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# Investigating the experimental limits of the Brewster's angle method

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**Abstract:** We present the method, analysis, and experimental results of the Brewster's angle method commonly used for determining the refractive indices of optical films. We show the significance of the intersection of reflectance curves, in that the necessity for substrate refractive index and film layer thickness knowledge are both eliminated. We present the conditions for the existence of the second intersection of reflectance curves and introduce a method for determining the refractive index of the substrate layer by using the angular information alone. Analytical results reveal impressive practical sensitivity and accuracy limits for the method, where the experimental results also support the theoretical analysis.

**Key words:** Refractive index, nondestructive measurement, Brewster's angle, optical film

## 1. Introduction

Among the numerous reported techniques, ellipsometry and prism coupling (m-line method) are among the methods commonly used for the refractive index measurement of homogeneous nonabsorbent optical films. Ellipsometry requires a complex experimental setup and involves an iterative approach to determine the optical constants and film thickness using a predefined model of the material [1]. The prism coupling method requires that a prism with a suitable refractive index be available for measuring optical films with different refractive indices and that the film be guiding at the wavelength of operation. If the film supports only a single mode, then independent knowledge of the film thickness is necessary to determine the refractive index of the film [2]. Thickness measurements via a mechanical profilometer can be destructive for soft polymeric films, which are often used as optical materials. Scanning electron microscopy is a useful tool for thickness measurements and visual assessment of the surface quality. However, it should be noted that, depending on the material, samples may have to be coated with conductor materials and may become unusable for further measurements. For some materials like photopolymers, which is the most important issue in our research field, electron microscopy may itself modify the chemical structure of the material during the measurement. Therefore, refractive index measurement methods that eliminate the necessity for the film thickness and/or the substrate refractive index information are essential.

An alternative method for the refractive index measurement of polymer optical films involves computational fitting of recorded diffraction patterns, where refractive index modulated polymer films are used as phase objects in a plane-wave diffraction setup [3].

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Brewster's angle method is a technique based on very limited penetration, frequently used for measuring refractive indices of optical materials. Brewster's angle based methods are usually preferred over other measurement techniques due to the simple experimental setup and straightforward calculations required. With the appropriate selection of the employed laser wavelength, refractive indices of photosensitive films can be measured without exposing the photoactive materials, i.e. nondestructive measurements are achievable. Reliable results are easily obtained as long as the surface and interfaces of the measured samples are smooth.

While the method is a straightforward tool for measuring the refractive index of bulk systems [4], for the measurement of ambient-film-substrate systems, fitting is necessary in order to obtain precise values [5]. To measure the refractive index with high sensitivity, a combined approach involving a separate measurement of the film thickness has also been reported, where the results were enhanced using computer simulations and data-fitting for the measured values [6].

The Abeles–Brewster method is an improved version of the technique that eliminates the necessity to obtain the thickness of the optical film layer [7]. This method requires an additional reflectance measurement of the uncoated substrate layer to be compared with the system measurement [8]. Using Brewster's angle based methods, film refractive index measurement sensitivities of 0.0065, 0.0056 (relative error: 0.3%), 0.0050, and 0.0020 were reported in [9], [5], [10], and [8] respectively in accordance with descending order.

This paper presents a theoretical, analytical, and experimental investigation of the Brewster's angle method based on the significance of intersections of reflectance curves for samples with different thicknesses. The analysis reveals that no prior theoretical and empirical knowledge of the material properties is needed. In addition, the necessity to know the substrate refractive index and the film layer thickness is eliminated. It is also shown that it is possible to determine the substrate refractive index using the intersection angles alone, without having to know the properties of any other layers in the ambient-film-substrate system. Experimental sensitivity and accuracy limit analyses of the method have been performed using an analytical approach. Experimental measurements were carried out to support the theoretical studies and the results are presented.

## 2. Background

The reflection coefficient of p-polarized light from the interface of two dielectric media with refractive indices $n_1$ and $n_2$ is given by the Fresnel equations as [11]:

$$r_p=\frac{n_2\cos\theta_1-n_1\cos\theta_2}{n_2\cos\theta_1+n_1\cos\theta_2} \tag{1}$$

where $\theta_1$ and $\theta_2$ are the angles of incidence and refraction, respectively. If $n_1 < n_2$, the reflection coefficient completely vanishes at angle $\theta_1 = \theta_B$, i.e. the Brewster's angle, expressed as [11]:

$$\theta_B=\tan^{-1}\frac{n_2}{n_1} \tag{2}$$

Measurement of the refractive index using the Brewster's angle technique involves the illumination of the material with p-polarized light while the angle of incidence is varied to obtain the angle of minimum reflectance. The angle of minimum reflectance occurs at Brewster's angle, and if the ambient medium is air, i.e. $n_1 = 1$, the refractive index of the material is found simply by taking the tangent of the angle of minimum reflectance.

When the optical material is in the form of a film rather than bulk material, the reflection obtained from the material becomes the convolution of the ambient-film-substrate system (Figure 1).

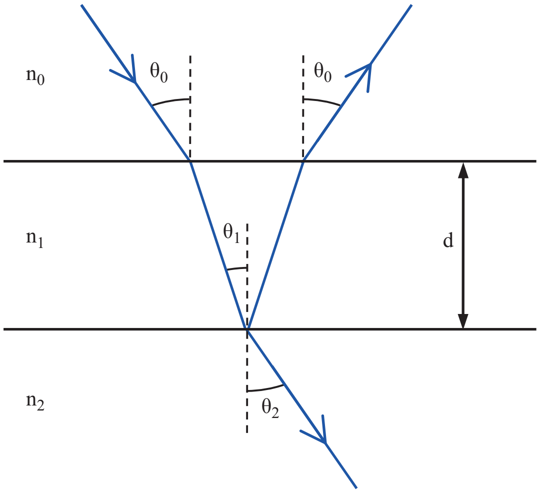

**Figure 1**. Ambient-film-substrate model used in the calculations.

Here $n_0$, $n_1$, and $n_2$ represent the ambient, film and substrate refractive indices, respectively; $d$ is the film thickness; $\theta_0$ and $\theta_1$ are the angle of incidence for the incoming beam and the angle of refraction for the beam refracted in the film layer, respectively; and $\theta_2$ is the angle of refraction in the substrate region. All layers are assumed to be homogeneous and nonabsorbent.

The reflectance $R(\theta)$ for p-polarized light of an ambient-film-substrate system has the following form [12]:

$$R(\theta)=\left|\frac{r_{01}+r_{12}e^{-2i\beta}}{1+r_{01}r_{12}e^{-2i\beta}}\right|^2 \tag{3}$$

where $r_{01}$ and $r_{12}$ are Fresnel reflection coefficients at the ambient-film and film-substrate interfaces, respectively, and $\beta=(2\pi/\lambda_0)\,n_1 d\cos\theta_1=(2\pi/\lambda_0)\,d\sqrt{n_1^2-n_0^2\sin^2\theta_0}$, where $\lambda_0$ is the wavelength in vacuum. In order to estimate the refractive indices, the substrate refractive index and film thickness, namely $n_2$ and $d$, must be known.

## 3. Limits of the method

### 3.1. Theory

The reflectivity of a homogeneous dielectric film located between two homogeneous media for p-polarized light is expressed as given in Eq. (3). For mathematical convenience, the reflectance expression can be written as [12]:

$$R(n_0,n_1,n_2,\theta_0,d,\lambda_0)=\frac{r_{01}^2+r_{12}^2+2r_{01}r_{12}\cos 2\beta}{1+r_{01}^2r_{12}^2+2r_{01}r_{12}\cos 2\beta} \tag{4}$$

where $\beta=\beta(n_0,n_1,\theta_0,d,\lambda_0)$, $r_{01}=r_{01}(n_0,n_1,\theta_0)$, and $r_{12}=r_{12}(n_0,n_1,n_2,\theta_0)$. Using Snell's law and trigonometric identities, the Fresnel coefficients can be written as:

$$r_{01}=\frac{n_1\cos\theta_0-n_0\sqrt{1-\left(\frac{n_0}{n_1}\sin\theta_0\right)^2}}{n_1\cos\theta_0+n_0\sqrt{1-\left(\frac{n_0}{n_1}\sin\theta_0\right)^2}} \tag{5}$$

$$r_{12}=\frac{n_2\sqrt{1-\left(\frac{n_0}{n_1}\sin\theta_0\right)^2}-n_1\sqrt{1-\left(\frac{n_0}{n_2}\sin\theta_0\right)^2}}{n_2\sqrt{1-\left(\frac{n_0}{n_1}\sin\theta_0\right)^2}+n_1\sqrt{1-\left(\frac{n_0}{n_2}\sin\theta_0\right)^2}} \tag{6}$$

Considering the case where the angle of incidence of the incoming beam is exactly the Brewster's angle for the ambient-film interface, i.e. $\theta_0=\tan^{-1}(n_1/n_0)=\theta_B$, it can easily be seen that:

$$r_{01}\left(\theta_B=\theta_0\right)=0, \tag{7}$$

$$r_{12}\left(\theta_B=\theta_0\right)=\frac{n_1n_2-\frac{n_1}{n_2}\sqrt{n_0^2n_2^2+n_1^2n_2^2-n_0^2n_1^2}}{n_1n_2+\frac{n_1}{n_2}\sqrt{n_0^2n_2^2+n_1^2n_2^2-n_0^2n_1^2}} \tag{8}$$

$$\beta\left(\theta_B=\theta_0\right)=\frac{2\pi}{\lambda_0}dn_1^2\frac{1}{\sqrt{n_0^2+n_1^2}} \tag{9}$$

Substituting Eq. (7) in the reflectance equation (Eq. (4)) yields the expression for the reflection when the angle of incidence equals the Brewster's angle for the ambient-film interface:

$$R\left(\theta_B=\theta_0\right)=r_{12}^2 \tag{10}$$

which is a function of $n_0$, $n_1$, and $n_2$, i.e. $R(\theta_B=\theta_0)=R(n_0,n_1,n_2)$ (see Eq. (8)). This means that when the angle of incidence is equal to the Brewster's angle for the ambient-film interface, the value of the reflection does not depend on the thickness of the film, $d$. i.e. if $\theta_0=\theta_B$, then:

$$\frac{\partial R}{\partial d}=\frac{\partial r_{12}^2}{\partial d}=0. \tag{11}$$

The interpretation of the above expression implies the following: for a set of reflectance measurements performed on different samples with the same material in the film layer, the measured reflectance value at the Brewster's angle will always be the same, regardless of the film thickness. Consequently, by a set of measurements performed on samples with different thicknesses, the refractive index of the film can be found without any necessity to know the actual thickness of the film layers themselves. This property was also observed in [13] and partially analyzed in [14] and [15]. In this study, which was outlined in [16], mathematical properties of the above technique are utilized for further investigation; in addition, the significance of the method is demonstrated based on consequent derivations, numerical analysis, and experimental studies.

It should be noted here that, for the configuration presented in Figure 1, if $\theta_0=\theta_B$, obtaining a zero reflection coefficient for any given film thickness requires either $n_2=n_0=1$ or $n_1=n_2$ in Eq. (8), which implies the structure to be either a film layer sandwiched by ambient on both sides or a single-interface structure. Therefore, if $\theta_0=\theta_B$, it is not possible to reduce the reflection coefficient of a film-covered substrate to zero for any film thickness.

### 3.2. Calculations

In order to illustrate the uniqueness of the intersection point, reflectance curves of various samples with film layer thicknesses varying between 0.5 $\mu$m and 300 $\mu$m have been calculated for a laser wavelength of 633 nm, where an angular measurement resolution of 0.01° was assumed (Figure 2).

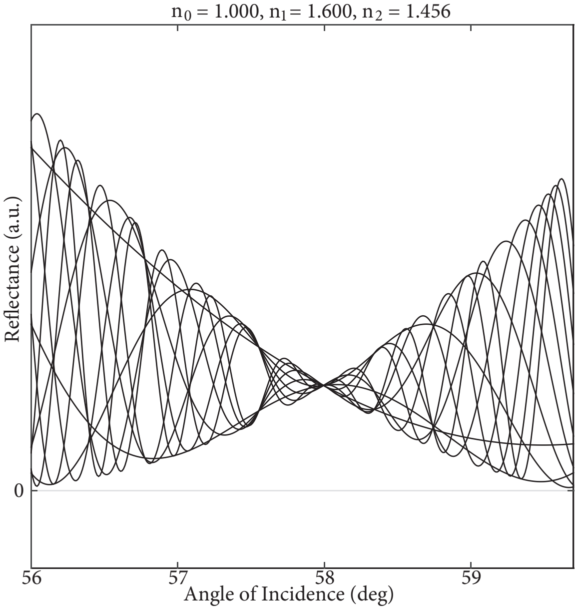

**Figure 2**. Intersection of reflectance curves for different thicknesses.

Although measuring only two samples with different thicknesses would be sufficient to determine the refractive index of the film, a higher number of measurements leads to assurance in determining the exact angular point of intersection.

Moreover, if the same dielectric film is coated on an alternative substrate with a different refractive index, the angular location of the intersection point remains the same. The difference is only in the value of reflectance at the point of intersection. This variation can be visualized as the intersection point shifting upwards or downwards on the reflectance graph with shift direction depending on the substrate refractive index. What this means is that with this method neither the thickness of the film layer nor the refractive index of the substrate needs to be known in order to calculate the refractive index of the film layer. As stated previously, once the angle of intersection is determined, the refractive index of the film layer can easily be calculated since the angle of intersection corresponds to the Brewster's angle for the ambient-film interface. That is:

$$\tan\theta_{int}=\frac{n_1}{n_0}\Longrightarrow n_1=n_0\tan\theta_{int} \tag{12}$$

In addition, further investigation shows that a second intersection of the reflectance curves occurs due to $r_{12}$ becoming zero at the angle of incidence, calculated by:

$$\theta_0=\sin^{-1}\left(\frac{n_1 n_2}{n_0}\frac{1}{\sqrt{n_1^2+n_2^2}}\right) \tag{13}$$

if a structure as in Figure 1 satisfies either of the below conditions:

$$n_1\leq\frac{n_2}{\sqrt{n_2^2-1}} \tag{14}$$

$$n_2\leq\frac{n_1}{\sqrt{n_1^2-1}} \tag{15}$$

The conditions of existence for this second intersection are illustrated in Figure 3. The second intersection, an example of which is depicted in Figure 4, occurs because either of the conditions given in Eqs. (14) and (15) makes the Brewster's angle for the second intersection smaller than the critical angle for the interface, permitting it to exist within the measurable range.

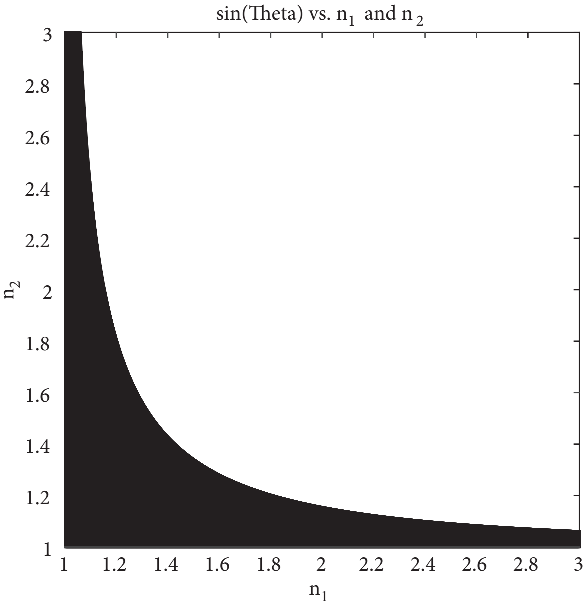

**Figure 3**. Region of existence (shaded area) for the second intersection.

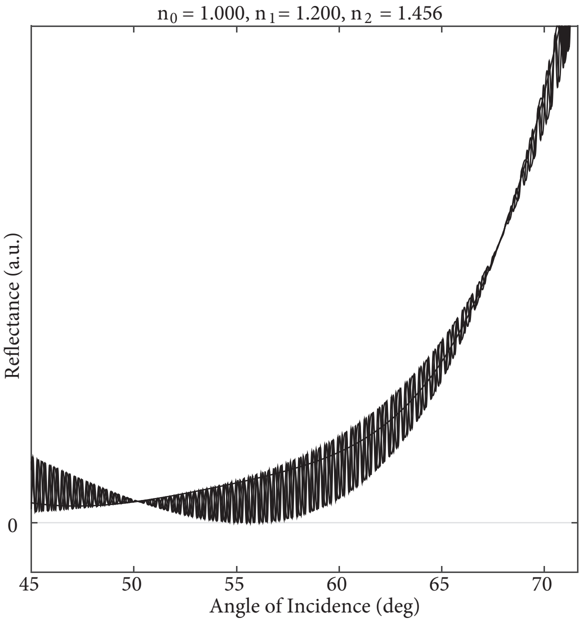

**Figure 4**. Intersection of reflectance curves for different thicknesses: intersection angles correspond to the Brewster's angles for $n_0$-$n_1$ and $n_1$-$n_2$ interfaces.

### 3.3. Substrate refractive index

If two intersection points exist for the reflectance curves (as in Figure 4) of an ambient-film-substrate configuration, it can be shown that the substrate refractive index can be determined using the angular data alone. If the intersection angles of the curves are obtained using the measurements, it is possible to determine the substrate refractive index without having to know either the refractive index ($n_1$) or the thickness ($d$) of the film layer. It can be shown that, using Eqs. (12) and (13) and assuming the ambient is air ($n_0$= 1), the substrate refractive index can be found using:

$$n_2 = \frac{\alpha\beta}{\sqrt{\alpha^2 - \beta^2}} \tag{16}$$

where $\alpha = \tan(\theta_{i1})$, $\beta = \sin(\theta_{i2})$, and $\theta_{i1}$ and $\theta_{i2}$ are angular values of the first and second intersection points, respectively ($\theta_{i1} < \theta_{i2}$).

### 3.4. Sensitivity

The sensitivity of the method was investigated by studying the reflectance curves at the intersections in detail. Sensitivity was defined by the angular width of the uncertainty region, which is determined by the loss of linearity in the envelope enclosing multiple reflectance curves. The angular width was taken as the error margin determining the ambiguity of the results experimentally achievable by the method, since mathematically there can be no ambiguous region defined for the theoretical intersection point.

For the configuration including a single intersection of reflectance curves as in Figure 2, the close study of the intersection indicates an angular uncertainty of $\Delta\theta = 0.000815^\circ$, as shown in Figure 5. Accordingly, for the single intersection configuration, the maximum error in the refractive index measurement corresponds to $\Delta \mathrm{n} = 1.4224 \times 10^{-5}$.

For the configuration including two intersections as shown in Figure 4, the angular width of uncertainty for the first intersection was determined as $\Delta\theta = 0.00163^\circ$, which corresponds to a maximum error in refractive index of $\Delta \mathrm{n} = 2.8449 \times 10^{-5}$ (see Figure 6).

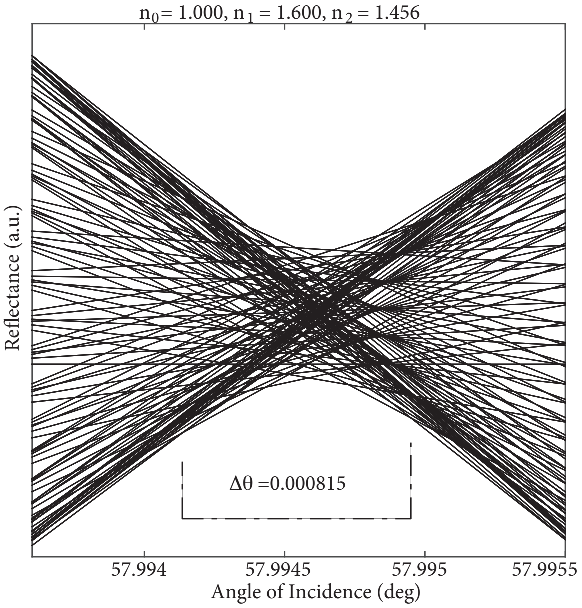

**Figure 5**. Close study of the intersection point for the single-intersection configuration.

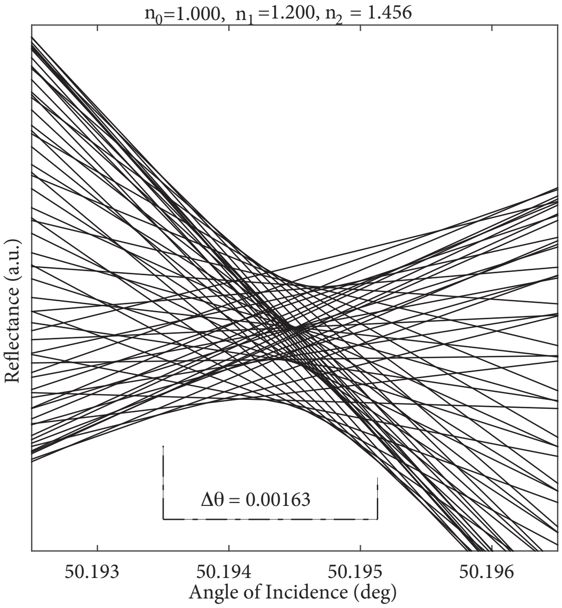

**Figure 6**. Close study of the first intersection for the two-intersection configuration.

For the same configuration, the angular width uncertainty for the second intersection was determined as $\Delta\theta = 0.0014^\circ$, corresponding to a maximum error in substrate refractive index of $\Delta \mathrm{n} = 2.0541 \times 10^{-3}$ (see Figure 7).

### 3.5. Accuracy

Accuracy analysis of the method was performed by studying the exact location of intersection points using only the obtained reflectance plots. Intersection locations were determined by tracing the linear sections of the envelopes enclosing the reflectance curves and then the refractive indices corresponding to central intersection points were calculated. For film refractive index $n_1$, relative errors of the analytically determined refractive indices were calculated with respect to the actual refractive indices. In configurations with two intersection points, substrate refractive indices $n_2$ were obtained by using the intersection points $\theta_{i1}$ and $\theta_{i2}$ with Eq. (16), and relative errors were calculated with respect to the actual values of the substrate refractive index.

The analysis of the intersection point for the configuration with a single intersection point given in Figure 5 is presented in Figure 8. Results indicate a relative error rate of $1.5464 \times 10^{-5}\%$ in determining the optical film refractive index.

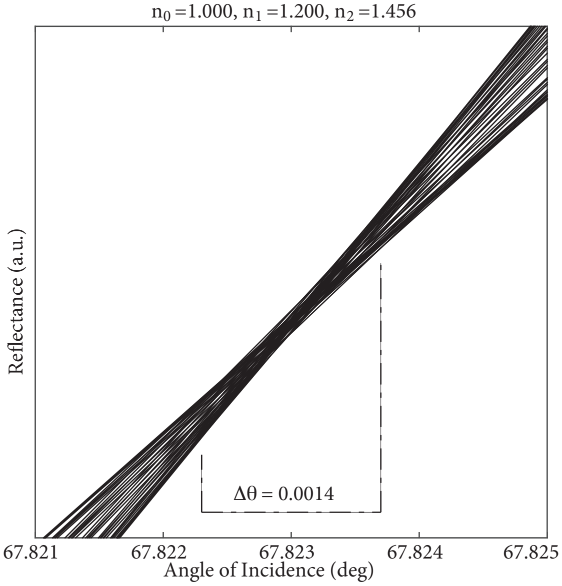

**Figure 7**. Close study of the second intersection for the two-intersection configuration.

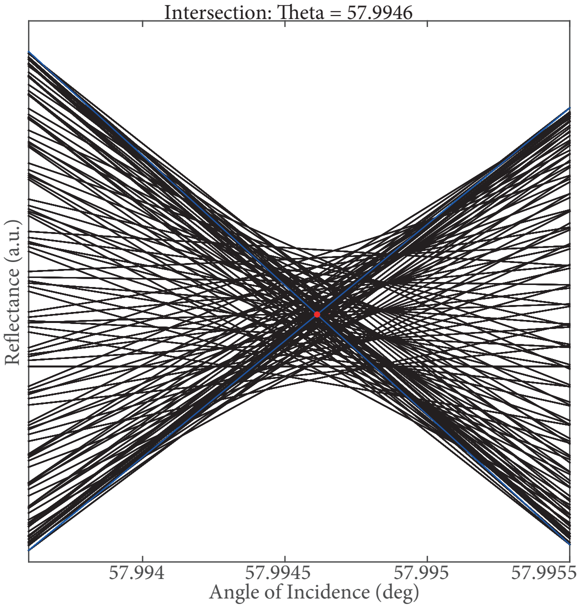

**Figure 8**. Exact point of intersection for the single-intersection configuration.

For the configuration with two intersections presented in Figures 6 and 7, the analysis of the exact intersection points indicates a relative error rate of $2.9897 \times 10^{-5}\%$ and $1.2772 \times 10^{-5}\%$ in determining the film and substrate refractive indices, respectively (see Figures 9 and 10). The results given above, as well as calculations for additional configurations, are presented together in the next section.

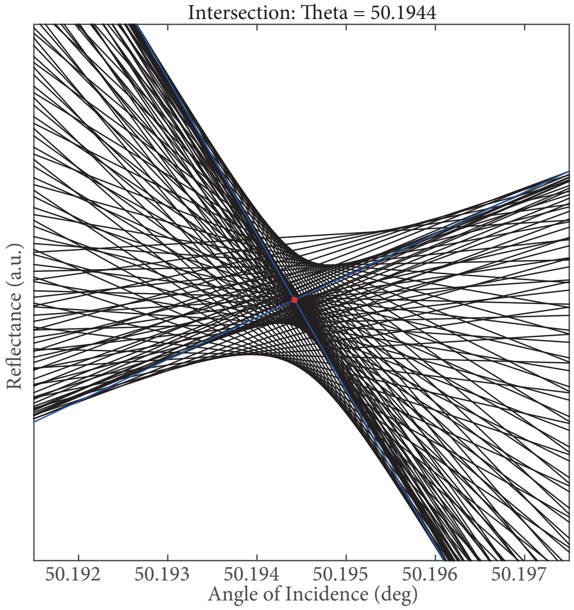

**Figure 9**. Exact point of intersection for the first intersection of the two-intersection configuration.

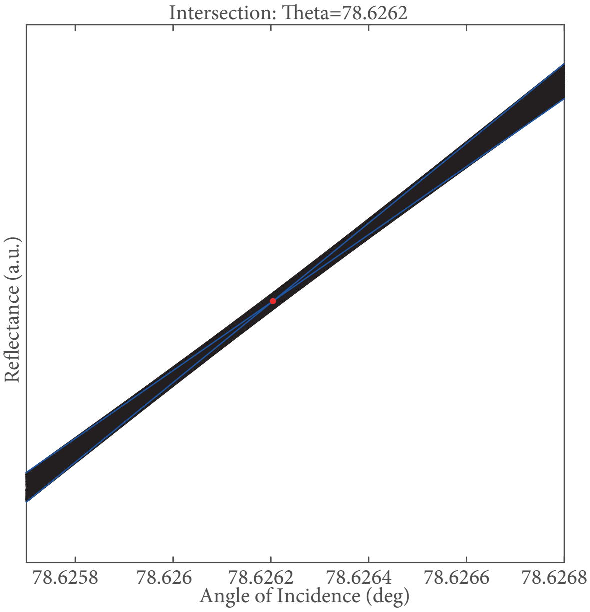

**Figure 10**. Exact point of intersection for the second intersection of the two-intersection configuration.

## 4. Numerical results

Results of the sensitivity analysis are presented in Table 1. Corresponding to an angular resolution of 0.01° in the measurements, results indicate a maximum experimental refractive index sensitivity of up to 0.000014 for the measurement of the film layer, which is well beyond the sensitivity figures reported earlier. It also presents a maximum experimental sensitivity for the measurement of substrate refractive index of 0.002054.

**Table 1**. Sensitivity analysis of the method.

| Configuration | | | Calculation | | | |
|---|---|---|---|---|---|---|
| $n_0$ | $n_1$ | $n_2$ | $\Delta\theta_{i1}$ | $n_1$ sensitivity | $\Delta\theta_{i2}$ | $n_2$ sensitivity |
| 1.000 | 1.600 | 1.456 | 0.000815° | $1.4224 \times 10^{-5}$ | - | - |
| 1.000 | 1.200 | 1.456 | 0.001630° | $2.8449 \times 10^{-5}$ | 0.001400° | $2.0541 \times 10^{-3}$ |

Results of the accuracy analyses for the film refractive index (RI) based on the first intersection and the substrate refractive index based on the second intersection are presented in Tables 2 and 3, respectively. In both tables, determined values for the intersection angles are only shown up to the third digit and error figures are only shown up to the fourth digit for convenience.

**Table 2**. Accuracy analysis of the method for the first intersection.

| Configuration | | | Calculation | | Relative error (%) |
|---|---|---|---|---|---|
| $n_0$ | $n_1$ | $n_2$ | $\theta_{i1}$ | Absolute RI error | |
| 1.000 | 1.200 | 1.456 | 50.194° | $9.8055 \times 10^{-7}$ | $8.1713 \times 10^{-5}$ |
| 1.000 | 1.200 | 1.700 | 50.194° | $3.5876 \times 10^{-7}$ | $2.9897 \times 10^{-5}$ |
| 1.000 | 1.245 | 1.483 | 51.228° | $4.2595 \times 10^{-6}$ | $3.4214 \times 10^{-4}$ |
| 1.000 | 1.245 | 1.513 | 51.228° | $1.1016 \times 10^{-7}$ | $8.8487 \times 10^{-6}$ |
| 1.000 | 1.455 | 1.522 | 55.499° | $1.2927 \times 10^{-7}$ | $8.8846 \times 10^{-6}$ |
| 1.000 | 1.581 | 1.483 | 57.686° | $7.4596 \times 10^{-7}$ | $4.7183 \times 10^{-5}$ |
| 1.000 | 1.600 | 1.456 | 57.994° | $2.4741 \times 10^{-7}$ | $1.5464 \times 10^{-5}$ |
| 1.000 | 1.875 | 1.513 | 61.927° | $1.4585 \times 10^{-8}$ | $7.7789 \times 10^{-7}$ |
| 1.000 | 1.900 | 1.700 | 62.241° | $1.0293 \times 10^{-7}$ | $5.4174 \times 10^{-6}$ |

**Table 3**. Accuracy analysis of the method for the second intersection.

| Configuration | | | Calculation | | Relative error (%) |
|---|---|---|---|---|---|
| $n_0$ | $n_1$ | $n_2$ | $\theta_{i2}$ | Absolute RI error | |
| 1.000 | 1.200 | 1.456 | 67.822° | $6.3753 \times 10^{-7}$ | $4.3787 \times 10^{-5}$ |
| 1.000 | 1.200 | 1.700 | 78.626° | $1.2002 \times 10^{-6}$ | $7.0603 \times 10^{-5}$ |
| 1.000 | 1.245 | 1.483 | 72.464° | $6.4601 \times 10^{-6}$ | $4.3561 \times 10^{-4}$ |
| 1.000 | 1.245 | 1.513 | 74.021° | $8.0807 \times 10^{-6}$ | $5.3409 \times 10^{-4}$ |

Each of the configurations listed in Table 2 was checked via Eqs. (14) and (15), for the existence of a second intersection of reflectance curves. For the configurations in which a second intersection exists, angular values of second intersection $\theta_{i2}$ were calculated and corresponding substrate refractive indices were calculated using $\theta_{i1}$ values from Table 2 and Eq. (16).

For the optical film measurement, results of the accuracy analysis show a minimum accuracy of 4.25 $\times$ $10^{-6}$ in refractive index value, corresponding to a relative error rate of 3.42 $\times$ $10^{-4}$%. In determining the substrate refractive index, results show a minimum accuracy of 8.08 $\times$ $10^{-6}$ in refractive index value, corresponding to an error rate of 5.34 $\times$ $10^{-4}$%. In addition to the minimum accuracy, the optical film measurement results also indicate that it is possible to reach a maximum accuracy of 1.45 $\times$ $10^{-8}$ using the method, corresponding to a minimum relative error of 7.77 $\times$ $10^{-7}$%.

## 5. Experimental results

For the experimental verification of the calculated sensitivities, the angular reflectance measurement system illustrated in Figure 11 was set up. The computer controlled setup includes a 632.8 nm He-Ne laser for measuring the angular reflectance of the samples, where the measured reflectance values were simultaneously normalized against the source output to eliminate any noise factors and variations in the source power.

In the study, angular reflectance characteristics of three samples of AA/PVA (acrylamide/polyvinyl alcohol) film-covered glass substrates with film thicknesses of 75 $\mu$m, 80 $\mu$m, and 90 $\mu$m were measured and the obtained results are presented in Figure 12. It is known from earlier bulk measurement studies that the polymer has a refractive index of 1.455 at 632.8 nm wavelength. The chemical composition and preparation process of the polymer film samples were described elsewhere [3].

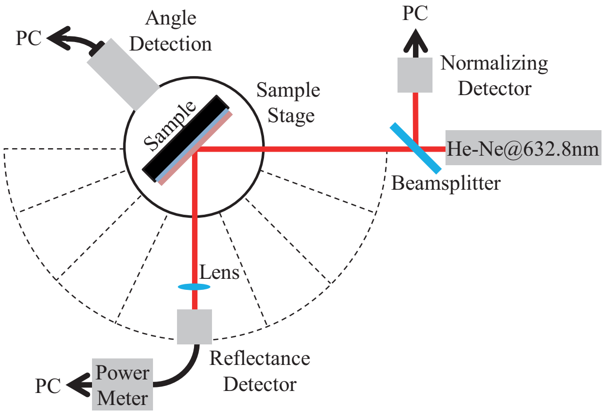

**Figure 11**. Experimental setup.

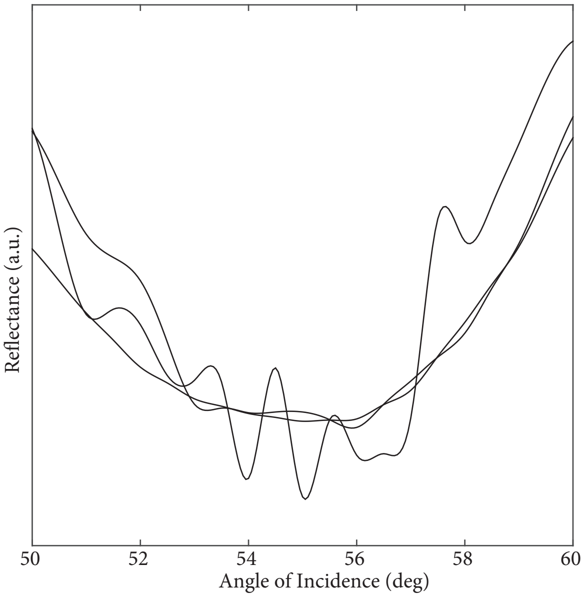

**Figure 12**. Reflectance curves of the three film-on-glass samples.

Close study of the reflectance curves presented in Figure 13 reveals the intersection point at $\theta_i = 55.5012^\circ$, corresponding to an absolute refractive index error of 7.1707 $\times$ $10^{-5}$ and a relative error of 4.9283 $\times$ $10^{-3}$%. (It is worth noting that a mild zoom into the resultant figure reveals that the data point close to 53.5° is not an actual intersection.) The configuration under investigation and the overall results are reviewed in Table 4.

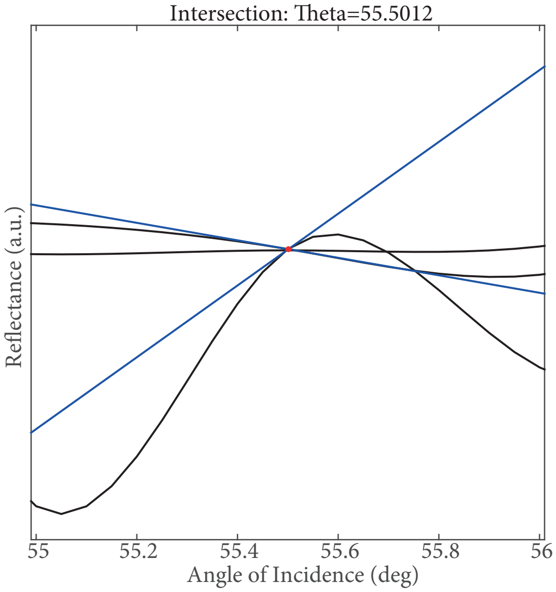

**Figure 13**. Exact point of intersection analysis for the reflectance curves.

**Table 4**. Experimental results.

| Configuration | | | Measurement results | | Relative error (%) |
|---|---|---|---|---|---|
| $n_0$ | $n_1$ | $n_2$ | $\theta_i$ | Absolute RI error | |
| 1.000 | 1.455 | 1.522 | 55.501° | $7.1707 \times 10^{-5}$ | $4.9283 \times 10^{-3}$ |

Considering the absolute and relative error values achieved through the experimental results, it can be seen that the calculations regarding the maximum accuracy of the method hold. Even though only three samples were involved in the experimental studies, the results obtained demonstrate that highly accurate results are readily achievable. Increasing the number of samples used in the measurements would further improve the achieved results and bring the experimental results closer to the calculated limits.

While the experimental results indicate that the measured sample structure leads to a single intersection of reflectance curves, this result is also confirmed by cross-checking the conditions of Eq. (14) or (15) with the obtained results.

## 6. Conclusion

In this work, with the Brewster's angle method it has been shown theoretically and experimentally that neither the thickness of the film layer nor the refractive index of the substrate needs to be known in order to obtain the film refractive index. The conditions for the existence and the angular value of the second intersection were presented and a new method was introduced for the measurement of the substrate refractive index by only using the intersection points.

Assuming an angular measurement resolution of 0.01° in the setup, the method has been shown to have a maximum experimental film refractive index sensitivity of 0.000014 and maximum substrate refractive index sensitivity of 0.002054. Furthermore, experimental verification of the study led to an absolute error in film refractive index measurement of 0.000072 and a relative error of 0.0049%, where both error values are subject to further improvement by increasing the number of samples employed in the measurements.

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