THE PRISM-COUPLED WAVEGUIDE METHOD EXTENDED TO ANISOTROPIC FILMS

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SUMMARY

In that area, limited correlation is usual among quantitative results obtained from different techniques such as ellipsometry, prism-coupled waves, Abeles method or spectrophotometry [3], [4]. A joint work by seven prominent laboratories, each employing its favorite technique, showed that, for Sc2O3 films deposited in the same run and under very similar conditions, refractive index results showed significant discrepancies [5]. That comparison indicated the need for methods employing more realistic models which depart from the standard homogeneous and isotropic film assumption.

Earlier reports on electron microscopy [6], computer simulation [7], humidity adsorption [8] and light scattering [9] had already given evidence for porous film microstructures, different from those found in bulk materials, which agree with our hypothesis.

1. INTRODUCTION

Improvement of optical components and systems based on the technology of thin films has imposed an increasing demand for accurate knowledge of their optical constants. High reflectance all-dielectric mirrors, wide-band dichroic beam-splitters and narrow-band interference filters, for example, show optical performances which are strongly dependent on the individual behavior of the layers they are composed of [1].

Advances in vacuum instrumentation, deposition parameters control and design techniques for interference filters have altered the way to high quality and tight tolerance in film-based optical components. The main challenge has shifted to the investigation of the material properties of coatings [2].
2. THEORETICAL BACKGROUND

2.1 Prism-Coupler Waveguide Method

Consider a planar waveguide surrounded by two parallel interfaces labelled as a and b, as shown in Fig. 1. For an isotropic waveguide with index of refraction \( n \), the well known [11] mode equation applies:

\[
\frac{2\pi}{\lambda} \beta \Gamma = \theta_a + \theta_b + m \pi, \tag{1}
\]

where \( \beta \Gamma \) is the propagation parameter defined by:

\[
\beta \Gamma = [n^2 - \alpha^2]^\frac{1}{2}, \tag{2}
\]

with \( \alpha = \omega c / \gamma \), while for

\( s \)-polarization (TE):

\[
\tan(\theta_i) \equiv \frac{\beta_i}{n_s} \tag{3}
\]

\[
n_s = [n^2 - \alpha^2]^\frac{1}{2} \tag{4}
\]

\[
\beta_i \equiv [\alpha^2 - n_i^2]^\frac{1}{2} \tag{5}
\]

\( i = a, b. \)

When the waveguide material is anisotropic, provided one of its principal axes of symmetry is orthogonal to the plane of incidence (i.e., in the \( s \)-polarization direction) equation (1) remains valid. This has been rigorously shown through a 4 x 4 matrix formalism for both uniaxial [12] and biaxial [13] cases. Under such condition, the \( s \)- and \( p \)-polarization modes are uncoupled, and thus can be treated as propagating independently in the waveguide. Expressions above can still be used, except for (2) and (4) which are replaced by [13]:

\[
\beta \Gamma \equiv [n_i^2 - \alpha^2]^\frac{1}{2},
\]

\[
\beta_p \equiv \frac{n_{11} n_3 [n_2^2 \sin^2(\phi) + n_3^2 \cos^2(\phi) - \alpha^2]^\frac{1}{2}}{n_1^2 \sin^2(\phi) + n_3^2 \cos^2(\phi)} \tag{6}
\]

and

\[
n_s \equiv [n_i^2 - \alpha^2]^\frac{1}{2},
\]

\[
n_p \equiv \frac{n_{11} n_3}{n_1^2} \frac{[n_2^2 \sin^2(\phi) + n_3^2 \cos^2(\phi) - \alpha^2]^\frac{1}{2}}{n_1^2 \sin^2(\phi) + n_3^2 \cos^2(\phi)} \tag{7}
\]

where conventions used for the principal refractive indices are according to Fig. 1.

2.2 Envelope Method

Consider a system which consists of a thin film on a transparent substrate surrounded by air as shown in Fig. 2.
Incoherent summation over the multiple reflections that occur at both substrate interfaces leads to the overall transmissivity at normal incidence:

\[ T = \frac{T_I T_s}{1 - R_I R_s} \quad (8) \]

where \( R_I \) and \( T_s \) refer to the substrate-air interface, while \( T_I \) and \( R_s \) refer to the air-film-substrate subsystem. These are determined by use of a coherent treatment and Fresnel relations, so that in the weak absorption regime (8) becomes:

\[ T \approx \frac{A'_1 x}{A'_2 - A'_3 x \cos (2\gamma) + A'_4 x^2}, \quad x \equiv e^{-2\gamma} \quad (9) \]

as in Swanepoel [14], where phase thickness \( \gamma = 2\pi nd/\lambda \), absorption coefficient \( \xi = 2\pi kd/\lambda \), and \( A' \) coefficients are explicit functions of the real part of the refractive indices of the media.

For a high index film with \( n > n_s \), for example, \( T_{\text{max}} \) or \( T_{\text{min}} \) envelope curves are obtained from equation (9) with \( \cos (2\gamma) \) equal to +1 or -1, respectively. Those values correspond to the following values of \( \lambda \):

\[ \lambda = \frac{4 \pi nd}{2m} \quad \text{for} \quad T = T_{\text{max}} \quad (10) \]

or

\[ \lambda = \frac{4 \pi nd}{2m + 1} \quad \text{for} \quad T = T_{\text{min}}. \quad (11) \]

Through a combination of \( T_{\text{max}} \) and \( T_{\text{min}} \) so that \( x \) is eliminated, the real part of the film refractive index is given by

\[ n = \left[ N \pm \left( N^2 - n_s^2 n_a^2 \right)^{\frac{1}{2}} \right]^{\frac{1}{2}}, \quad (12) \]

with

\[ N \equiv 2 n_0 n_s \left[ \frac{1}{T_{\text{min}}} - \frac{1}{T_{\text{max}}} \right] + n_s^2 + n_a^2/2 \]

If the value of \( n \) is known, \( d \) is determined by using at least any two identities (10) or (11), so that \( m \) is eliminated. From \( n \) and \( d \), \( m \) is immediately found. Furthermore \( n \) and \( d \) results can be iteratively refined through the Swanepoel [14] procedure. The interference order \( m \) is an integer, the closest to that value calculated previously.

In the anisotropic case, provided the incident polarization is taken as along or orthogonal to one of the principal axes of symmetry, those two components will traverse the film uncoupled, and therefore can be treated independently as it was described. Referring to Fig. 1 and identity (12), the effective refractive indices obtained by this method are related to the principal refractive indices, as in (7) with \( \sigma = 0 \), through the relations:

\[ n_{||} = n_0 \]

\[ n_{\perp} = \frac{n_1 n_3}{[n_1^2 \sin^2 (\phi) + n_3^2 \cos^2 (\phi)]^{\frac{1}{2}}} \quad (14) \]

corresponding to an incident polarization parallel and perpendicular to the second principal axis of symmetry, respectively.

3. EXPERIMENTAL PROCEDURE

Films for this work were thermally deposited on clean transparent glass in a vacuum chamber with ground-pressures of 10E-6 mbar. Resistive heating of a W boat was employed for the PbF\(_2\) films, while the CeO\(_2\) films were reactively deposited from Mo boats in an oxygen atmosphere at typical working pressures of 10E-4 mbar. Deposition geometry was carefully controlled in each run, with particular attention to the vapor angle of incidence.

In the waveguide experiment, the main Otto [15] configuration was used, as shown in Fig. 3. However the prism-coupler was a hemisphere made of Schott SF-11 optical glass with refractive index \( n_n = 1.77862 \) at the He-Ne laser wavelength. It was supported by a \( \theta - \phi \) positioner over an X-Y translator. The Zeiss-Jena goniometer could provide readings better than 0.5 minute of arc.

![Figure 3: Experimental configuration with hemispherical coupler.](image)
4. RESULTS AND DISCUSSION

Data from a PbF$_2$ film, deposited at normal vapor incidence and measured by the waveguide technique, are shown in Fig. 4.

![Figure 4: Thickness versus refractive index for a PbF$_2$ film as seen by the waveguide method. Two TE-TM pairs refer to left and right sides of the hemispheric prism-coupler.](image)

The very small spread of values for the refractive index and thickness, from $TE$ and $TM$ modes, left and right measurements as well, enforces the experimental procedure and indicates that the isotropic model presents reasonably good performance here.

In order to test possible discrepancies due to the orientation of the sample, another set of measurements was made after it was rotated by an azimuthal angle of 90°. Good agreement was reached for refractive index results, although thickness values differed slightly more than the measurement uncertainty [13]. This is probably due to thickness nonuniformity and a small shift in position as the sample was rotated.

In order to obtain more pronounced anisotropic behavior, films of CeO$_2$, with high bulk index, were deposited at oblique incidence. A vapor angle of 58.3° by the tangent rule [16] implied in a columnar angle of 39.0°, both with respect to the surface normal.

Even starting from an isotropic model, very significant refractive index differences $\Delta n$ between $TE$ and $TM$ modes resulted. Right (R) and Left (L) typical data are present in Fig. 5, as well as mean standard deviations $\sigma$ representing the remaining uncertainties after a large number of measurements. Relative values are $(\Delta n/\sigma)_R \geq 20$ and $(\Delta n/\sigma)_L \geq 56$.

Thickness values are not in satisfactory agreement even when measurement imprecisions are considered.

![Figure 5: Thickness versus refractive index for a PbF$_2$ film as seen by the waveguide method.](image)

That whole picture illustrates the need for models which take explicitly into account the anisotropic microstructure. Moreover careful analysis of the theory shows that an "equivalent isotropic index" can be taken for the $TE$ polarization along one of the principal axes of symmetry, but that a similar association is incorrect for the $TM$-polarization [13].

Next step was to consider a uniaxial model by assuming $n_1 = n_2$, whose results are also shown in Fig. 5. Now the birefringence is even more pronounced with $(\Delta n)_R = 0.098 \pm 0.007$ and $(\Delta n)_L = 0.093 \pm 0.004$, while all thickness data now agree within the experimental uncertainties.

For the more general and accurate biaxial columnar model, since the CeO$_2$ film would guide only two $TM$-modes, an additional measurement by the envelope method was performed. Here the $TE$-polarization is still along one of the principal axes of symmetry so that $n_{TE} = n_2$.

Consistency between the two experiments was enforced by a comparison of $TE$ measurements, which lead to good agreement between $n_2^{EN} = 1.78 \pm 0.03$ (envelope method) and $n_2^{WG} = 1.794 \pm 0.002$ (waveguide method) at 632.8 nm.

Besides reaching thickness results even closer to each other than those obtained by the uniaxial model, the biaxial provided the following $TM$-related principal refractive indices: $n_1 = 1.65 \pm 0.05$ and $n_3 = 1.97 \pm 0.06$. Here expressions (1), (6) and (7) where used for the waveguide method, and (14) for the envelope method. Detailed description of the matching between those two treatments can be found in Ref. [13]. The values obtained for $n_1$ and $n_3$, compared to that of $n_2$, are consistent with the relative molecular filling factors along different directions, as expected from self-shadowing considerations.
5. CONCLUSION

Starting from ideal isotropic behavior, the waveguide and the envelope methods were extended to general uniaxial and biaxial structures, provided a proper sample orientation is chosen to assure uncoupled propagation of linearly polarized modes. That requirement can be met in practically all experiments with monolayer films, and retains most simplicity of the isotropic treatments.

It was shown that the isotropic model still performs reasonably well for moderate-index PbF₂ films deposited at normal vapor incidence. For high-index, obliquely deposited CeO₂ films, however, anisotropic models of microstructure are required. Uniaxial and biaxial cases considered, TE and TM polarization results were consistent within the waveguide experiment, as well as with those from the envelope method.

Such consistency indicates that both methods are appropriate for anisotropic films when the columnar model is explicitly used in their interpretation.

6. ACKNOWLEDGMENTS

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7. REFERENCES


Figure Captions

Fig. 1 Representation of the columnar model for an anisotropic film with principal indices shown along the axes of symmetry [10].

Fig. 2 Thin film deposited on a transparent substrate surrounded by air, with conventions shown for the envelope method calculations.

Fig. 3 Configuration of the waveguide experiment with hemispherical coupler.

Fig. 4 Thickness versus refractive index for a normally deposited PbF₂ film as seen by the waveguide method. Two TE - TM pairs refer to left and right sides of the hemispheric prism-coupler.

Fig. 5 Thickness versus refractive index for an obliquely deposited CeO₂ film as seen by isotropic (O) and uniaxial (O) models. TE (O) results from both cases are coincident.