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Characterization of thin films based on reflectance and transmittance measurements at oblique angles of incidence

Antti Lamminpää, Saulius Nevas, Farshid Manoocheri, and Erkki Ikonen

The optical parameters of a SiO_2 thin-film coating determined from the spectral reflectance and transmittance measurements at various incidence angles, including the normal incidence and the Brewster's angle, are compared in this paper. The high-accuracy measurements were carried out through visible–near-infrared spectral regions by using our purpose-built instruments. The optical parameters obtained from the reflectance and the transmittance data are consistent over the angles of incidence and agree within 0.2%. The effect of important systematic factors in the oblique-incidence spectrophotometric measurements is also discussed. © 2006 Optical Society of America

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1. Introduction

In the optical metrology of thin-film coatings, characterization techniques based on least-squares fitting to the measured reflectance or transmittance spectra are widely used.[1–3] The fitted spectral characteristics usually consist of normal-incidence transmittance measurement results. However, the analysis may also include spectrophotometric measurement data collected at oblique angles of incidence. Moreover, the reflectance measurements are almost always made at oblique incidence. Thus to compare the reflectance and the transmittance measurement results it may be necessary to include the transmittance data at the oblique incidence. Furthermore, the oblique-incidence data can increase the sensitivity of the characterization methods.[1]

An important condition for the characterization procedures to yield reliable results is the accuracy of the spectrophotometric measurements[4,5] to be performed with well-collimated beams, at precisely determined incidence angles, and with properly aligned polarization planes of the beam.[6] At the Helsinki University of Technology (TKK), we perform spectrophotometric measurements that satisfy such requirements.[7–9] Our purpose-built high-accuracy reference spectrophotometer features a collimated beam in a single-beam measurement configuration. In our previous report,[6] we studied the effect of important systematic factors in oblique-incidence measurements and presented preliminary experimental results for a SiO_2 thin-film sample based on spectral transmittance measurements at several angles of incidence over the spectral range from 400 to 950 nm. The derived optical parameters of the coating were consistent over the incidence angles from 0 to 56.4 degrees and demonstrated a high accuracy of the measurements.

In this contribution, we present experimental results for the SiO_2 thin-film sample based on spectral reflectance data at 10, 30, and 56.4 degree angles of incidence. The oblique-incidence transmittance measurements were also extended to cover the spectral range 950–1600 nm. The optical parameters of the SiO_2 coating derived from the reflectance results are compared with those determined from the normal- and oblique-incidence transmittance data. The results of the characterization are consistent and agree within 0.2%. The consistency among the determined optical parameters of the thin-film layer confirms the accuracy of the spectrophotometric measurements. The effect of the systematic factors in the measurements is also discussed.

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2. Spectrophotometric Characterization of Optical Thin-Film Coatings

In the analysis of thin-film samples, we use a mathematical model that considers a thin homogeneous layer of a dielectric material deposited on a macroscopically thick plane-parallel substrate.[5] The spectral transmittance and reflectance characteristics of such a sample are derived as a result of sums of geometric series of the coherent interreflections in the thin-film layer and noncoherent interreflections in the substrate slab. During the analysis, published refractive-index values were used for the fused-quartz substrate.[10,11] In the model, the complex index of refraction of the layer is described by the use of appropriate dispersion equations. We used the Cauchy formula to describe the refractive index $n(\lambda)$ of the SiO_2 thin-film layer,

$$n(\lambda) = x_1 + \frac{x_2}{\lambda^2} + \frac{x_3}{\lambda^4}, \tag{1}$$

where x_1, x_2, and x_3 are freely selectable constants.[5] Equation

$$k(\lambda) = x_4 \exp(-x_5\lambda), \tag{2}$$

where x_4 and x_5 are also adjustable constants, could be used for the description of the extinction coefficient $k(\lambda)$ of the material.

The optical parameters of thin-film coatings are obtained as a result of least-squares fitting to the measured reflectance or transmittance spectra. The constants of the dispersion equations, $x_1, \ldots x_5$, and the film thickness are adjusted during the minimization of the discrepancy function expressing the difference between the modeled and the measured values.

It is obvious that the accuracy of the characterization procedures based on curve fitting to the spectrophotometric data is highly related to the quality of the measurements. The uncertainty in the determined optical parameters of a thin-film layer is a result of various uncertainty components in the measurements, especially of those having systematic origin.[4,5] Furthermore, there are additional systematic factors that require careful consideration when the spectrophotometric measurements are performed at oblique angles of incidence, such as misalignment between the polarization and the incidence planes, angle determination error, and quality of the beam collimation.[6] However, it may be difficult to have these factors under control if ordinary commercially available spectrophotometers are used.

3. Experimental Procedures and Results

The measurements of spectral regular reflectance were made for the SiO_2 coating deposited on a fused-quartz substrate, the same sample as in Ref. 6. The SiO_2 coating was produced with the ion-beam sputtering process, Balzer BAK 640 plant, where a Kaufman-type ion source with Xe ions and the reactive atmosphere of O_2 were used. The base pressure was 10^{-6} mbar, and the deposition pressure was between 1×10^{-4} and 2×10^{-4} mbar. Measurements were performed at 10, 30, and 56.4 degree incidence angles defined within ± 0.1 degree over the wavelength range from 400 to 1480 nm. The upper wavelength limit in the reflectance measurements is defined by the current set of gratings in the monochromator. The incident angles correspond to those at which the oblique-incidence transmittances were earlier measured over the wavelengths from 400 to 950 nm.[6] Now the transmittances in the spectral region from 950 to 1600 nm were also measured.

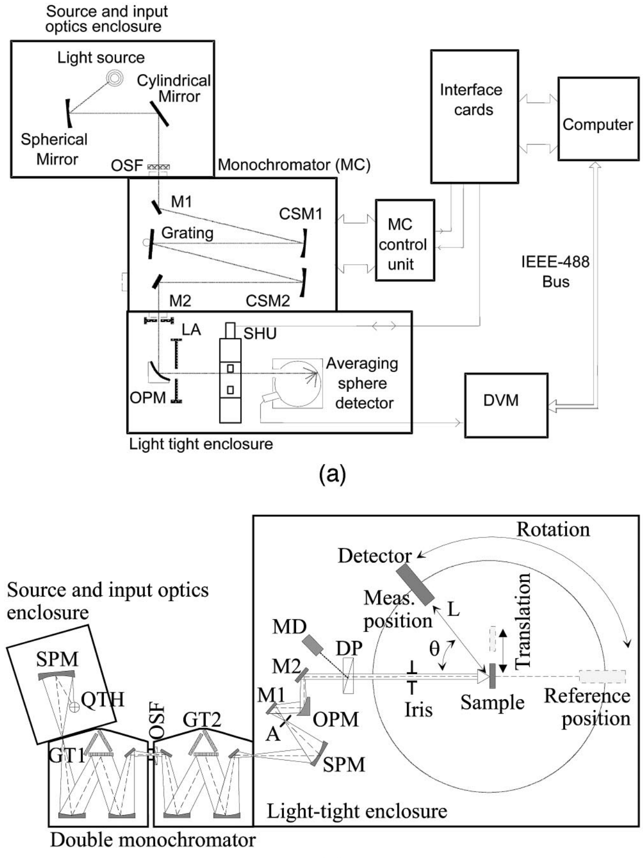

Fig. 1. (a) Schematic of the reference spectrophotometer setup: OSF, order-sorting filter; M1, M2, flat mirrors; CSM1, CSM2, collimating spherical mirrors; LA, limiting aperture; OPM, off-axis parabolic mirror; SHU, sample holder unit; DVM, digital voltage meter. (b) Schematic of the gonioreflectometer setup: QTH, quartz-tungsten-halogen lamp; SPM, spherical mirror; GT1, GT2, grating turrets; OSF, order-sorting filter; M1, M2, flat mirrors; A, aperture; OPM, off-axis parabolic mirror; DP, polarizer; MD, monitor detector.

These and the earlier transmittance measurements were carried out with the reference spectrophotometer [Fig. 1(a)] built at TKK.[9] The instrument features a well-collimated beam in a single-beam measurement configuration. It consists of a wavelength-selective light source system built around a single grating monochromator with reflecting input and output optics and a versatile detection system. A Glan–Taylor

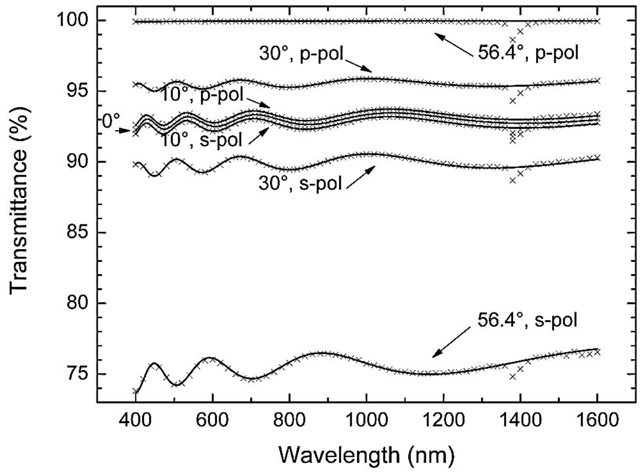

Fig. 2. Transmittance spectra of the SiO_2 thin-film coating on a fused-quartz substrate for 0, 10, 30, and 56.4 degree incidence angles. Solid lines and crosses represent modeled and measured spectral transmittance values, respectively. The measurements were carried out for both *s* and *p* polarization of the spectrophotometer beam.

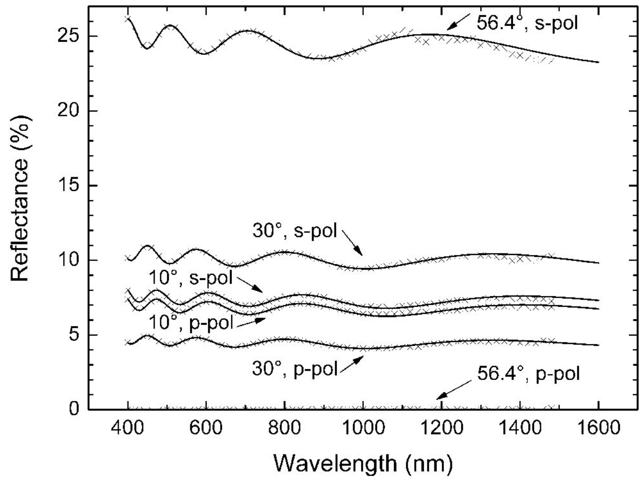

Fig. 3. Reflectance spectra of the SiO_2 thin-film coating on a fused-quartz substrate for 10, 30, and 56.4 degree incidence angles. Solid lines and crosses represent modeled and measured spectral reflectance values, respectively. The measurements were carried out for both *s* and *p* polarization of the reflectometer beam.

polarizing prism is employed for achieving the linear polarization states of the beam. A linear translator is used for alternately positioning the sample and the block in the path of the beam that is directed toward an averaging sphere detector unit with a possibility of mounting either silicon or InGaAs photodiodes. The silicon photodiode was used in the earlier measurements throughout the wavelength range from 400 to 950 nm. In the current measurements for the 950–1600 nm spectral range, the InGaAs photodiode was utilized. The relative standard uncertainty of the normal-incidence transmittance measurements ranges from 0.05% to 0.2%.

The reflectance measurements were performed with an absolute gonioreflectometer [Fig. 1(b)] that has been recently built at TKK.[12] The instrument has a double monochromator-based source system providing a collimated beam as well. The polarization state of the beam is defined by the Glan–Taylor polarizing prism. The linearly polarized beam is incident on the test sample mounted in a sample holder that consists of a rotary turntable and a linear translator. The intensity of the incident and the reflected beams is measured alternately by a detector mounted on another turntable that has a common axis of rotation with that of the sample holder unit. Silicon and InGaAs photodiodes were used as the detector for the measurements in the wavelength ranges from 400 to 900 nm and from 900 to 1480 nm, respectively. The estimated standard uncertainty in the reflectance measurements for the wavelengths from 400 to 900 nm is 0.2% and varies between 0.5% and 1% at the longer wavelengths.

With the measurements completed, the optical parameters of the SiO_2 layer were determined from the measured transmittance spectra separately for each angle of incidence by fitting simultaneously the results obtained with *s* and *p*-polarization over the 400–1600 nm wavelength range. The same analysis procedure was repeated for the reflectance measurement data. The measured and modeled transmittance and reflectance spectra are plotted in Figs. 2 and 3, respectively. Because of the water absorption, the wavelength region around 1400 nm was excluded from the analysis. The determined refractive indices are shown in Fig. 4 for each set of the reflectance and the transmittance data. The analysis did not reveal any reasonable absorption in the SiO_2 layer.

4. Comparison of the Results Derived from the Oblique-Incidence Spectrophotometric Measurements

When the refractive-index curves plotted in Fig. 4 are compared, it is obvious that there is a very good agreement among the results yielded by the reflectance data

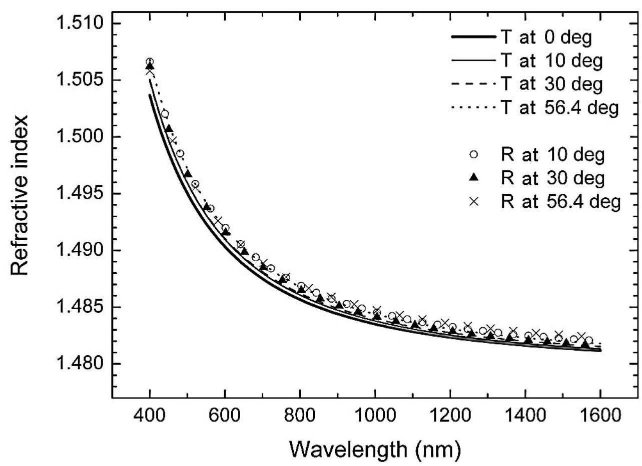

Fig. 4. Refractive indices of the SiO_2 layer determined from the spectral transmittance and reflectance measurements at the oblique-incidence angles. The refractive indices yielded by the oblique-incidence reflectance and transmittance measurements agree within 0.2%.

Table 1. Comparison of the Determined SiO_2 Layer Thicknesses

	Determined Layer Thickness			Difference when Compared to the Normal-Incidence Transmittance Result		
Measurement	Physical	Optical (550 nm)	Optical (1000 nm)	Physical	Optical (550 nm)	Optical (1000 nm)
Transmittance at 0°	715.9	1068.4	1062.1	–	–	–
Transmittance at 10°	715.9	1068.9	1062.2	0.00%	0.05%	0.02%
Transmittance at 30°	716.1	1069.2	1062.6	0.02%	0.07%	0.05%
Transmittance at 56.4°	715.0	1068.4	1061.4	−0.12%	0.00%	−0.06%
Reflectance at 10°	716.2	1070.2	1063.3	0.05%	0.17%	0.12%
Reflectance at 30°	717.0	1071.1	1064.2	0.16%	0.25%	0.20%
Reflectance at 56.4°	716.9	1071.1	1064.5	0.14%	0.25%	0.23%

at all the angles of incidence. The refractive indices determined from the reflectance measurements at 10° to 56.4° angles of incidence agree better than 0.1%, and thus the curves in the figure closely overlap. The determined indices also agree well with those from the spectral transmittance measurements over the incidence angles from 0° to 56.4°. The consistency among the refractive indices obtained from transmittance measurements at the different angles of incidence is within 0.2%. As illustrated in Fig. 4, all the obtained refractive indices fall between the values yielded from normal-incidence and Brewster's angle measurements. We conclude that the consistency among the results for the different incidence angles supports the consistency in setting the angle of incidence, the aligning of the plane of polarization with respect to the plane of incidence, and also a good collimation of the beam.[6] If not considered carefully, these factors could easily cause significant systematic errors.

The physical and optical thicknesses of the layer derived from the oblique-incidence spectrophotometric data are compared in Table 1. As in the case of the refractive indices, both physical and optical thicknesses yielded by the reflectance data agree within 0.1%. Comparison of the reflectance and the transmittance results also shows a good agreement among the determined physical thicknesses. An interesting feature is that the agreement for the optical thicknesses is somewhat worse than that in the case of the physical parameter comparison. Typically, the calculated optical thickness of a coating is affected by the measurement errors less than the physical thickness and the refractive index alone, as the effect on these two parameters tends to be of the opposite sign: if one of them is overestimated then the other one is underestimated and vice versa.[4,5] However, considering that the reflectance and the transmittance measurements were carried out with two different instruments having different wavelength calibrations, this could be possibly explained by a small discrepancy between the wavelength scales of the instruments. A shift in the measurement wavelengths causes an error in the determined layer thickness and a small change in the refractive index at shorter wavelengths.[5]

5. Conclusions

We have measured with high accuracy the spectral regular transmittance and reflectance of a SiO_2 thin-film sample over the visible–near-infrared wavelength regions, 400–1600 nm, with collimated linearly polarized beams incident on the sample at incidence angles ranging from normal incidence to the Brewster's angle. The optical parameters of the layer determined from each of the oblique-incidence reflectance spectra were compared with those derived from the transmittance measurements. The intercomparison showed a good agreement among the optical parameters derived from the reflectance data at the different angles of incidence and among those from the normal- and oblique-incidence transmittance characterization. The agreement among the results is within 0.2%. Thus the good agreement among the determined optical parameters supports the accuracy of the measurements with the two instruments and the consistency in determining the incidence angles, the alignment of the plane of polarization with respect to the plane of incidence, and also a good collimation of the measurement beams. These factors must be considered carefully in the oblique-incidence spectrophotometric measurements, especially when ordinary commercial spectrophotometers with converging beam geometries are used. Otherwise, they could cause significant systematic errors in the determined optical parameters of the layer.

An interesting finding was that the optical thicknesses determined from the reflectance and the transmittance data were in a slightly worse agreement than the derived physical thicknesses and the refractive indices of the layer, if considered separately. However, taking into account that two different instruments were utilized in the reflectance and the transmittance measurements, this could be possibly attributed to a small discrepancy between the wavelength calibrations of these instruments.

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