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Measurement of optical constants of thin polymer films using spectrally resolved white light interferometry

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Abstract. Using the spectrally resolved white light interferometry we present our experimental results on the measurement of the optical constants of thin polymer films coated on a transparent substrate. As an extension to our previous work (*J. Opt. Soc. Am.* **B12**, 1559 (1995)) on thick glass plates, we have shown here that this technique can be effectively applied to very thin polymer films also. We have improved the accuracy of our results by using the Sellmeier dispersion formula for fitting the data. From the width and position of the zero-order fringe and the frequency of modulations in the white light spectrum, the refractive index $n(\lambda)$ and thickness t of the thin polymer films are calculated. To study the accuracies involved in the technique, PVA, PMMA and PS films of varied thicknesses are coated on glass plates and the measured values are compared with ellipsometer studies.

Keywords. Optical constants; polymer films; white light interferometry.

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1. Introduction

Thin films are used as planar waveguides for processing optical signals by modulation, switching, frequency conversion, etc. To make the best use of these thin films for devices, it is essential to measure their refractive indices as a function of wavelength $n(\lambda)$ and their geometric thickness t. Depending on the coating mechanism, the thickness of the film could vary from place to place. Hence it becomes important to measure the refractive index and the thickness quite accurately and non-destructively at any specific location of the sample. A variety of techniques is available for their measurement. Most important among them are prism coupling [1], m-line method [2], integrated optical technique [3], ellipsometry [4] and interferometric techniques [5].

In the prism coupling and m-line techniques, (a) the film must be thick enough to support at least two modes to propagate film for determining n and t simultaneously, (b) it is not contact less and is suited for hard films, (c) laser lines over the whole visible range are not easily available. Ellipsometer studies are best suited for very thin films as the reflection coefficients are periodic for phase change larger than 2π. As a result, to determine the accurate values of the thickness, one would require the approximate values of the thickness for thicker films before making the ellipsometric measurements.

Among the optical measuring techniques, interferometry is highly accurate and simple in operation. The technique of spectral analysis of the interferogram is used in

the measurement of differential refractive index of liquid solution [5], construction of a thickness gauge system [6], real time measurement of material dispersion of optical glass [7], and in the measurement of polarization mode dispersion in a single mode optical fiber [8].

When the optical path difference between the two interfering beams $c\tau$ is more than the coherence length $c\tau_c$ of the light beam used, one does not see interference in the spatial domain. However, if the spectrum of the interfering beams is monitored, one observes an interference pattern in the spectral domain, the periodicity of the fringes characteristic of the delay between the two beams. By properly adjusting the path difference between the two arms of the Michelson interferometer, after the introduction of the dispersive material, the zero-order fringe can be made to appear within the region of observation. Using the fringe width and its frequency in the spectral domain, the measurement of refractive index and thickness of thick glass plates has been proposed by us recently [9]. In this communication we extend our investigations to the measurement of optical constants ($n(\lambda)$ and t) of thin polymer films coated on a thick

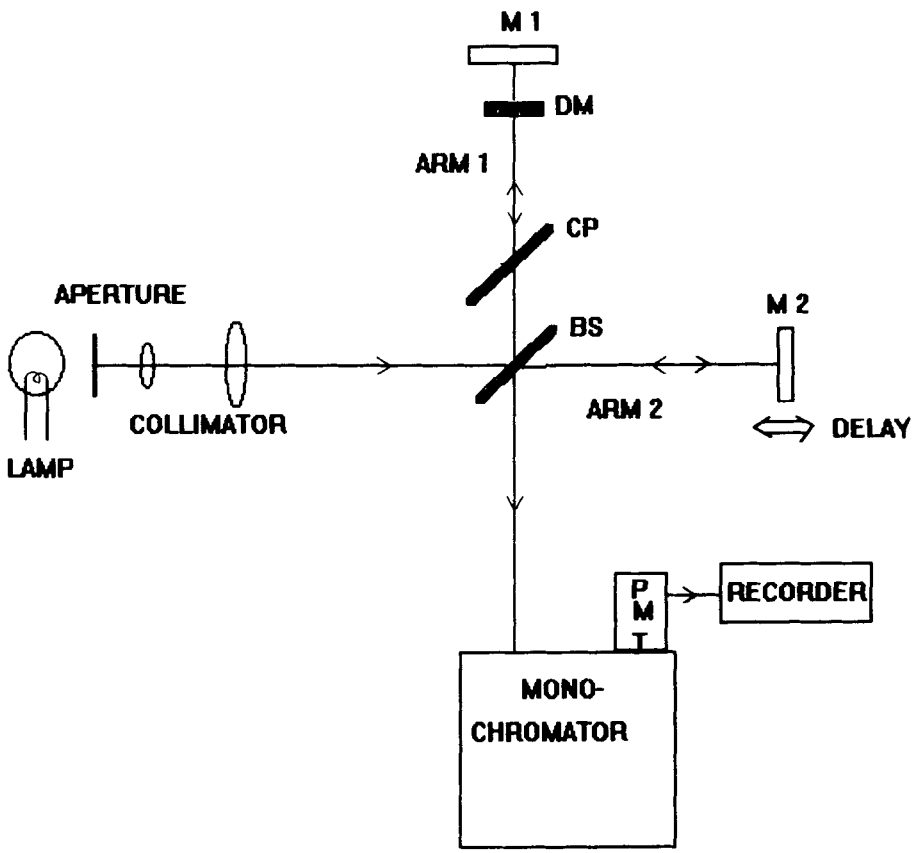

Figure 1. Schematic representation of the experimental set up: Michelson interferometer with the time delay between the interfering beams $\tau > \tau_c$ (τ_c is the correlation time of the source). BS, 50/50 beam splitter; CP, compensating plate; M1, M2, mirrors; DM, dispersive medium.

glass substrate using either spin coating technique or doctor-blade technique and show that this technique would be an alternative for the more expensive ellipsometer.

2. Experimental details

The experimental set up used for the measurements, shown in figure 1, consists of a Michelson interferometer (MI) configuration with a light source of 1 mm aperture illuminated by a 25 W tungsten filament lamp (white light source). The light beam is collimated using proper lens combination and is sent into the two arms of the MI by a 50–50 beam splitter (BS). The dispersion effects of the dielectric film coated beam splitter (with AR coating on the second surface) is compensated by introducing another one of the same material (CP) in the second arm. One mirror M_1 is fixed and another one M_2 is mounted on a translational stage with a movement accuracy of $\pm 5\,\mu$m. Light emerging from the interferometer is imaged onto the slit of a 50 cm grating spectrometer (Jobin-Yvon, HRS-2). The output from the spectrometer, collected with a PMT, is given to a strip-chart recorder (Packard-611). The recorded spectra are later digitized for analysis with a computer.

The glass plates, on which the polymer films are to be coated, are thoroughly cleaned with solvents and then dried with dry nitrogen. Care has been taken to prepare the polymer solution by filtering it. Commercially available polyvinyl alcohol (PVA) (0·3 gm) is dissolved in 10 ml of distilled water. The solution is warmed to make a clear homogeneous solution before filtering. The polymer solution is then coated on the glass substrate using the spin coating technique or doctor blade technique. The coated glass plates are kept in an oven at 40°C for 2 h to make sure that the solvents are completely removed. Similar procedure is followed to prepare the films of polystyrene (PS) dissolved in xylene and polymethyl meta acrylate (PMMA) dissolved in toluene.

3. Theory

Consider interference of two light beams described by the frequency dependent complex field amplitudes $\varepsilon_1(\omega)$ and $\varepsilon_2(\omega)$,

$$\varepsilon(\omega) = a(\omega)\varepsilon_1(\omega) + b(\omega)\varepsilon_2(\omega)\exp(-i\omega\Delta/c), \tag{1}$$

where Δ is the total path delay between the two interfering beams, $a(\omega)$ and $b(\omega)$ are the frequency dependent coefficients which depend on the properties of the beamsplitter.

In the case when both light beams $\varepsilon_1(\omega)$ and $\varepsilon_2(\omega)$ are derived from the same source, the observed spectrum at the output is given by the expression

$$S(\omega) = \tfrac{1}{2}S_0(\omega)[1 + \mathrm{Re}\{\mu_{12}(\omega)\}\cos(\omega\Delta/c + \theta)], \tag{2}$$

where $\mu_{12}(\omega)$ is the complex degree of spectral coherence of the two beams and θ is the constant phase factor. A detailed discussion of the theory has been given in our earlier communications [9, 10].

In terms of wavelength λ, the above expression can be written as

$$S(\lambda) = \frac{1}{2}S_0(\lambda)\left[1 + \mathrm{Re}\{\mu_{12}(\lambda)\}\cos\left(\frac{2\pi\Delta}{\lambda} + \theta\right)\right]. \tag{3}$$

When a glass substrate of refractive index $n(\lambda)$ and thickness t is introduced into one of the beams (say arm 1), the path difference becomes

$$\Delta = n(\lambda)2t - L_0, \tag{4}$$

where $n(\lambda)$ is the refractive index of the material over the entire visible region, given by the modified 5 constant Sellmeier dispersion relation

$$n(\lambda) = A\lambda^4 + B\lambda^2 + C + D/\lambda^2 + E/\lambda^4, \tag{5}$$

with A, B, C, D and E the dispersion constants of the glass substrate. As the refractive index is a function of λ, one can see from (3), a variation in the periodicity of the fringes with λ. The constant L_0 can be chosen such that the total phase difference becomes zero at some wavelength near the centre of the spectrum, where one sees a broad fringe with the periodicity of the fringes changing on either side. Experimentally this is achieved by translating the mirror in arm 2 and visually observing the fringes with a grating.

Equation (3) is modified as

$$S(\lambda) = \frac{1}{2}S_0(\lambda)\left[1 + \mathrm{Re}\{\mu_{12}(\lambda)\}\cos\left(\frac{2\pi(\Delta + \Delta')}{\lambda} + \theta\right)\right] \tag{6}$$

with, $\Delta' = n'(\lambda)2t'$ for the glass plate coated with a polymer film of refractive index n' and thickness t'.

4. Results and discussion

Figures 2(a) and 2(c) show the spectral interference fringes recorded with the transparent glass substrate alone, introduced into one of the arms. The intensity of the interference pattern represents the cosine of the phase at each wavelength of the radiation. After the introduction of the glass plate in arm 1, the broad fringe is brought to the centre of the spectrum by translating the mirror M_2. We refer to this broad fringe as zero-order fringe, which appears for $L_0 = n(\lambda_{st})2t + \theta\lambda_{st}/2\pi$, where λ_{st} is the stationary phase point and corresponds to the wavelength at the centre of the zero-order fringe. Since the variations of the phase i.e., $\mathrm{d}\phi/\mathrm{d}\lambda$ around this point is very slow, this zero-order fringe can be seen to be broad compared to other fringes. The stationary phase point can be located at any point in the spectrum by changing the path difference between the two arms using micrometer. From (3) and (4), one can see that the width of the fringes depends on the refractive index and thickness of the material. The modulated spectrum is fitted using (3) to get the refractive index $n(\lambda)$ and thickness t of the glass plate.

Without disturbing the inteferometer, the coated glass plate is reintroduced in the same arm. A special mount with an aperture is made for this purpose so that the same area of the glass plate is introduced into the interferometer. The spectrum recorded now (figures 2(b) and 2(d)), clearly shows the following interesting features: (1) the stationary phase point is red shifted, and (2) there are more number of fringes in the region of observation, as a result of the excess path seen in this arm. The modulated spectrum obtained with the polymer film coating is fitted with (5). The $n(\lambda)$ and t of the substrate are substituted from the previously calculated values. The values obtained for glass

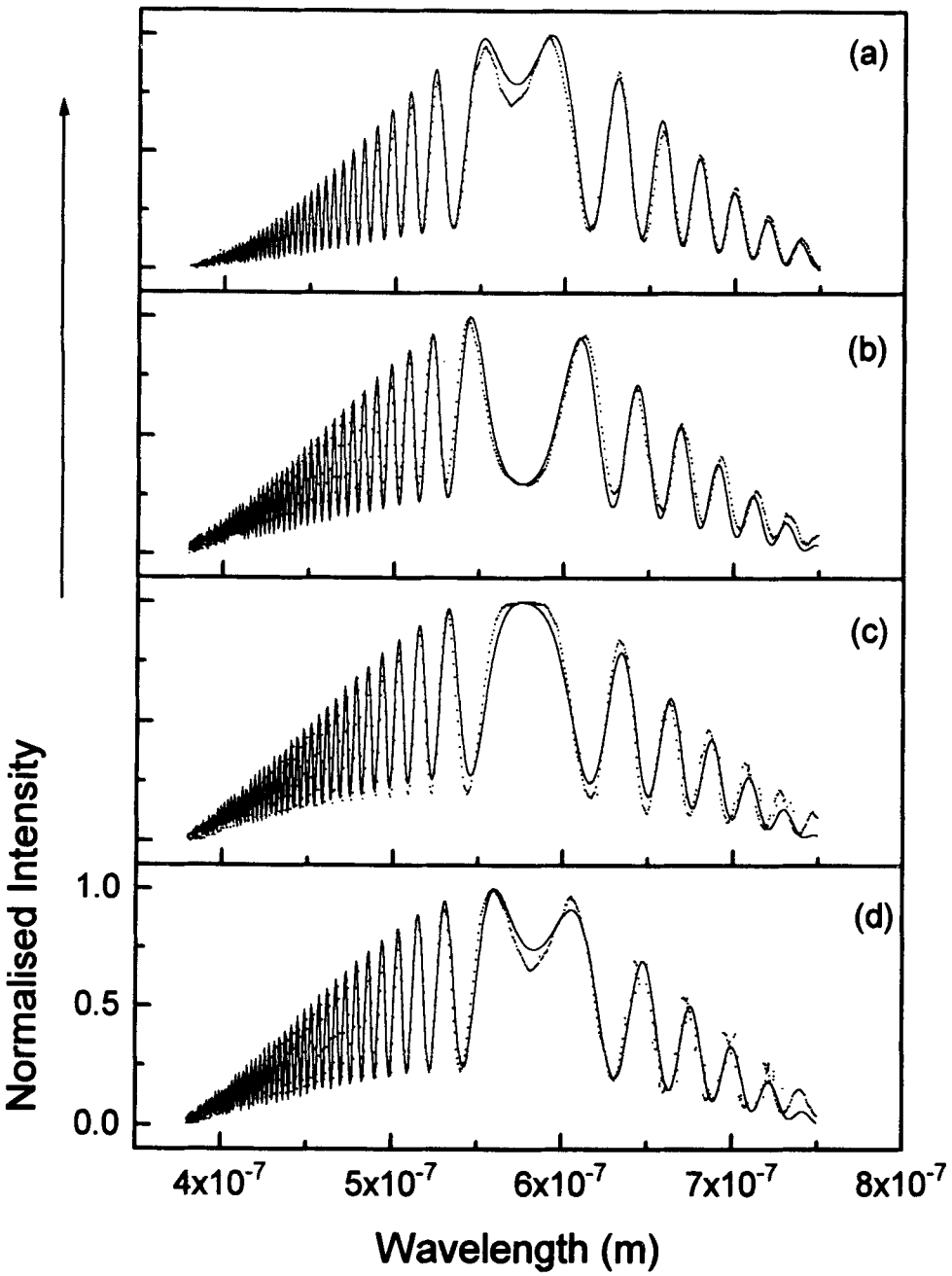

Figure 2. Normalized recordings of the spectral interference pattern (dotted line) and the theoretical fittings (continuous line) for (a) glass plate, (b) glass plate + PVA, (c) glass plate, (d) glass plate + PS.

plates and the polymer films are summarized in table 1. We estimate that the accuracies of the values so derived are in the range of 10^{-5}, that is $\pm$ 1 nm for t and $\pm$ 0·00001 for n. These accuracies are estimated through our theoretical fittings of the experimental data. A change in the fifth decimal place of either $n(\lambda)$ or t (in μm) is found to affect the fitting curves.

Table 1 also gives the values of λ_{st} for the glass substrates and the polymer films. Substituting the fitted values of the dispersion constants A, B, C, D and E of the glass plates and the corresponding constants of the polymer films in (5), the variation in the refractive index of these materials over the entire visible region is calculated and shown in figure 3.

In order to compare the accuracies of the results, we have measured n and t values with an ellipsometer. The back surface of the glass plate was properly grounded to eliminate the reflections from the second surface of the glass plate. The measured values are also given (*) in table 1. Transmission measurements are taken on a spectrometer prior to the ellipsometer studies to get the approximate thickness of the polymer films. Ellipsometer studies could not give any values for the PVA and PMMA films as their refractive indices are either very close to or lower than the glass substrate. This clearly demonstrates the usefulness of the spectral modulations technique for characterizing the thin film refractive indices and thickness over the other techniques. We have also measured the thickness and refractive indices of free standing films of PS of the order of 10–40 μm thickness. Except for the visibility (which was lower for free standing films, as they were not perfectly transparent) of the fringes, we obtained similar results. However, as there is no 1 mm glass plate, the measurement accuracies are expected to be much higher for free standing films.

5. Conclusions

We have developed a simple and relatively inexpensive method that uses a white light source, Michelson interferometer, and a spectrometer to measure simultaneously the thickness t and the refractive index $n(\lambda)$ of thin, transparent films over the entire visible region to an accuracy of about 10^{-5}. The results are compared with ellipsometer measurements and we have demonstrated that the spectral modulation technique gives more accurate values for n and t compared to the other techniques.

Table 1. Measured n and t values of the glass substrate and the polymer films.

Material	n (at 633 nm)	t ($\times 10^{-4}$ m)	λ_{st} ($\times 10^{-7}$ m) GP	λ_{st} ($\times 10^{-7}$ m) GP + PF
Glass plate	1·52075	9·99981		
PVA	1·50747	0·00333	5·67624	5·75079
PS	1·58294 (1·587)*	0·00323 (0·003237)*	5·78424	5·81699
PMMA	1·48889	0·01544	5·51117	5·72425

*Ellipsometer studies; GP: Glass plate; PF: Polymer film.

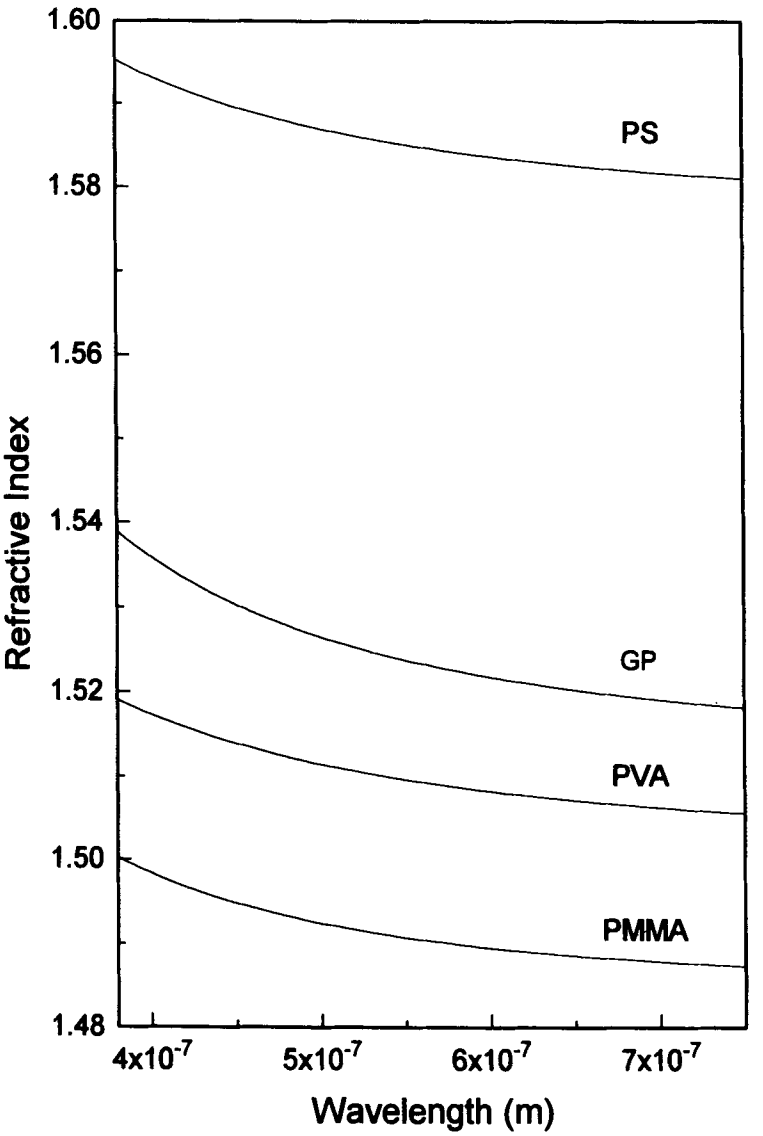

Figure 3. Plot of the refractive index variation with wavelength for the glass plate (GP) (average for all the plates) and the polymers PVA, PS and PMMA over the entire visible region.

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