Oscillations in spectral behavior of total losses \((1 – R – T)\) in thin dielectric films

Tatiana V. Amotchkina, Michael K. Trubetskov, Alexander V. Tikhonravov, Vesna Janicki, Jordi Sancho-Parramon, Olga Razskazovskaya, and Vladimir Pervak

Abstract: We explain reasons of oscillations frequently observed in total losses spectra \((1 – R – T)\) calculated on the basis of measurement spectral photometric data of thin film samples. The first reason of oscillations is related to difference in angles of incidence at which spectral transmittance and reflectance are measured. The second reason is an absorption in a thin film. The third reason is a slight thickness non-uniformity of the film. We observe a good agreement between theoretical models and corresponding measurements, which proves above statements on the origins of oscillations in total losses.

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References and links
1. Introduction

Design and production of high quality multilayer optical coatings require knowing optical constants of thin film materials with a good accuracy. Therefore it is not surprising that hundreds of papers devoted to optical characterization of thin films have been published so far. Characterization problem is still at the center of interest of the optical coatings community. The evidence of this fact is that at the last three Optical Interference Coatings (OIC) Topical Meetings in 2004, 2007, and 2010, special measurement problems related to this subject were considered [1–3].

In Ref. [4] we introduced a concept of characterization approach as a combination of the measurement data, thin film model, and algorithm for data processing. The most popular characterization approach is based on using normal and quasi-normal transmittance ($T$) and reflectance ($R$) spectra of an investigated sample. This approach is simple from an experimental point of view. Normal incidence transmittance can be measured with a sufficient accuracy on any UV-visible-IR spectrophotometer without the need for a polarizer or special accessory and quasi-normal reflectance can be also measured on most of spectrophotometers with a nonpolarizing accessory.

Knowing measurement data accuracy as well as possibilities and limitations of well-established measurement techniques are very important for characterization processes because they essentially impact characterization results [5, 6]. The errors in spectral photometric data can be divided into random and systematic ones [6]. Random errors (random noise) vary from one point to another in the measurement data set. It was shown in [5] that these errors almost do not influence characterization results. Systematic errors are those that result in an offset of spectral characteristics as a whole or cause large-scale wavelength variations of $T$ and $R$ curves. An example of systematic errors is a drift of spectral transmittance (reflectance) in time. Offsets and drifts can be caused by calibration drifts, incidence angle variations, monochromator offsets, finite optical bandwidth, detector nonlinearity, etc. Systematic errors are especially critical for the determination of thin film parameters [5].

Useful information about measurement data accuracy is provided by examining the spectral dependence of $TL(\lambda) = 100\% - R(\lambda) - T(\lambda)$ calculated from measurement normal or quasi-normal reflectance and transmittance data. This dependence is frequently called total losses ($TL$) in the investigated thin film sample [4, 7]. Typically, in the spectral range where the substrate and the thin film are non-absorbing and non-scattering, one expects zero total losses in the sample. In reality, systematic errors can be comprised in measurement data sets. In the simplest cases of presence of systematic offsets in $R$ or $T$ spectra, $TL(\lambda)$ deviate from zero by some constant.
Total losses spectral behavior gives also a priori information about presence of absorption in thin films. In the spectral range where investigated thin film is absorbing, one usually expects that $\text{TL} (\lambda)$ is monotonically decreasing with growing $\lambda$.

Frequently, analyzing total losses spectra in the range of interest, researchers observe oscillations. Presence of these oscillations may cause doubts about the quality of measurement data. Some researchers repeat measurements and observe that oscillations still remain in remeasured data sets. As a result, these oscillations are attributed either to small defects in spectral photometric measurements and ignored, or all measurement data are considered as unacceptable and rejected. Up to now, no comprehensive study on the origins of oscillations in total losses has been performed.

In the present paper we provide a special study aimed at revealing origins of oscillations in total losses spectra. We suppose three possible reasons of oscillations in $\text{TL}$. These three reasons are of completely different nature. The first reason of oscillations in $\text{TL}$ is related to difference in angles of incidence (AOI) at which $T$ and $R$ are measured. The second reason is an absorption in a thin film acting in combination with interference effects and therefore leading to oscillations in $\text{TL}$. The third reason can be attributed to a slight thickness non-uniformity of the film. In order to support our suppositions we obtain corresponding theoretical formulas and compare them with results of experiments. From a practical point of view the obtained results allow to check the measurement data accuracy, verify measurement arrangement and to select a proper characterization model at a very early stage of thin film characterization.

In Section 2, we provide theoretical study related to three possible reasons of oscillations in $\text{TL}$. Namely, we obtain approximate relations for $\text{TL}$ assuming that 1) AOI for separate $T$ and $R$ measurements are different; 2) film is slightly absorbing; and 3) there is a slight thickness non-uniformity. These approximate relations demonstrate qualitatively that oscillations can be caused by all reasons mentioned above. In Section 3, we describe experimental samples specially produced for our study, and $T$ and $R$ data sets measured using two different Perkin Elmer spectrophotometers. In Section 4, in order to validate our theoretical results, we compare theoretical and experimental data and discuss the correspondence between these data. In Section 5, we present more evidences of our theoretical results with the help of measurements performed with Cary spectrophotometers. Our final conclusions are presented in Section 6.

2. Reasons of oscillations: theoretical study

2.1. Influence of AOI on total losses spectral behavior

Theoretically, in order to calculate total losses $\text{TL} (\lambda)$, transmittance and reflectance values should be specified for the same values of AOI. Practically, it is not always possible, because in some spectrophotometers transmittance and reflectance of a sample are measured at slightly different AOI, $\theta_1$ and $\theta_2$, respectively. Typically, transmittance is measured at normal incidence ($\theta_1 = 0$) and reflectance is measured at a quasi-zero AOI ($\theta_2 = 5 - 10^\circ$). Nevertheless, it is required to estimate total losses on the basis of available $T$ and $R$ spectra. The most natural way to estimate total losses is to neglect difference between $\theta_1$ and $\theta_2$ and calculate $\text{TL}$ as follows:

$$\text{TL}(\lambda_j) = 100\% - T(\theta_1, \lambda_j) - R(\theta_2, \lambda_j), \quad j = 1, \ldots, L,$$

where $\{\lambda_j\}$ is the wavelength grid in the measurement spectral range. In Eq. (1) and in Sections 2-4 we assume that the light is non-polarized.

We suppose that difference in quasi-normal AOI may cause oscillations in total losses $\text{TL}(\lambda)$. In order to prove this, we expand $T(\theta_1)$ and $R(\theta_2)$ in the vicinities of $\theta_1 = 0$ and $\theta_2 = 0$ into
Taylor’s series. As a result, we obtain an approximate relation $T_{\text{AOI}}$ for $T_L$ defined by Eq. (1):

$$T_{\text{AOI}}(\lambda) = \frac{4\pi d n_s}{\lambda} \frac{\sin 2\varphi}{n} \left( \frac{n^2 - 1}{(1 + n_s)^2 \cos^2 \varphi + (n_s/n + n)^2 \sin^2 \varphi} \right) \left( \theta_2^2 - \theta_1^2 \right),$$

(2)

where $\varphi = (2\pi/\lambda) n d$, $n$ and $n_s$ are refractive indices of the film and the substrate, respectively, $d$ is the film geometrical thickness. For the sake of convenience, in Eq. (2) and in what follows, indication of wavelength dependencies of $n(\cdot), n_s(\cdot), \varphi(\cdot)$ are omitted.

It should be noted here, that the approximate relation (2) is obtained without taking into account the substrate back side. It is evident from a physical point of view that effect of the substrate back side reveals itself very weakly in $T_L$ calculations. The reason is that when back side is taken into account, reflectance is increased and transmittance is decreased by almost the same value, and in total losses these two variations compensate each other. The numerical confirmation of this fact is presented below.

In order to verify the accuracy of approximate relation (2), we compare the values $T_{\text{AOI}}$ and exact values of $T_L$ defined by Eq. (1). We calculate both functions for a model sample. Refractive index wavelength dependencies of the model film and model substrate are described by the Cauchy formula:

$$n(\lambda) = A_0 + A_1 (\lambda_0/\lambda)^2 + A_2 (\lambda_0/\lambda)^4,$$

(3)

where $A_0, A_1$ and $A_2$ are dimensionless parameters, $\lambda_0 = 1000$ nm, wavelength $\lambda$ is specified in nanometers. For $n(\lambda)$, we specify $A_0 = 2.18031, A_1 = 0.00586, A_2 = 0.00676$. For $n_s(\lambda)$, we take $A_0 = 1.50399, A_1 = 0.0049, A_2 = -0.00005$. We specify model film thickness $d = 500$ nm, AOI $\theta_1 = 0, \theta_2 = 7^\circ$. In Fig. 1 we compare spectral dependencies of $T_L$ and $T_{\text{AOI}}$. We calculate $T_L$ for two cases: when the substrate back side is taken into account and when it is not taken into account. In Fig. 1 we observe that the total losses calculated for both cases are almost undistinguishable. This figure demonstrate that $T_{\text{AOI}}(\lambda)$ approximates exact $T_L(\lambda)$ with a high accuracy.

In Fig. 1 oscillations are clearly seen. These oscillations are caused by trigonometric terms $\sin 2\varphi, \cos \varphi, \sin \varphi$ in Eq. (2). The magnitudes of oscillations are proportional to film thickness $d$: in thicker films oscillations in $T_L$ are higher. It is seen also that magnitudes of oscillations are dependent on the ratio $n_s/n$. Refractive indices of the substrates, typically used in visible

![Fig. 1. Comparison of theoretical $T_L(\lambda)$ Eq. (1) in a model sample with back side contribution, without back side contribution and approximate dependence $T_{\text{AOI}}(\lambda)$ calculated by Eq. (2). AOI $\theta_1 = 0, \theta_2 = 7^\circ$.](image-url)
spectral range, are 1.45–1.65. When \( n_s/n \) is close to 1, the numerator in Eq. (2) approaches to zero, \( T_{LAO} \) tends to zero, and, therefore, oscillations are vanished. Situations, when \( n_s/n \approx 1 \), correspond to films with low refractive index values, for example, SiO\(_2\) films. Situations, when \( n_s/n \) differs from one essentially, correspond to films with high and medium refractive indices, such as TiO\(_2\), Ta\(_2\)O\(_5\), Al\(_2\)O\(_3\), Nb\(_2\)O\(_5\), HfO\(_2\). In the case of the considered model sample, the ratio \( n_s/n \) is about 0.7, and oscillations in \( TL \) are quite high.

In this subsection, we demonstrate theoretically that oscillations in \( TL \) can be attributed to difference in AOI used for separate \( T \) and \( R \) measurements. The oscillations reveal themselves stronger for films with high refractive indices and for thicker films. In the case of film with refractive index \( n \) close to \( n_s \), the oscillations are negligible for any values of film thickness \( d \). From a practical point of view, oscillations in \( TL \) originated from difference in AOI, do not necessarily indicate poor quality of experimental data.

### 2.2. Influence of thin film absorption on total losses spectral behavior

In the cases of slightly absorbing thin films, we suppose that oscillations in total losses can be caused by absorption. Assume that the film extinction coefficient is \( \chi(\lambda) \) and consider total losses in a slightly absorbing film:

\[
TL(\chi, \lambda) = 100\% - T(\chi, \lambda) - R(\chi, \lambda).
\] (4)

We find the first term of the Taylor’s series for \( TL(\chi, \lambda) \) in the vicinity of \( \chi = 0 \). In the case of normal incidence, we obtain an approximate relation \( TL_{Abs} \) for \( TL(\chi, \lambda) \):

\[
TL_{Abs} = \frac{4\pi d \chi}{\lambda} \left\{ n_s (n_s + 1) \left( \frac{n_s}{n} + 1 \right) + \frac{(n_s^2 - 1) \cos^2 \varphi + [n^2 - (n_s/n)^2] \sin^2 \varphi}{(1 + n_s^2 \cos^2 \varphi + (n_s/n + n)^2 \sin^2 \varphi)^2} \right\}.
\] (5)

In order to verify the accuracy of approximate relation (5), we compare the values of \( TL_{Abs} \) and exact values of \( TL(\chi) \) defined by Eq. (4). We calculate the latter values for the case when the substrate back side is taken into account and when the back side is not taken into account. It is seen from Fig. 2 that \( TL(\chi) \) calculated for these two cases are almost undistinguishable. We provide calculations of \( TL(\chi) \) and \( TL_{Abs} \) values for a model sample. Refractive indices \( n \) and \( n_s \) as well as film thickness \( d \) are the same as in Subsection 2.1. Extinction coefficient of
the model film is described by the exponential model:

\[ \chi(\lambda) = B_0 \exp \{ B_1 \lambda_0 / \lambda + B_2 \lambda / \lambda_0 \}, \]

where \( B_0, B_1 \) and \( B_2 \) are dimensionless parameters, \( \lambda_0 = 1000 \) nm, wavelength \( \lambda \) is specified in nanometers. For \( \chi(\lambda) \), we specify \( B_0 = 66686.77, B_1 = 0, B_2 = -42.55 \). In Fig. 2 we compare spectral dependencies of \( TL(\chi, \lambda) \) and \( TL_{\text{Abs}}(\lambda) \) in the spectral range from 380 to 500 nm where \( \chi(\lambda) \) differs from zero, and observe an excellent agreement between two dependencies.

In Fig. 2 one can observe damped oscillations in total losses. Presence of these damped oscillations is explained by the analysis of Eq. (5). Function \( TL_{\text{Abs}}(\lambda) \) is presented as a sum of two terms: the first one is monotonically decreasing term and the second one is the oscillating term. It is seen from Eq. (5) that for thicker films oscillations magnitude and frequency are higher. From Eq. (5) one can see also that oscillations are dependent on the ratio \( n_s/n_p \). If this ratio essentially differs from one, the second term in Eq. (5) provides oscillations in total losses. For example, damped oscillations in total losses are expected when someone analyzes data related to samples of TiO\(_2\), HfO\(_2\), Nb\(_2\)O\(_5\) films. If the film refractive index \( n_s \) is close to the substrate refractive index \( n_s \), the second term in brackets in Eq. (5) tends to the constant \((n_s - 1)/(n_s + 1)\), and the whole expression becomes a monotonically decreasing function. It means that in the case of a slightly absorbing thin film with low refractive index, damped oscillations in total losses are very small and they cannot be observed. As an example of slightly absorbing film with low refractive index we can consider a film composed of a mixture of TiO\(_2\) and SiO\(_2\), with a small content of TiO\(_2\) [8].

In this subsection, we have shown theoretically that damped oscillations in total losses can be attributed to a slight absorption in the considered film.

### 2.3. Influence of thickness non-uniformity on total losses spectral behavior

Our third supposition is that oscillations in total losses can be attributed to thickness non-uniformity. In many spectrophotometers, reflectance and transmittance measurements are performed using different attachments. This requires the manipulation of the sample. Thus, it might be for reflectance and transmittance measurements, different areas of the surface are tested. If the deposition process produces films with non-uniform thickness, it is reasonable to expect that reflectance and transmittance measurements are affected by different film thickness.

Assume that transmittance is measured at a point where film thickness equals to \( d \) and reflectance is measured at a point where film thickness equals to \( d + \Delta d \). In order to verify our supposition we consider total losses

\[ TL(d + \Delta d, \lambda) = 100\% - T(d, \lambda) - R(d + \Delta d, \lambda) \]

in the frame of the first-order perturbation theory with small parameter \( \Delta d \). As a result, in the case of non-absorbing thin film, we derive an approximate relation \( TL_{\Delta d}(\lambda) \) for \( TL(\Delta d, \lambda) \):

\[ TL_{\Delta d}(\lambda) = -\frac{\pi \Delta d}{4\lambda} q^{(s)} \sin 2\varphi \left[ f^{(s)} + f^{(p)} \right], \]

\[ f^{(s,p)} = T^2 \left[ (q^{(s,p)2} - (q_{a}^{(s,p)})^2) \left( 1 - \frac{q_{a}^{(s,p)}}{q_{a}^{(s,p)}} \right)^2 \right] \]

where \( q^{(s,p)}, q_{a}^{(s,p)} \) are effective refractive indices for \( s \)- and \( p \)-polarization, \( \varphi = (2\pi/\lambda) q^{(s)} d \) (see, for example, [9]). In the case of normal incidence, relation (8) is simplified:

\[ TL_{\Delta d}(\lambda) = -\frac{\pi \Delta d}{2\lambda} n \sin 2\varphi \left[ T^2 (n^2 - 1) \left( 1 - \frac{n_s^2}{n^2} \right) \right] \]

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In order to verify the accuracy of approximate relation (8), we compare exact values of $TL(\Delta d, \lambda)$ and values of $TL_{\Delta d}(\lambda)$. We calculated the exact values for two cases: when the substrate back side is taken into account and without the substrate back side. In Fig. 3 one can see that $TL(\Delta d, \lambda)$ calculated for two cases are almost undistinguishable. We provide calculations of $TL(\Delta d, \lambda)$ and $TL_{\Delta d}(\lambda)$ values for a model sample. Refractive indices $n$ and $n_s$ as well as substrate and film thickness $d$, are the same as in Subsection 2.1. We take $\Delta d$ value equal to 0.1% of the film thickness, i.e. $\Delta d = 0.5$ nm. In Fig. 3 we compare spectral dependencies of $TL(\Delta d, \lambda)$ and its approximation $TL_{\Delta d}(\lambda)$ for AOI $\theta = 7^\circ$. An excellent agreement between these two dependencies is an evidence that relation (8) accurately approximates function (7).

In Fig. 3 one can observe oscillations with magnitudes reaching 1%. This is consistent with theoretical spectral behavior of $TL_{\Delta d}$ (Eq. (8)), where oscillating terms are included. Analyzing Eqs. (8) and (9), one can notice that magnitudes of oscillations are proportional to the ratio $n_s/n$. It means that oscillations in total losses appear only if a sufficient contrast between $n$ and $n_s$ is provided. Such contrast may be achieved when films with high and medium refractive indices, such as TiO$_2$, Ta$_2$O$_5$, Nb$_2$O$_5$, Al$_2$O$_3$, HfO$_2$, on glass or quartz substrates, are studied. In the cases when thin film samples with low contrast between $n$ and $n_s$ are considered, oscillations caused by thickness non-uniformity can not be observed in total losses.

In this section, on the basis of qualitative analysis of approximate relations for total losses, we proved theoretically that the presence of oscillations in total losses can be caused either by differences in AOI used for separate $T$ and $R$ measurements, or by slight absorption in the thin film, or by slight thickness non-uniformity. We found that in all three cases, oscillations appear only in thin film samples with a sufficient contrast between refractive indices of the film and substrate. In the next section we will consider experiments supporting these results.

3. Experimental samples and measurement data

In order to confirm theoretical conclusions obtained in the previous section, we produced special experimental samples and measured several sets of reflectance and transmittance data. In the course of our study we produced five samples. First three samples were produced with magnetron-sputtering Leybold Optics HELIOS plant. These samples are Ta$_2$O$_5$, Nb$_2$O$_5$ and SiO$_2$ films on Suprasil substrates. Geometrical thicknesses of the films are about 515 nm, 490 nm and 400 nm, respectively. Substrate thickness is 6.35 mm, substrate diameter is 25 mm. Deposition rates for Ta$_2$O$_5$ were 0.35 nm/sec, for Nb$_2$O$_5$— 0.4 nm/sec, for SiO$_2$— 0.45 nm/sec.
Two other samples were prepared by e-beam evaporation in a modified Varian 3117 chamber. These samples are TiO$_2$ and HfO$_2$ films on Suprasil substrates. The base pressure was $8 \cdot 10^{-6}$ Torr, partial pressure of oxygen was $9 \cdot 10^{-5}$ Torr for both materials. Deposition rates were 0.5 Å/s for HfO$_2$ and 2 Å/s for TiO$_2$. The films were deposited on 1 mm thick Suprasil substrates that were placed onto rotating calotte to insure better uniformity of film thickness and pre-heated to 250°C. Geometrical film thicknesses were about 504 nm and 400 nm, respectively.

The choice of the sample structures is explained as follows. We chose thin film materials (Ta$_2$O$_5$, Nb$_2$O$_5$, and TiO$_2$) with refractive indices providing a high contrast with refractive index of the substrate and also thin film material (SiO$_2$) with refractive index having low contrast with substrate refractive index. Three thin films (Ta$_2$O$_5$, Nb$_2$O$_5$ and SiO$_2$) produced by magnetron sputtering are non-absorbing in the spectral range from 380 to 800 nm (Nb$_2$O$_5$ may have very slight absorption in the range from 380 to 400 nm). Two thin films (TiO$_2$ and HfO$_2$) produced with e-beam evaporation are slightly absorbing at short wavelengths of the considered spectral range. The film thicknesses were chosen large enough in order to obtain noticeable oscillations.

For each sample, transmittance and reflectance were measured using two spectrophotometers: Perkin Elmer Lambda 950 and Perkin Elmer Lambda 25. At Perkin Elmer 950, $T$ spectra were taken at normal incidence and also at AOI $\theta_1 = 7^\circ$, $R$ data were measured at AOI $\theta_2 = 7^\circ$. Measurements were performed in the visible spectral range. Reflectance of the Suprasil 6.35 mm thick substrate (with back side into consideration) was taken as the reference for $R$ measurements.

At Perkin Elmer Lambda 25, transmittance data were taken at normal incidence and reflectance data were measured at AOI $\theta_2 = 6^\circ$ in the spectral range from 325 nm to 1100 nm. The measurements were done with 1 nm step. Beam size was $10 \times 2$ mm$^2$, slit size 1 nm, scan rate 240 nm/min. Reflectance measurements were done with a standard relative specular reflectance accessory, meaning that the sample has to be moved between $T$ and $R$ measurements. A calibrated Ag mirror was used as a reference for reflectance measurements.

4. Experimental verification on the basis of $R$ and $T$ measurements taken at different points of sample

In this section we check our theoretical conclusions using experimental data.

4.1. Total losses spectral behavior in the case of different AOI

Our first theoretical conclusion was that different AOI used in the course of separate $T$ and $R$ measurements may cause oscillations in total losses. According to theoretical predictions obtained in Subsection 2.1, we expect oscillations in the cases of films with high refractive index values. Crosses in the left and right sides of Fig. 4 present experimental total losses calculated as $TL(\lambda_j) = 100\% - T(\theta_1 = 0^\circ, \lambda_j) - R(\theta_2 = 7^\circ, \lambda_j)$ for Ta$_2$O$_5$ and Nb$_2$O$_5$ samples, respectively. We do not present total losses in TiO$_2$ and HfO$_2$ samples here because thin films in these samples are slightly absorbing, and we do not want to mix effects of two different factors: deviations in AOI and absorption.

In Figs. 4(a)–4(b) one can observe noticeable oscillations with approximately 0.8% magnitude. Solid curves in Fig. 4 show theoretical function $TL_{AOI}(\lambda)$ calculated using Eq. (2) for Ta$_2$O$_5$ and Nb$_2$O$_5$ samples. Function $TL_{AOI}(\lambda)$ is calculated for $n(\lambda)$ and $d$ values determined in the course of characterization process based on normal incidence transmittance data. Determined refractive index of Ta$_2$O$_5$ is described by Cauchy model (3) with $A_0 = 2.075463, A_1 = 0.0172, A_2 = 0.00166$, dispersion of Nb$_2$O$_5$ refractive index is described by Cauchy model with $A_0 = 2.250124, A_1 = 0.0176, A_2 = 0.00463$. Film thicknesses of Ta$_2$O$_5$
Fig. 4. Comparison of $TL(\lambda) = 100\% - R(7^\circ, \lambda) - T(0^\circ, \lambda)$ calculated from experimental data and $TL_{AOI}(\lambda)$ calculated by Eq. (2): (a) Ta$_2$O$_5$, (b) Nb$_2$O$_5$.

Fig. 5. Total losses $TL(\lambda) = 100\% - T(0^\circ, \lambda) - R(7^\circ, \lambda)$ calculated from experimental data of SiO$_2$ sample taken by Perkin Elmer Lambda 950.

and Nb$_2$O$_5$ films were found equal to 517 nm and 480 nm, respectively. Refractive index of Suprasil substrate [10] in the visible range can be described by Cauchy formula with $A_0 = 1.448, A_1 = 0.00386, A_2 = -0.000397$.

In Figs. 4(a)–4(b) one can observe a satisfactory agreement between experimental and theoretical curves. Generally, the curves are consistent: it is seen that oscillations magnitudes are in a good agreement and positions of extrema are quite close. This correspondence is a convincing argument for our theoretical conclusion about influence of difference in AOI used for separate $T$ and $R$ measurements on $TL$ spectral behavior. Observed discrepancy between theoretical and experimental curves will be commented later in Subsections 4.3 and 4.4.

In Fig. 5 one can observe absence of oscillations in total losses related to SiO$_2$ sample. This is in a full agreement with theoretical predictions obtained in Subsection 2.1 that difference in AOI in the cases of samples with $n \approx n_s$ cannot cause oscillations in total losses.

At the next step of our study, we check whether oscillations in total losses disappear if AOI, used for $T$ and $R$ measurements, are equal. Crosses in Figs. 6(a)–6(b) present total losses $TL(\lambda_j) = 100\% - T(7^\circ, \lambda_j) - R(7^\circ, \lambda_j)$ calculated for Ta$_2$O$_5$ and Nb$_2$O$_5$ samples. It is seen that despite of the equality in AOI oscillations in total losses are still clearly observed. Presence of oscillations means that not only difference in AOI may cause oscillations, there is another reason. According to our theoretical study, oscillations in total losses can be also explained by
4.2. Total losses spectral behavior in the case of slightly absorbing thin film

In Subsection 2.2 we made a supposition that oscillations in total losses can be caused by absorption in considered thin films. In Figs. 7(a)–7(b) we show total losses calculated from T and R measurement data related to TiO$_2$ and HfO$_2$ samples, respectively. In this case we calculate total losses as $\text{TL}(\lambda_j) = 100\% - T(7^\circ, \lambda_j) - R(7^\circ, \lambda_j)$, where $T$ and $R$ were taken by Perkin Elmer Lambda 950. In Figs. 7(a)–7(b) in the shorter wavelength ranges from 350-380 nm to 600 nm one can clearly observe damped oscillations in total losses.

Solid curves in Fig. 7 present theoretical values of $\text{TL}_{\text{Abs}}(\lambda_j)$ calculated using Eq. (5) for TiO$_2$ and HfO$_2$ samples. Refractive indices $n$ and extinction coefficients $\chi$ in Eq. (5) were found in the course of characterization process based on $T$ and $R$ measurements [4, 11]. For data processing we used a homogeneous thin film model that incorporates Cauchy formula for $n(\lambda)$ and exponential formula for $\chi(\lambda)$.

For refractive index and extinction coefficient of TiO$_2$ film in the spectral range from 380 nm to 800 nm, we obtained $A_0 = 2.16895, A_1 = 0.01206, A_2 = 0.00588, B_0 = 3.07241, B_1 = 0, B_2 = -18.22825$. For refractive index and extinction coefficient of HfO$_2$ film in the spectral range from 350 nm to 800 nm, we obtained $A_0 = 1.927513, A_1 = 0.0072, A_2 = 0.000596,
$B_0 = 0.0573, B_1 = 0, B_2 = -8.621482$. Physical thicknesses of TiO$_2$ and HfO$_2$ films were determined equal to 504 nm and 400 nm, respectively.

In Figs. 7(a)–7(b) one can observe a good agreement between experimental data and theoretical predictions at the shorter wavelength spectral ranges where absorption reveals itself noticeably. Positions of the first maximum in theoretical and experimental curves are quite close, positions of the second maxima are close for TiO$_2$ sample. It is clearly seen from Fig. 7 that these oscillations in total losses are caused not only by absorption, but by another reason as well. According to our theoretical study, oscillations in total losses can be also attributed to slight thickness non-uniformity. We shall check this supposition in the next subsection.

### 4.3. Total losses spectral behavior in the case of slight thickness non-uniformity

Our third supposition made in Subsection 2.3 is that the presence of oscillations in total losses can be explained by a slight thickness non-uniformity. In Fig. 8(a) (Ta$_2$O$_5$) and Fig. 8(b) (Nb$_2$O$_5$) crosses indicate total losses $TL(\lambda_j) = 100\% - R(\theta, \lambda_j) - T(\theta, \lambda_j)$ calculated from $T$ and $R$ data measured at Perkin Elmer Lambda 950. Solid curves in Figs. 8(a)–8(b) present theoretical spectral dependencies $TL_{\Delta d}(\lambda)$ calculated using Eq. (8). For calculation we used the same refractive indices $n(\lambda)$ and the same physical thicknesses $d$ of Ta$_2$O$_5$ and Nb$_2$O$_5$ thin films as in Subsection 4.1. We specify $\Delta d$ as a value providing the best fit with experimental data, namely, we take $\Delta d = 1$ nm for Ta$_2$O$_5$ film and $\Delta d = 0.5$ nm for Nb$_2$O$_5$ film. It should be mentioned here that according to Eq. (8), $\Delta d$ values affect only magnitudes of the oscillations, but not positions of extrema.

Both $\Delta d$ values are less than 0.2% of respective film thicknesses $d$. This is consistent with estimated thickness non-uniformity in the HELIOS plant. The thickness non-uniformity is estimated as 0.1–0.2% of total geometrical thickness.

In Fig. 8 one can observe a good agreement between experimental and theoretical dependencies of total losses. This agreement can be considered as a convincing argument for our supposition about the influence of the thickness non-uniformity on total losses spectral behavior.

Let us consider another evidence of thickness variation. In Fig. 9(a) we compare two normal incidence transmittance measurements $T^{(1)}(\lambda_j)$ and $T^{(2)}(\lambda_j)$ related to Ta$_2$O$_5$ sample. These measurements were performed on two different days. Both measurements were taken at the same Perkin Elmer Lambda 950 spectrophotometer. Although these two transmittance spectra
look undistinguishable, they are shifted one with respect to another. The evidence of this shift is confirmed by the plot in Fig. 9(b), where difference between two normal incidence transmittance measurements $\delta T(\lambda_j) = T^{(1)}(\lambda_j) - T^{(2)}(\lambda_j)$ is presented by crosses. In Fig. 9(b) oscillations of about 0.8% magnitude are clearly observed. The observed shift between two measured transmittance curves cannot be explained by any film structural changes because HELIOS plant produces dense and stable films [12, 13].

In Fig. 9(b) solid curve presents approximate relation for $\delta T(\lambda)$. This relation coincides with $T_{Ld}(\lambda)$ described by Eq. (9). Let us assume that the film thickness in the course of the first measurement was $d$ and in the course of the second measurement it was to $d + \Delta d$. Then, taking into account that $T = 1 - R$, we obtain:

$$\delta T(\lambda) = T(d, \lambda) - T(d + \Delta d, \lambda) \approx - \frac{\partial T(\lambda)}{\partial \Delta d} \bigg|_{\Delta d = 0} \Delta d = \frac{\partial R(\lambda)}{\partial \Delta d} \bigg|_{\Delta d = 0} \Delta d = T_{Ld}(\lambda) \quad (10)$$

The best fit between magnitudes of experimental oscillations and theoretical oscillations predicted by Eq. (8) is achieved for $\Delta d = 0.5$ nm. This value is less than 0.1% of the geometrical thickness of Ta$_2$O$_5$ film, that is in accordance with estimated thickness non-uniformity in Helios plant.

4.4. Effect of systematic errors in $R$ and $T$ data on the correspondence between experimental and theoretical total losses

As it has been mentioned above, in Figs. 4, 7 one can observe a discrepancy between experimentally found total losses and their theoretical approximations. Namely, in Fig. 4 theoretical $T_L(\lambda)$ values are shifted to the shorter wavelengths with respect to experimental total losses. In Fig. 7 in the wavelength range above 400-450 nm we observe oscillations while theoretically predicted level of oscillations is very small.

The first explanation of the observed discrepancies is that oscillations may be caused not only by difference in AOI $\theta_2$ and $\theta_1$ or only by thin film absorption but by combinations of these effects and effect of thickness non-uniformity.

The second explanation can be addressed to systematic errors in measurement spectral photometric data. In order to calculate approximate functions $T_{L\text{AOI}}(\lambda)$ and $T_{\text{Abs}}(\lambda)$, we substituted refractive indices $n$ and thicknesses $d$ determined in the course of characterization process. This process in its turn is based on measurement data. Accuracy of characterization results $n$ and $d$
are dependent on the accuracy of measurement data [5, 14]. Particularly, systematic offset in spectral photometric data causes offset in $n$ values.

According to estimation obtained in [14], systematic offset in measured transmittance spectra causes the following offset of film refractive index $\Delta n$:

$$\Delta n = \frac{(n^2 + n_s)^3}{8n_m(n_s - n^2)} \Delta T. \quad (11)$$

Our characterization process is based on the minimization of a standard discrepancy function estimating the closeness between experimental and model spectral characteristics of the investigated sample [4, 11, 15]. In the course of minimization process, refractive index changes by a value close to that given by Eq. (11) in order to compensate for the measurement data offset.

Systematic offset in experimental $T/R$ data can be estimated on the basis of comparison of the data related to a bare substrate and theoretical transmittance/reflectance data calculated for this substrate [16]. In the course of characterization of Ta$_2$O$_5$ and Nb$_2$O$_5$ samples, we used only normal incidence $T$ data. Taking refractive index of Suprasil from Heraeus Quartzglas Catalog [10], we estimated systematic offset in transmittance data as -0.6%. According to Eq. (11), possible offsets in refractive indices of Ta$_2$O$_5$ and Nb$_2$O$_5$ equal to 0.018, that is about 0.8% of $n$ for both films.

In Figs. 10(a)–10(b) we compare experimental total losses $TL(\lambda_j) = 100\% - T(\theta_1 = 0^\circ, \lambda_j) - R(\theta_2 = 7^\circ, \lambda_j)$, calculated for Ta$_2$O$_5$ and Nb$_2$O$_5$ samples, theoretical approximations of these losses $TL_{AOI}(\lambda)$ calculated by Eq. (2) with $n(\lambda)$ values presented in Subsection 4.1, and total losses $TL_{AOI}(\lambda)$ calculated by Eq. (2) with $n(\lambda) + \Delta n$, where $\Delta n = 0.018$. In Figs. 10(a)–10(b) one can observe that $TL_{AOI}(\lambda)$ curve calculated with $n(\lambda) + \Delta n$ are in a better agreement with experimental total losses than $TL_{AOI}(\lambda)$ dependence calculated with $n(\lambda)$. Residual discrepancy should be addressed to a slight thickness non-uniformity.

5. Experimental verification on the basis of $R$ and $T$ measurements taken at the same point of sample

As it has been demonstrated above, measurement setup impacts presence of oscillations in total losses. Namely, $T$ and $R$ are typically measured at different AOI due to specific features of optical registration scheme, i.e., transmittance is usually measured at normal incidence, while reflectance has to be measured at oblique incidence, depending on the specific attachment.

Another reason of oscillations, thickness non-uniformity, may cause oscillations in measurements taken by spectrophotometers where the measurement of reflectance and transmittance
requires sample manipulations. In these setups, it is not guaranteed that transmittance and reflectance are measured at the same point of the studied sample. There are, however, experimental configurations where no sample manipulation is required between $R$ and $T$ measurements [16, 17] and the area of the studied sample is the same for both measurements. In this case, oscillations caused by thickness non-uniformity cannot contribute to the total losses. Oscillations in total losses in the cases when measurements are performed using such setups, can be caused either by difference in AOI or by film absorption.

Let us demonstrate the latter statement using experimental examples. As the first measurement device we chose an advanced accessory to the double beam UV-Vis-NIR spectrophotometer Cary 5000, developed by Agilent Technologies [16]. In this absolute variable angle transmittance and reflectance accessory, the linearly polarized beam that illuminates the sample can be measured in transmission, and by rotating the detector assembly about an axis through the sample and perpendicular to the plane of incidence, in reflection. Thus the same spot on the sample is used for both $R$ and $T$ measurements. As the second device we use a different unit of the same advanced accessory installed in the same Cary 5000. The measurements were made using different sample mounts.

As an experimental sample, we consider a sample of Ta$_2$O$_5$ film of 292 nm thickness on Suprasil substrate of 25 mm diameter and 6.35 mm thickness. The film was deposited using HELIOS deposition plant mentioned in Section 3. Two Ta$_2$O$_5$ samples, one chosen for this example and another one described in the previous text, differ only by film thickness. For our demonstration, we consider transmittance data measured at $7^\circ$ and $10^\circ$, and reflectance data measured at $10^\circ$, both measurements for $s$-polarized light.

In the first experimental example we consider $T$ and $R$ spectra taken by use of the advanced accessory. First, we consider total losses calculated on the basis of $T$ and $R$ measurements taken at different AOI: $TL(s)(\lambda_j) = 100\% - T(s)(7^\circ, \lambda_j) - R(s)(10^\circ, \lambda_j)$. These total losses are shown in Fig. 11(a) by crosses. Oscillations in experimental total losses of about 0.4% magnitude are clearly observed. Solid curve in Fig. 11(a) presents approximation of total losses $TL_{AOI}(\lambda)$ calculated from experimental data.
series:

\[
TL_{\text{AOI}} = \left\{ \frac{4\pi d}{\lambda} \frac{n_s}{n} \sin^2 \varphi \left[ \frac{(n^2 - 1) \left( 1 - \frac{n_s}{n} \right)^2}{\left[ (1 + n_s)\cos \varphi + \frac{n_s}{n} + n \right] \sin \varphi} \right]^2 + \right. \\
\left. + \frac{2n_s}{\left( 1 + n_s \right)^2 \cos^2 \varphi + \frac{n_s}{n} + n \sin^2 \varphi} \left( \frac{(\theta_2^2 - \theta_1^2)}{n^2} \right) \right\} \\ a(n, n_s) = -(1/n_s - n_s)^2, \\
b(n, n_s) = \left( (1 + n_s)^2 + (n_s/n + n)^2 + (n_s/n)^2 - \left( n^2/n_s^2 \right)^2 + n^2 - n^4 \right)/n^2
\]

In Fig. 11(a) one can observe a good agreement between theoretical and experimental data. This agreement confirms that the effect of thickness non-uniformity does not contribute to total losses spectral behavior.

Secondly, in Fig. 11(b) we show experimental total losses, calculated from \( T \) and \( R \) measurements taken at equal AOI: \( TL^{(s)}(\lambda) = 100\% - T^{(s)}(\gamma, \lambda_j) - R^{(s)}(\gamma, \lambda_j) \). Absence of any oscillations is clearly observed. This is another confirmation of the fact that thickness non-uniformity does not reveal itself in total losses in the cases when \( T \) and \( R \) measurements are performed at devices providing \( T \) and \( R \) measurements at the same point of the investigated sample.

In the second experimental example we performed measurement of reflectance and transmittance using different unit of the advanced accessory in Cary 5000. The measurements were made using different sample mounts. In Fig. 12(a) the black curve shows the first transmittance measurements. Crosses show the same sample re-measured in a different accessory unit 4 months later. Visually these measurements are undistinguishable. At the same time, in Fig. 12(b) we show the difference \( TL^{(s)}(\lambda_j) = T^{(s,1)}(\gamma, \lambda_j) - T^{(s,2)}(\gamma, \lambda_j) \) between two transmittance spectra marked by crosses. Oscillations of about 0.15% magnitude are clearly observed. Solid curve in Fig. 12(b) indicates approximation of total losses calculated using Eqs. (10), (12) with \( \Delta d = -0.3 \) nm. This thickness non-uniformity value corresponds to about 0.1% of geometrical film thickness \( d = 292 \) nm. This is in a full agreement with thickness non-uniformity level in HELIOS plant.

\[\text{Fig. 12. (a) Comparison of two transmittance spectra taken by Cary 5000 spectrophotometer with two different accessories. The data are related to Ta}_2\text{O}_5 \text{sample described in Section 5, AOI=7°, s-polarization case. (b) Comparison of } \delta T^{(s)}(\lambda) = T^{(s,1)}(\gamma, \lambda) - T^{(s,2)}(\gamma, \lambda) \text{ calculated from experimental data and its approximation by Eqs. (10), (12).}\]
6. Conclusions

In our study we found out several reasons of oscillations in total losses spectra. These reasons are originated from either measurement device arrangement either from film optical properties or from combinations of these two factors. The first reason of the oscillations is difference in AOI used for separate transmittance and reflectance measurements.

The second reason of the oscillations in total losses is thin film absorption. Such oscillations appear independently on measurement device because the origin of these oscillations is related to optical properties of investigated thin film.

The third reason of oscillations in total losses is slight thickness non-uniformity. Such oscillations are caused by thin film non-uniformity and measurement arrangement as well. We demonstrated that these oscillations do not appear when measurement devices allowing transmittance and reflectance measurement at the same point of investigated sample are used.

We demonstrated that frequently combined effects of the reasons listed above reveal themselves in total losses spectral behavior. Our results may help researchers dealing with characterization and measurement problems to distinguish between good and poor-quality measurement data. Using approximate relations (2), (5), (8) and (9), one can estimate, at least roughly, possible oscillations in total losses. Particularly, relations described by Eqs. (2), (5), (8) and (9) allow one to estimate magnitudes of oscillations and extrema positions. As optical constants of the investigated thin film, one can substitute to Eqs. (2), (5), (8) and (9) refractive indices and extinction coefficients obtained previously or determined by other authors for the films of the same materials and deposited by the use of the same deposition technique (see, for example, [4, 14, 18]). Geometrical thickness $d$ is typically known with a sufficient accuracy.

If total losses behavior differs significantly from that predicted by relations (2), (5), (8) and (9), namely, extrema positions are completely different or oscillations magnitudes are essentially higher, this may be considered as an indication of poor quality of measurement data. In particular the presence of oscillations in total losses in the cases of samples with low ratio between film and substrate refractive indices reveal problems in measurement data.

Our work benefits from using diverse experimental basis, including samples, produced by two different deposition techniques, and comprising spectral photometric data, made by four spectrophotometers of two different types: Perkin Elmer spectrophotometers and variable angle Cary spectrophotometers. In our study, we considered films with different optical properties (with and without absorption) and samples with high and low ratios of film and substrate refractive indices.

In this work we presented a comprehensive study of total losses spectra. These minor effects play an important role and balance on the limits of modern measurement devices. We believe that our results will facilitate further perfection of characterization and measurement processes.

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