunique solutions. Attempts have been made by Denton *et al* and Bennett and Booty [10, 11] to obtain unique and correct solutions, but this is difficult particularly for thick films.

The idea to obtain the optical constants (thickness and refractive index) from thin film interference data has been addressed by many authors [4, 12–14]. Some of these suggest that the information can be determined from the spectral positions of the maxima and/or minima of interference fringes when the data are taken at two or more angles of incidence of the probe beam, assuming that any change that this causes in the refractive index is negligible. This method is applicable to transmittance or reflectivity spectra but if the substrate is opaque, or the film surface and/or film substrate interface is not optically flat, serious problems can arise.

By substituting the optical detector by an OPC microphone in the transmittance detection configuration (figure 1(a)), we have been able to obtain pseudo transmittance spectra for films on both transparent and opaque substrates (figures 1(b) and 1(c)). From this we have named the obtained transmittance OPC spectra as photoacoustic transmission spectra (PATS). From the use of this technique, we have developed a novel procedure to determinate n and d, for low absorption films. An additional advantage of the technique is that it provides a simple method to find the variation of the refractive index as a function of the wavelength within the experimental spectral range.

2. Experiment

The optical analysis of thin films deposited on opaque substrates is difficult. As shown in figure 1(c) the light passing through the film is immediately absorbed in the substrate. This limits the optical detection techniques but favours the application of the photoacoustic method since the absorbed light is converted into heat, and under appropriate conditions this heat energy can be detected as an acoustic signal.

If the incident illumination is modulated at a suitably low frequency, such that the acoustic signal is large, then the local temperature changes and the generated thermal

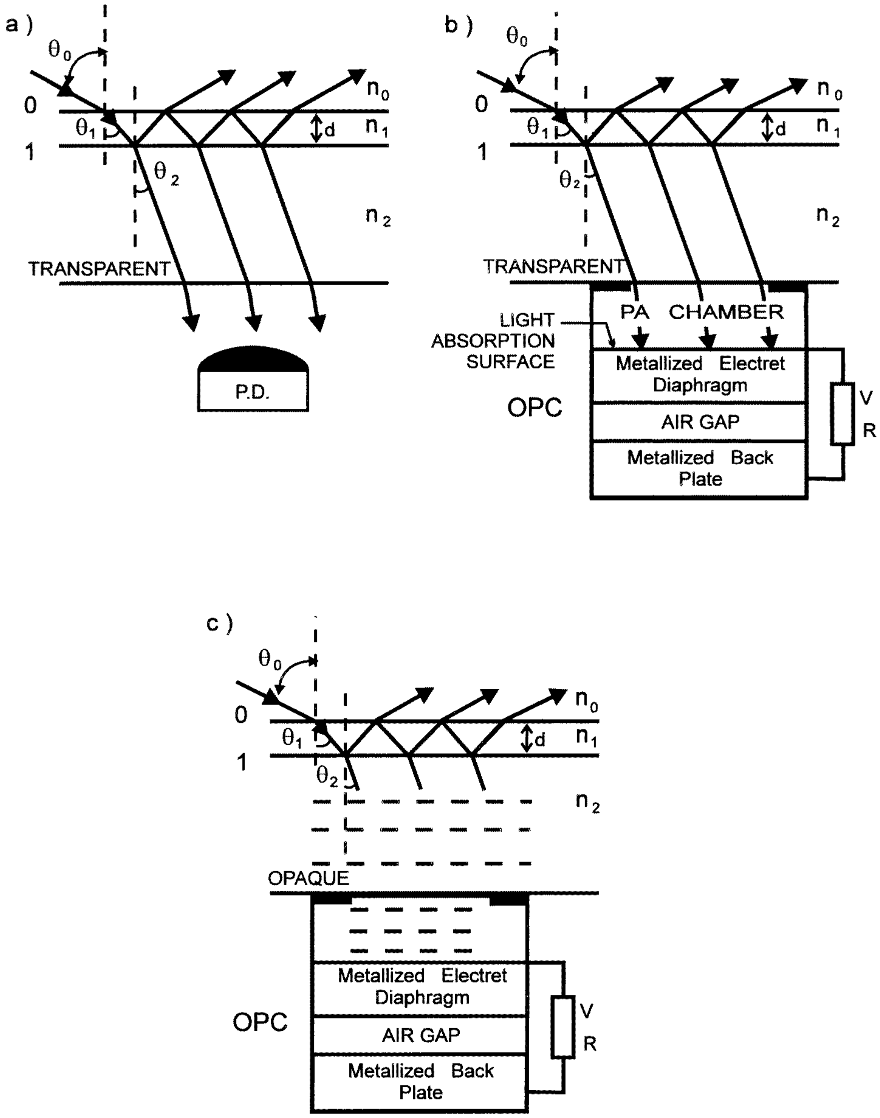

Figure 1. Schematic drawings of the detection schemes for thin films, of thickness *d*, on substrates: (a) conventional transmission spectroscopy of a transparent substrate; P.D. is a photodetector; (b) photoacoustic scheme to obtain thin film PATS on transparent substrates, and (c) same as (b) by opaque substrate; the horizontal dashed lines indicate thermal waves registered by the OPC microphone.

expansion of the substrate vary with the same modulation frequency. This thermal expansion is transmitted to the gas in contact with the substrate and detected by the microphone in the OPC cell. A complete description of the theory that outlines the generation and detection of this type of signals, called photoacoustic (PA) signals, has been developed by various authors [15–17]. Additional details are reported in [18, 19].

The theory establishes that the dependence of the PA signal amplitude, P_{TH}, on the incident radiation, is linear, $P_{TH} \propto I_T$. This means that the PA amplitude depends directly on the transmission of the film because this determines the fraction of the incident energy which reaches the substrate, $I_T = T I_I$, where I_I is the intensity of the incident light and T is the transmission of the film and includes the effect of the air–film and film–substrate interfaces. This is given by the Airy function [20],

$$T = \frac{(t_0 t_1)^2}{(r_0 r_1 - 1)^2 + 4 r_0 r_1 \sin^2(\delta/2)}. \tag{1}$$

This transmission equation is a direct function of the corresponding transmission and reflection Fresnel coefficients t_j and r_j, where the subindex $j = 0$ corresponds to the air–film interface and $j = 1$ to the film–substrate interface.

Thus the angles of the light path relative to the normal to the surface at incidence, within the film and within the substrate are θ_{0i}, θ_{1i} and θ_{2i} (=θ_{li}, $l = 0, 1, 2$), respectively, where the subindex i is used to indicate the different incident angles used. Therefore, the oscillating term is given by

$$\delta = \frac{4\pi n d}{\lambda} \cos\theta_{1i} = 4\pi \nu n d \cos\theta_{1i} \tag{2}$$

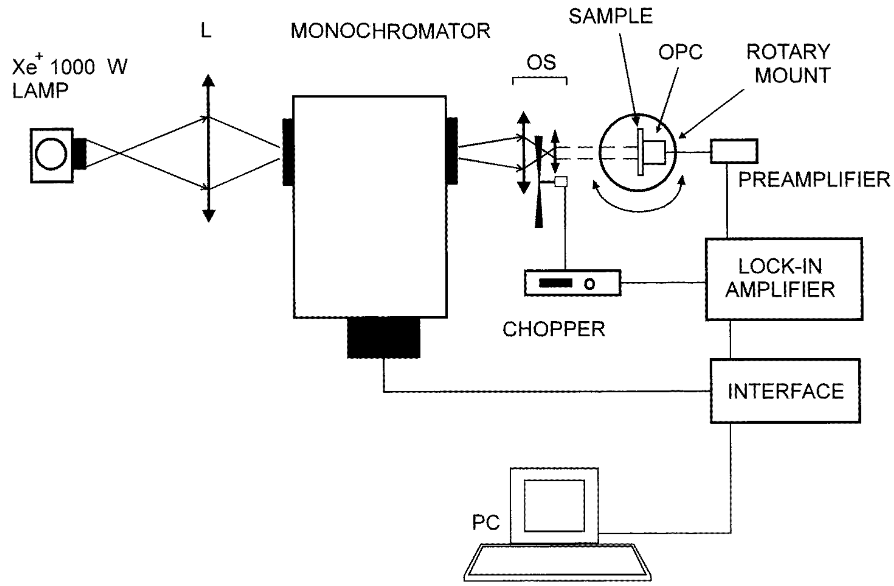

Figure 2. Experimental arrangement.

where d is the film thickness, λ is the wavelength of the illumination, and $v = \omega/2\pi$ is the optical frequency. In this way the maxima in the transmission spectra occur when $\delta = m\pi$, with $m = 0, \pm 1, \pm 2,$ or as normally expressed,

$$2nd\cos\theta_{1i} = m\lambda. \tag{3}$$

At each interface we have from Snell's law

$$n_{inc}\sin\theta_{inc} = n_{tra}\sin\theta_{tra}. \tag{4}$$

The subindices inc and tra refer to the incident and transmission medium respectively. As will be described in more detail in later sections, the wavelength values of the maxima of the experimental photoacoustic interference spectra are the same as for the transmission interference spectra, indicating that the photoacoustic effect is related to the transmission of the light through the film. This can easily be understood for the opaque substrate but is not so obvious for the transparent case. In this latter case the light traverses the sample and is absorbed by the matt surface of the microphone (figure 1(b)) placed directly below the sample. The veracity of this idea was demonstrated by applying carbon black to the back surface of the transparent substrate with a resulting increase in the intensity of the photoacoustic signal.

The acquisition of the PATS data was carried out using the experimental arrangement shown in figure 2 for thin films of AlN deposited on fused quartz and single crystal silicon substrates. In this set-up the light is supplied from a 1 kW Xe^+ discharge lamp (Oriel Mod. 66023) which was focused on the entrance slit of a 0.27 m Acton Research Corporation (ARC) SpectraPro 275 monochromator with 1200 lines/mm grating. The light was then focused, using quartz lenses, onto the sample using the optic system OS. The beam was modulated at a frequency of 15 Hz using a Stanford Research System (SRS) chopper (model SR-540). The samples were mounted on the OPC microphone (Radio Shack, Cat. No. 270-092B) chamber which in turn was mounted on a Newport rotary base (Mod. 481-Series) with an angular resolution of 0.02°. This equipment allowed the selection of the angle of incidence of the probe beam; for this work the following values were used $\theta_{01} = 0°$ and $\theta_{02} = 50°$. The signal from the microphone was amplified with a SRS preamplifier (Mod. SR550) and then sent to a SRS Lock-in Amplifier (Mod. SR-850); the modulation signal was provided by the chopper. The monochromator was controlled by a 486 PC using an ARC interface (Mod. PS-445).

Spectra were taken of the quartz and silicon substrates with and without (reference spectra) the AlN film. The final PATS data were obtained by dividing the film spectra by the reference substrate data so that the wavelength response of the experimental arrangement was eliminated.

Figure 3 shows the obtained spectra where curve (a) is the transmission spectrum of the quartz with AlN taken using a spectrophotometer (Mod. UV160U Shimadzu); these data are included as a comparison to the OPC spectrum for the quartz substrate (curve (b)), and the OPC spectrum for the silicon (curve (c)). For these figures the angle of incidence is 0°. It should be noted that the three spectra are of the same form; the main difference is that the generation process of the PA signal is distinct for the two substrates and this gives rise to the observed signal offset. The spectrum for the 50° angle of incidence was qualitatively similar to the first but with an appropriate shift in the position of the maxima and minima.

3. Analysis of the spectra

The following analysis was developed to obtain expressions which facilitated the derivation of the film thickness, d, and the refractive index, n, for the spectral range where this is only weakly dependent on the wavelength. For the

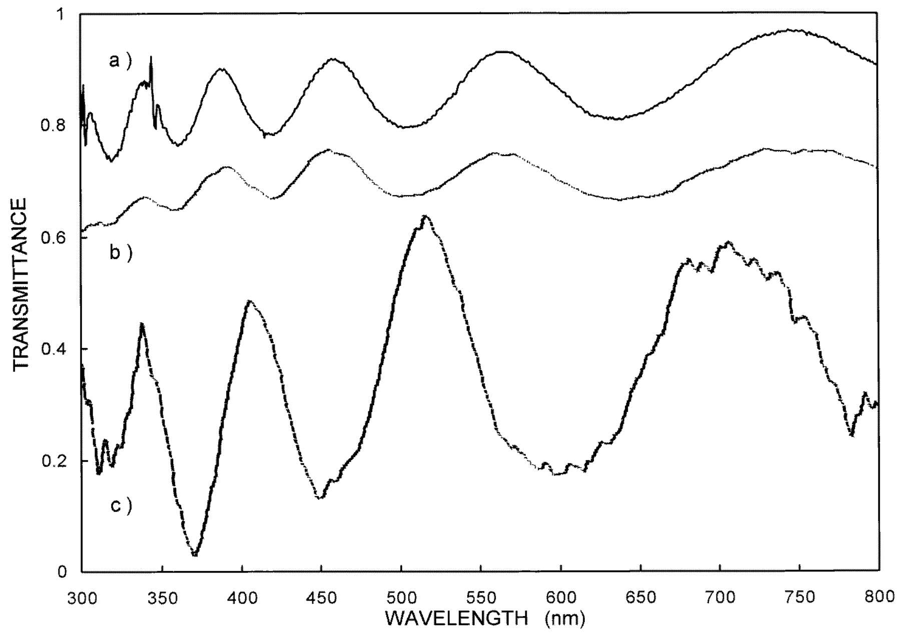

Figure 3. Transmission spectra of AlN thin films on: (a) a quartz substrate using conventional optical detection, (b) a quartz substrate with carbon black using PA detection, and (c) crystalline Si substrate using PA detection.

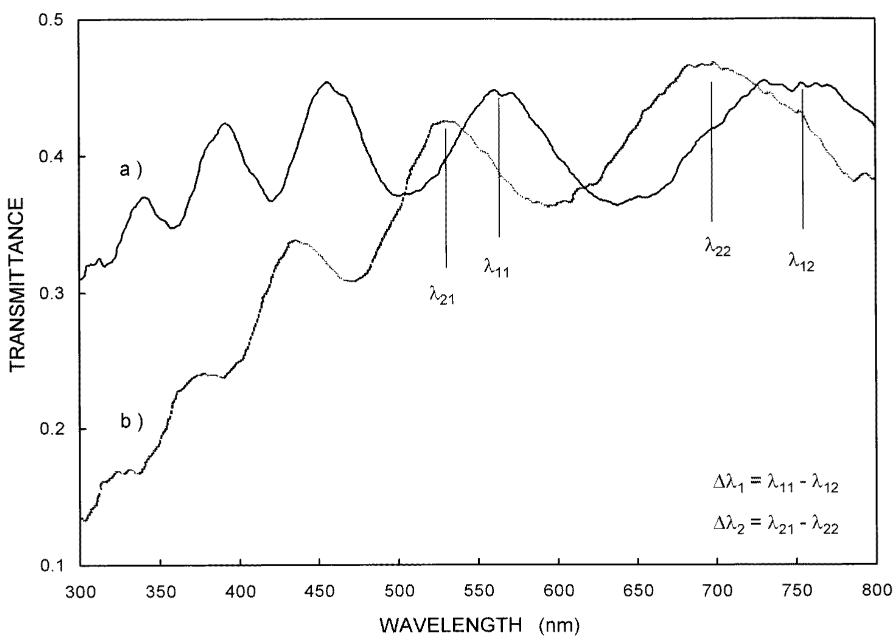

Figure 4. Thin film PATS of a quartz substrate with carbon black. Curve (a) corresponds to normal incidence (θ_{01} = 0°) and curve (b) to an angle of incidence of θ_{02} = 50°. The marks indicate the wavelength of the interference maxima.

range where $n = n(\lambda)$ a model was developed based on the harmonic oscillator model, through the optic dispersion theory [21].

Figure 4 shows the PAT data for the AlN films on quartz coated with carbon black for (a) 0° and (b) 50° angles of incidence, the wavelength positions λ_{11} and λ_{12} for spectrum 4(a) correspond to the same maxima (or minima) m and $(m+1)$ as λ_{21} and λ_{22} in the spectrum 4(b), in a range where n is approximately constant. Through a procedure similar to those used by Harrick [12], the film thickness can be expressed from equations (3) and (4), in terms of the wavelength shift of the points, $\Delta\lambda_1 = \lambda_{11} - \lambda_{12}$ and $\Delta\lambda_2 = \lambda_{21} - \lambda_{22}$, together with the ratios, $A = (\lambda_{11}\lambda_{12})/\Delta\lambda_1$ and $B = (\lambda_{21}\lambda_{22})/\Delta\lambda_2$, as

$$d = \frac{1}{2}\left(\frac{A^2 - B^2}{\sin^2\theta_{02} - \sin^2\theta_{01}}\right)^{1/2} \qquad (5)$$

where this relationship is independent of n.

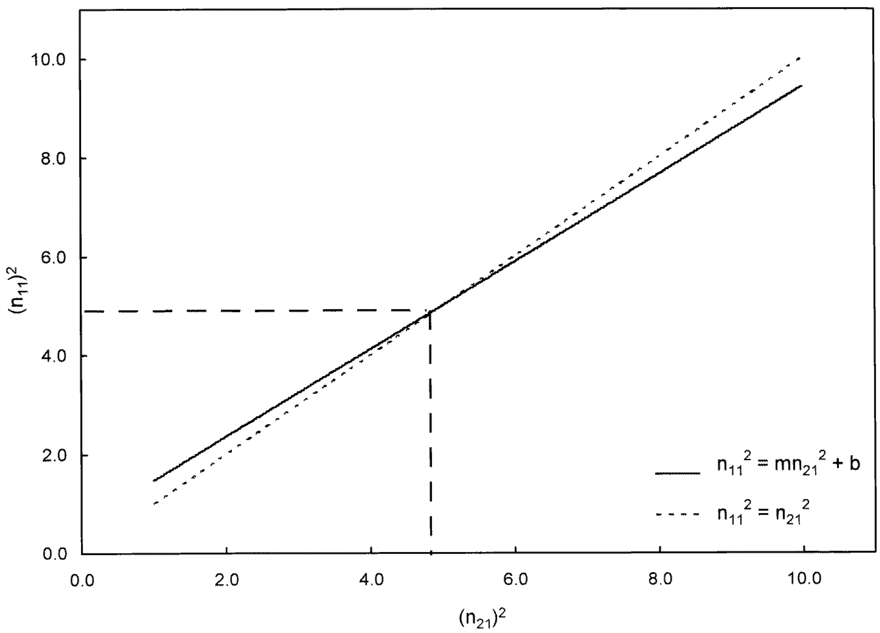

Figure 5. The solid line represents the numerical evaluation of the linear relation between n_{11}^2 and n_{21}^2. The dashed line corresponds to the relation $y = x$. The point of intersection gives the n_{11}^2 value for each interference maxima or minima in the PA spectra.

The precision of the value of d increases as more maxima (minima) are considered, as long as the distance between the sequence of maxima (minima) is a constant number of frequency units [12] and that there are no strong bands of absorption. In the contrary case it is advisable to use consecutive maxima (minima), i.e. $\Delta m = 1$.

From the same data, again using equations (3) and (4), and from a similar procedure, we obtain

$$n = \left(\frac{(A \sin\theta_{02})^2 - (B \sin\theta_{01})^2}{A^2 - B^2}\right)^{1/2} \tag{6}$$

which does not depend on d. Using these relationships we obtain values of $d = 567.07$ nm and $n = 2.14$, for the quartz substrate with uncertainties of 4 and 5%, respectively. By ellipsometry measurements of the same samples, using a He–Ne laser 632.8 nm, we obtain $d = 600$ nm and $n = 2.1$ with an uncertainty of $\sim$3%.

3.1. Variable refraction index

When $n = n(\lambda)$, then $T = T(n(\lambda))$, and the interference spectra are altered in terms of both the amplitude and the positions of the maxima. Under these circumstances equation (6) fails and $n(\lambda)$ must be found by other means. From the theory of optical dispersion, $n(\lambda)$ can be expressed in terms of the real and imaginary parts of the dielectric constant $\varepsilon = \varepsilon_1 + i\varepsilon_2$, as follows:

$$n(\omega) = \frac{1}{2}\left[(\varepsilon_1(\omega) + \sqrt{\varepsilon_1^2(\omega) + \varepsilon_2^2(\omega)})^{1/2}\right] \tag{7}$$

with

$$\varepsilon_1(\omega) = 1 + \omega_p^2 \frac{\omega_0^2 - \omega^2}{(\omega_0^2 - \omega^2)^2 + \Gamma^2\omega^2} \tag{8}$$

and

$$\varepsilon_2(\omega) = \omega_p^2 \frac{\Gamma\omega}{(\omega_0^2 - \omega^2)^2 + \Gamma^2\omega^2}. \tag{9}$$

Here ω_0 is the resonant frequency of the medium, and it takes a value of around $\omega_0 = 9.41 \times 10^{14}$ s^{-1} [22]; ω_p is the plasma frequency [23] and can be determined from the known parameters. Far from absorption region, $\varepsilon_1 \gg \varepsilon_2$, $n = n_{min}$ (the minimum value of n was obtained from equation (6)), and the resonance term in equation (8) can be considered negligible. Then equation (7) reduces to $n_{min} = 1 + \omega_p^2/(\omega_0^2 - \omega^2)$, and is satisfied by ω values outside the resonance region, where the difference $\omega_0^2 - \omega^2$ does not change substantially. Finally, Γ is a qualitative factor associated with the damping ratio of each frequency in the medium [20, 21]. It is the term responsible for the modulation of the amplitude and the variation in the position of the maxima of the PAT data.

To estimate analytically the $n(\omega)$ values within the experimental range, an iterative procedure was developed. We selected the experimental points associated with the maxima and minima in the interference figures in the PATs spectra, and built up a tendency-shape curve of Γ as function of ω using a polynomial fit and least mean squares procedure described below.

Initially for the angles of incidence θ_{01} and θ_{02}, equation (3) can be rewritten as

$$2d\sqrt{n_{11}^2 - \sin^2\theta_{01}} = m\lambda_{11} \tag{10a}$$

$$2d\sqrt{n_{21}^2 - \sin^2\theta_{02}} = m\lambda_{21}. \tag{10b}$$

For the terms n_{il} and λ_{il} the subindices i show the angle of incidence considered, whilst l is related to the value of

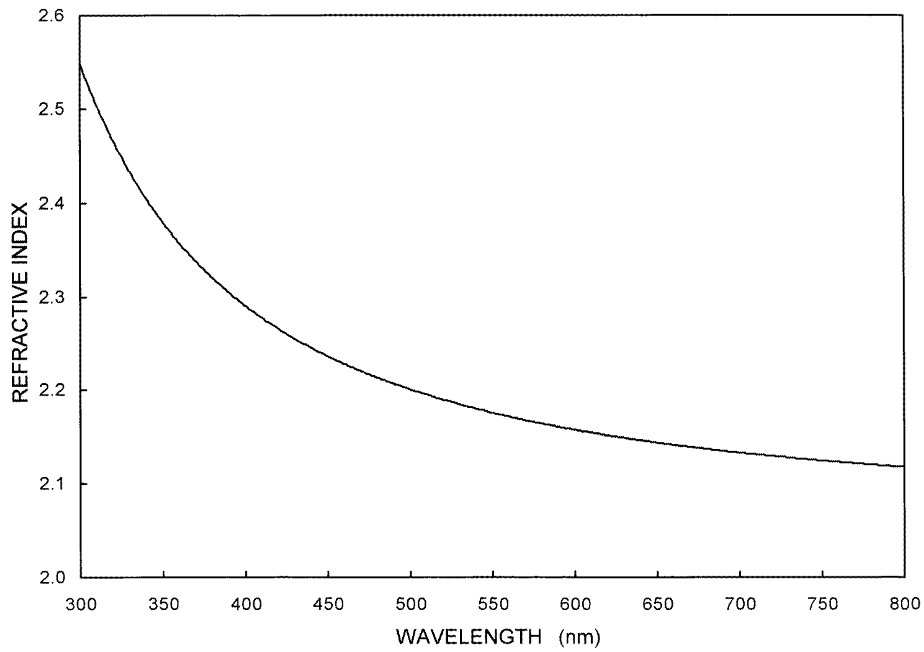

Figure 6. Calculated refractive index curve of AlN on a quartz substrate. The experimental data used in the numerical evaluation were taken from the PA spectra in figure 4.

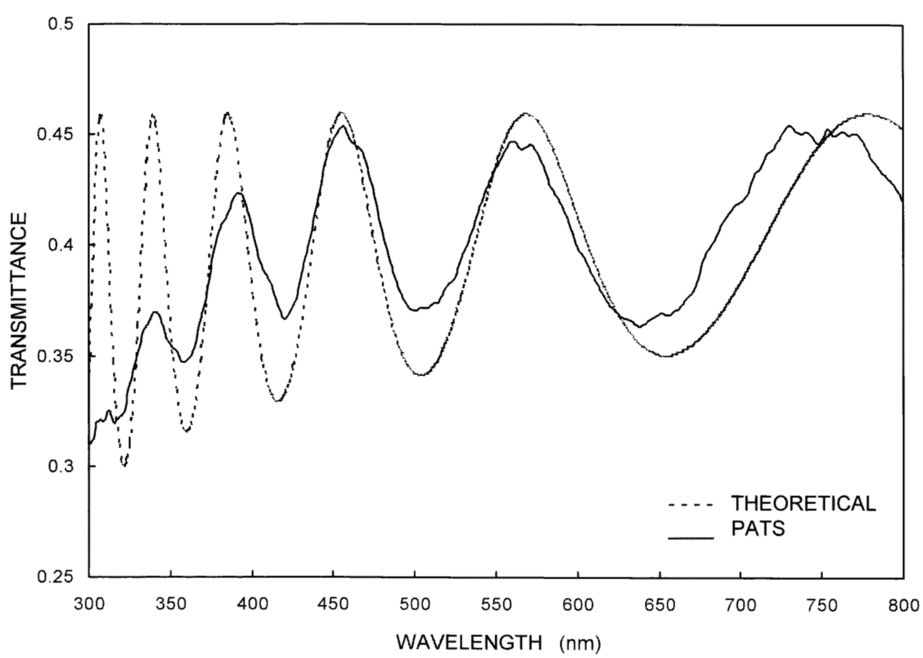

Figure 7. The experimental PA pseudo transmission spectrum (solid curve) and the numerically calculated transmittance spectrum (dashed curve), for a AlN film on a quartz substrate darkened with carbon black. The angle of incidence is 0°.

n^2 for the corresponding λ at which there is a maximum or minimum in the interference spectra. By combining these relationships we obtain

$$n_{11}^2 = \frac{\lambda_{11}^2}{\lambda_{21}^2}(n_{21}^2 - \sin^2\theta_{02}) + \sin^2\theta_{01} \qquad (11a)$$

$$n_{21}^2 = \frac{\lambda_{21}^2}{\lambda_{11}^2}(n_{11}^2 - \sin^2\theta_{01}) + \sin^2\theta_{02}. \qquad (11b)$$

It can be seen that there is a linear relation between n_{11}^2 and n_{21}^2, as shown in figure 5. The value of n^2 for each maximum (minimum) can be found from where the line in the figure crosses a 45° gradient line [13]. The distribution of these points, evaluated for the various maxima or minima as a function of λ, provides a numerical expression for Γ. Figure 6 is the $n(\lambda)$ curve obtained from these values of Γ.

Substituting the numerical n values in the equation (1), a theoretical transmittance spectrum can be generated and we have seen that this is in good agreement with the results obtained experimentally, see figure 7. We believe that this procedure establishes the uniqueness of the calculated data.

The method can be used to analyse the optical properties of thin films even if the substrate is opaque; as mentioned earlier the substrate generates the acoustic signal and this can be detected using the OPC set-up. However, if as in our case the refractive index of the substrate varies within the wavelength range used then this changes the reflection coefficient of the film–substrate interface and this must be included in the calculation. Basically, this involves using the variable refractive index analysis procedure for both the film and the substrate; this calculation is somewhat more complicated and results will be reported in the near future.

The advantage of the present procedure is that it gives values of n and d independent of the properties of each medium and this aspect means the method is quite general.

4. Conclusions

The ability of the method to successfully regenerate the experimental spectra indicates that the method is a convenient procedure to obtain the optical properties of thin films. The method can be used for both opaque and transparent substrates. Additionally, the method gives the variation of the refractive index as a function of the wavelength and the plasma frequency of the material. The model is based on physical properties of the material under study and is simpler than the conventional methods based on curve fitting by numerical analysis. Finally, the method is quite general and can be used with any type of interference spectra, absorption, reflection or transmission.

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