Quality control of oblique incidence optical coatings based on normal incidence measurement data

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Abstract: We demonstrate selection of reliable approaches for post-production characterization of oblique incidence multilayer optical coatings. The approaches include choice of input information, selection of adequate coating model, corresponding numerical characterization algorithm, and verification of the results. Applications of the approaches are illustrated with post-production characterization of oblique incidence edge filter, oblique incidence beam splitter and oblique incidence 43-layer quarter-wave mirror.

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1. Introduction

The determination of layer parameters of produced multilayer coatings (reverse engineering (RE) of optical coatings) provides a feedback for the design-production chain. The main intention of the RE is to detect systematic and random errors in layer parameters, to improve deposition control and, therefore, to raise the quality of optical coating production [1–4]. In order to properly characterize produced coatings, a reliable RE approach should be chosen.

We refer to a combination of the experimental data set, coating model, corresponding numerical algorithm, and verification method as a RE approach. It is known that optical coatings parameters (levels of thickness errors, stability of optical constants of layers, degree of inhomogeneity, etc.) are dependent on the deposition process and on the monitoring technique used in the course of the deposition. Hence, coatings deposited under different deposition conditions correspond to different mathematical models. RE procedures, therefore, are different. Evidently, the RE approach should be the same for all coatings deposited under the same deposition conditions. Once carefully chosen, the approach can be applied further to post-production characterization of all similar optical coatings. Such selection is especially important for mass production of optical coatings, allowing one to improve the quality of manufactured coatings and the production yield.

The most typical RE approaches are based on the numerical analysis of normal incidence or quasi-normal incidence transmittance (T) and/or reflectance (R) data related to a produced coating sample. This approach is quite simple from an experimental point of view because normal incidence transmittance and quasi-normal incidence reflectance can be measured using a wide set of spectrophotometers. The approach, however, may easily lead to unreliable results. This unreliability can be attributed to an insufficient information content of some experimental data sets and to the influence of measurement errors on the obtained results. Data related to broadband reflectors or to broadband antireflection coatings (AR) can be considered as examples of such low-informative data sets. Some examples of insufficient information content of measurement data are presented in our recent publication [2].

Modern state-of-the-art thin film coatings are becoming more complicated and as demands on optical coating performance continue to increase, the ambiguity of RE continues to grow with the typically growing number of layers. Generally, in order to overcome
ambiguity in RE, approaches utilizing larger sets of measurement data should be developed. Recently, RE approaches based on \textit{in situ} data analysis have been actively developed \cite{2,3,5,6}.

In order to obtain reliable RE results one should take care that not only a sufficient amount of input information is considered but also that reasonable optical coating model and numerical algorithm is used for data analysis. All \textit{a priori} information on a produced coating, such as the course of the deposition process, deposition conditions and accuracy of measurement data should be taken into account by the model and algorithm. In the first turn, the algorithm should provide RE results which can be used for adjusting a deposition process (for example, calibration of quartz crystal monitors) or for correcting the optical coating model (for example, changing optical constants of layer materials). Achievement of perfect fitting of experimental data by model spectral characteristics is not necessarily an indication of good quality RE results. For example, in many cases a perfect fit can be achieved with physically non-sensible models.

The important issue concerning RE process is the validation of the results. For this purpose, various approaches can be used. Recently, we reported on verification of RE results using specially produced coatings with imposed errors to layer thicknesses \cite{2,5}. In \cite{6} the verification of the RE results with the help of group delay measurements was demonstrated.

In the present paper we consider the selection of reliable RE approaches in the case of multilayer coatings working at oblique incidence. Typically, RE processes for such coatings are based on the analysis of normal incidence data (\textit{ex situ} and/or \textit{in situ}). It is well known, however, that a good fit of normal incidence data does not guarantee the correct spectral performance at oblique incidence. Ideally it would be useful to increase the amount of input information by adding oblique incidence measurement data. Unfortunately, as discussed below, respective measurements are more complicated than normal incidence ones and it is therefore desirable to minimize them. Therefore, development of RE approaches which allow to obtain reliable oblique-incidence spectral characteristics on the basis of normal or quasi-normal incidence measurements only is an actual problem.

Accurate oblique-incidence \textit{R} and \textit{T} measurements are typically more complicated than normal and quasi-normal incidence measurements \cite{4,7}. Multiangle photometric measurements of real samples typically need to measure not only the directly transmitted or reflected light beam but also sufficient of the internally reflected beams in order to provide an accurate measurement of the total transmittance or reflectance of the sample being studied. At normal and near-normal incidence the aperture required to collect the internal reflections is typically not much larger than that needed to acquire the directly transmitted or reflected beam. However, as the angle of incidence at the sample surface increases, the lateral separation of the internally reflected beams increases through a maximum, near 50° angle of incidence (AOI), before decreasing again as the beam approaches grazing incidence. Further, the lateral separation of the beams depends not only on the AOI but also on the thickness of the sample to be measured. The lateral separation of the internally reflected beams will be greater for a thick sample than for a thin sample. Lastly, as the AOI increases, the area on the surface of the sample on which the incident beam impinges grows as the lateral dimension increases as $1/\cos(AOI)$. Hence optical systems for accurate measurement in the range from normal to oblique incidence need to incorporate sufficient detector aperture size to collect the wide range of lateral beam spread that will be encountered. As a result careful consideration needs to be given to the physical dimensions of the sample so that all the expected internal reflections can be generated and escape the sample surface (front and rear) and be measured. For example, the direct reflection from a fused silica flat plate is approximately 3.5% of the incident flux; the first internal reflection is nearly as great at 3.2%. The second internal reflection falls dramatically to well below 0.01%, Similarly the direct transmission through a fused silica plate is approximately 93.2% and transmitted portion of the first internal reflection is approximately 0.1% and the second is essentially zero. Clearly, measurement systems that collect only the directly reflected or transmitted beam but neglect the internally reflected and transmitted portions provide a skewed estimate of the total reflectance and
transmittance of the sample. Ideally an instrument or measurement technique specially configured for oblique incidence measurements or special accessories to devices typically used for normal incidence measurements, are required.

In our present work we propose to perform careful oblique-incidence measurements only for several test coatings and to use the obtained experimental data for adjusting RE process. The RE process is to be performed on the basis of normal incidence data, oblique incidence measurements are used only for verification of the results. For further oblique incidence coatings produced under the same deposition conditions, the adjusted RE procedure can be used without additional verification.

In Section 1 we show an example of unreliable RE approach. In the rest of the current work we demonstrate selection of reliable RE approach using three quite complicated problems. We perform post-production characterization of coatings deposited under different deposition conditions and, therefore, obtain different RE approaches. In Sections 2–4 we characterize oblique incidence edge filter, oblique incidence beam splitter and oblique incidence 43-layer quarter-wave mirror, respectively. Our final conclusions and recommendations are presented in Section 5.

2. Example of unreliable reverse engineering of 21-layer quarter-wave mirror

In order to demonstrate an unreliable RE approach we consider an example of post-production characterization of 21-layer quarter-wave mirror for the central wavelength of 400 nm. We used HfO₂ and SiO₂ as high (H) and low (L) index materials, respectively. Substrate is BK7 of 1 mm thickness. Nominal refractive indices of the materials and the substrate are presented in Table 1.

Table 1. Refractive indices of layer materials and substrates (λ should be expressed in μm).

| Material  | Deposition process/plant | Dispersion formula |
|-----------|--------------------------|--------------------|
| HfO₂      | e-beam evaporation/Varian 3117 | $n(\lambda) = 1.876067 + 1.04448781 \times 10^{-2} / \lambda^2 + 1.7964229 \times 10^{-4} / \lambda^4$ |
| SiO₂      | e-beam evaporation/Varian 3117 | $n(\lambda) = 1.44765 + 3.8009553 \times 10^{-3} / \lambda^2 - 5.379729 \times 10^{-5} / \lambda^4$ |
| TiO₂      | e-beam evaporation/SYRUSPro 1100 | $n^2(\lambda) = 1 + 4.062408 \lambda^2 / (\lambda^2 - 0.048857) + 1.401662 \lambda^2 / (\lambda^2 - 80)$ |
| SiO₂      | e-beam evaporation/SYRUSPro 1100 | $n^2(\lambda) = 1 + 1.158586 \lambda^2 / (\lambda^2 - 0.010727) + 0.5 \lambda^2 / (\lambda^2 - 61)$ |
| Nb₂O₅     | Magnetron sputtering/Helios | $n(\lambda) = 2.218485 + 0.021827 / \lambda^2 + 0.003999568 / \lambda^4$ |
| SiO₂      | Magnetron sputtering/Helios | $n(\lambda) = 1.465 + 4.710810 \times 10^{-4} / \lambda^4$ |
| HfO₂      | e-beam evaporation/SYRUSPro 710 | $n(\lambda) = 1.871707 + 6.84525161 \times 10^{-3} / \lambda^2 + 4.83324061 \times 10^{-4} / \lambda^4$ |
| SiO₂      | e-beam evaporation/SYRUSPro 710 | $n(\lambda) = 1.456819 + 1.993508 \times 10^{-3} / \lambda^2 + 7.181666 \times 10^{-10} / \lambda^4$ |
| BK7       | | $n(\lambda) = 1.504167 + 4.79862931 \times 10^{-3} / \lambda^2 - 4.17832610^{-5} / \lambda^4$ |
| Fused Silica | | $n(\lambda) = 1.448187 + 3.56075971 \times 10^{-3} / \lambda^2 - 7.487681 \times 10^{-6} / \lambda^4$ |
| Glass B260 | | $n(\lambda) = 1.502671 + 5.3319 \times 10^{-3} / \lambda^2 + 4.9185 \times 10^{-4} / \lambda^4$ |
| Suprasil   | | $n(\lambda) = 1.443268 + 4.0610 \times 10^{-7} / \lambda^2 + 6.94817641 \times 10^{-10} / \lambda^4$ |

The first layer of the mirror is a HfO₂ layer. The experimental sample was prepared at Rudjer Boskovic Institute (Zagreb, Croatia). We denote this sample as HR400-BK7. Layer thicknesses were controlled by a quartz crystal monitor. Reflectance and transmittance photometric data were taken in the spectral range from 300 nm to 2500 nm at AOI of 7°, 10°, 20°, 30°, 40°, 50°, and 60° for s- and p-polarized light. The accuracy of measurement data, estimated in [4], is 0.2%.
In Fig. 1(a) we compare theoretical and experimental reflectance data (s-polarization case, AOI = 7°). In Fig. 1(a) one can observe deviations between theoretical and measured data. These deviations can be attributed to various reasons, such as imprecise calibration of a quartz crystal monitor, inaccurate knowledge of high and low refractive indices, instability of the layer materials, etc.

In order to estimate closeness between model and experimental data we introduce a discrepancy function:

$$DF^2 = \frac{1}{L} \sum_{j=1}^{L} \left[ R(X; \lambda_j) - \hat{R}(\lambda_j) \right]^2,$$

where $X$ is the vector of model parameters describing the considered coating. Here and in what follows, we shall refer to “theoretical spectral curves” as to spectral curves of the initial design and to “model spectral curves” as to curves of the model design. In other words, theoretical and model can be called “unfitted” and “fitted” spectral characteristics. We perform post-production characterization of the HR400-BK7 sample on the basis of an analysis of experimental reflectance data taken in the spectral range [300;1100] nm on the wavelength grid $\{\lambda_j\}$, $j = 1, \ldots, L$, $L = 537$. If we know refractive indices, $n_H(\lambda)$ and $n_L(\lambda)$, with high accuracy and assuming also that the layers are homogeneous, in our RE algorithm we minimize function Eq. (1) and search for relative errors in layer thicknesses only. In this case:

$$R(X, \lambda) = R((1+\delta_1) d_1, \ldots, (1+\delta_N) d_N; n_H(\lambda), n_L(\lambda); \lambda),$$

where $\delta_1, \ldots, \delta_N$ are relative errors in layer thicknesses, $N$ is the number of layers.

In Fig. 1(b) we demonstrate the achieved excellent fit of the experimental data by model data. The estimated errors are shown in Fig. 2.
At the same time the correspondence between oblique-incidence experimental and model reflectance data, presented in Fig. 3, is not good. In Fig. 3 one can observe noticeable deviations between model and measured data indicating unreliability of RE results. There are at least two reasons for this unreliability. The first reason is that input information is not sufficient for post-production characterization of the considered coating because the most informative ripples at the left side of high-reflection zone are not included to the measurements. The second reason is insufficiency of the applied coating model. More accurate model should include variations of the refractive indices, inhomogeneities of layers, etc. Further attempts of the RE of the sample are not possible because we cannot provide sufficient input experimental information (for example, multiscan in situ data or ex situ data in the UV spectral range). We show this example only as an illustration of unreliable RE approach.

### 3. Reverse engineering of an edge filter

The second problem is the RE of an edge filter working at the AOI of 45°. The filter should provide the transmittance more than 93.5% in the spectral range from 670 nm to 710 nm, and reflectance more than 99% in the range from 585 nm to 643 nm. Substrate back side reflections are taken into account. Incidence light is unpolarized. In the design process TiO₂ as high-index material and SiO₂ as low index material were used. Substrate was Suprasil of 1 mm thickness. Refractive indices \( n_p(\lambda) \), \( n_s(\lambda) \) of the materials, and substrate refractive index \( n_s(\lambda) \) are presented in Table 1. Extinction coefficient \( k(\lambda) \) of TiO₂ is estimated as \( k(400) = 0.0036 \), \( k(450) = 0.0015 \), \( k(500) = 0.0006 \), \( k(550) = 0.0003 \), \( k(600) = 0.0001 \).
$k = 0$ for $\lambda > 600$ nm with piece-wise linear interpolation to intermediate $\lambda$. In [8] a good solution to this design problem was found using OptiLayer thin film software [9]. This solution is a 28-layer design with total physical thickness of 4032.5 nm. The design structure is presented in Table 2.

Table 2. Design structures considered in Sections 3–5, layer numbers LN starts from the substrate, physical thicknesses and layer materials are listed.

| Edge filter | Beam splitter | AR coating |
|-------------|---------------|------------|
| LN | Th. | Mat. | LN | Th. | Mat. | LN | Th. | Mat. |
| 1 | 189.02 TiO$_2$ | 1 | 114.27 Nb$_2$O$_5$ | 29 | 99.59 Nb$_2$O$_5$ | 1 | 241.76 SiO$_2$ |
| 2 | 314.64 SiO$_2$ | 2 | 185.02 SiO$_2$ | 30 | 179.48 SiO$_2$ | 2 | 170.17 Nb$_2$O$_5$ |
| 3 | 74.08 TiO$_2$ | 3 | 105.92 Nb$_2$O$_5$ | 31 | 112.33 Nb$_2$O$_5$ | 3 | 339.81 SiO$_2$ |
| 4 | 139.71 SiO$_2$ | 4 | 126.83 SiO$_2$ | 32 | 187.63 SiO$_2$ | 4 | 117.87 Nb$_2$O$_5$ |
| 5 | 188.73 TiO$_2$ | 5 | 44.84 Nb$_2$O$_5$ | 33 | 113.97 Nb$_2$O$_5$ | 5 | 140.52 SiO$_2$ |
| 6 | 324.15 SiO$_2$ | 6 | 118.57 SiO$_2$ | 34 | 187.54 SiO$_2$ | 6 | 68.74 Nb$_2$O$_5$ |
| 7 | 78.93 TiO$_2$ | 7 | 103.91 Nb$_2$O$_5$ | 35 | 112.26 Nb$_2$O$_5$ | 7 | 117.58 SiO$_2$ |
| 8 | 134.9 SiO$_2$ | 8 | 184.17 SiO$_2$ | 36 | 177.45 SiO$_2$ | 8 | 84.53 Nb$_2$O$_5$ |
| 9 | 87.48 TiO$_2$ | 9 | 114.71 Nb$_2$O$_5$ | 37 | 92.98 Nb$_2$O$_5$ | 9 | 197.91 SiO$_2$ |
| 10 | 270.27 SiO$_2$ | 10 | 189.85 SiO$_2$ | 38 | 87.27 SiO$_2$ | 10 | 22.22 Nb$_2$O$_5$ |
| 11 | 92.3 TiO$_2$ | 11 | 114.57 Nb$_2$O$_5$ | 39 | 56.45 Nb$_2$O$_5$ |
| 12 | 144.39 SiO$_2$ | 12 | 185.51 SiO$_2$ | 40 | 153.63 SiO$_2$ |
| 13 | 172.38 TiO$_2$ | 13 | 107.92 Nb$_2$O$_5$ | 41 | 108.68 Nb$_2$O$_5$ |
| 14 | 139.39 SiO$_2$ | 14 | 152.95 SiO$_2$ | 42 | 185.52 SiO$_2$ |
| 15 | 76.85 TiO$_2$ | 15 | 59.12 Nb$_2$O$_5$ | 43 | 113.88 Nb$_2$O$_5$ |
| 16 | 154.46 SiO$_2$ | 16 | 89.86 SiO$_2$ | 44 | 186.96 SiO$_2$ |
| 17 | 152.11 TiO$_2$ | 17 | 174.97 SiO$_2$ | 45 | 111.19 Nb$_2$O$_5$ |
| 18 | 156.86 SiO$_2$ | 18 | 174.97 SiO$_2$ | 46 | 170.4 SiO$_2$ |
| 19 | 76.44 TiO$_2$ | 19 | 111.43 Nb$_2$O$_5$ | 47 | 78.36 Nb$_2$O$_5$ |
| 20 | 149.57 SiO$_2$ | 20 | 186.86 SiO$_2$ | 48 | 399.82 SiO$_2$ |
| 21 | 163.40 TiO$_2$ | 21 | 113.71 Nb$_2$O$_5$ | 49 | 95.56 Nb$_2$O$_5$ |
| 22 | 155.08 SiO$_2$ | 22 | 187.52 SiO$_2$ | 50 | 176.15 SiO$_2$ |
| 23 | 70.33 TiO$_2$ | 23 | 112.4 Nb$_2$O$_5$ | 51 | 110.07 Nb$_2$O$_5$ |
| 24 | 122.78 SiO$_2$ | 24 | 180.15 SiO$_2$ | 52 | 118.71 SiO$_2$ |
| 25 | 33.82 TiO$_2$ | 25 | 100.79 Nb$_2$O$_5$ |
| 26 | 102.97 SiO$_2$ | 26 | 123.45 SiO$_2$ |
| 27 | 58.05 TiO$_2$ | 27 | 51.56 Nb$_2$O$_5$ |
| 28 | 209.38 SiO$_2$ | 28 | 118.92 SiO$_2$ |

The coating was produced using e-beam evaporation with ion-assistance at Leybold SYRUSPro 1100 deposition plant equipped with APS Pro. For controlling layer thicknesses broad band monitoring system (BBM) developed at the Laser Zentrum Hannover was used [10–12]. The produced sample is named as EF-Suprasil. In Fig. 4(a) theoretical and experimental near-normal incidence transmittance data of EF-Suprasil sample are compared.
We perform RE on the basis of analysis of transmittance in situ spectra $\hat{T}^{(i)}(\lambda_j)$ recorded after deposition of each layer $i = 1, ..., N$. These data were acquired using BBM device at the spectral range $[400;1000]$ nm on the wavelength grid $\{\lambda_j\}$, $j = 1, ..., L, L = 1354$.

Denote as $T(X_i; \lambda_j)$ the model transmittance data for the first $i$ deposited coating layers, $X_i$ is the vector of parameters describing considered coating. In our RE approach we utilize all transmittance scans simultaneously and use the triangular algorithm for layer thicknesses determination [5]. In order to estimate closeness between model and experimental data, we introduce a multiscan discrepancy function:

$$MDF^2 = \frac{1}{NL} \sum_{i=1}^{N} \sum_{j=1}^{L} \left[ T^{(i)}(X_i; \lambda_j) - \hat{T}^{(i)}(\lambda_j) \right]^2. \quad (3)$$

It is known from previous multiple experiments, that slight variations of optical constants of TiO$_2$ and SiO$_2$ materials are possible in the course of the deposition process [13]. Hence, in our RE algorithm we assume that refractive indices can be shifted with respect to the nominal refractive indices used in the theoretical design. We introduce two unknown parameters, namely, refractive index offsets $h_H$ and $h_L$:

$$T(X_i; \lambda) = T(d_1, ..., d_j; n_H(\lambda)(1+h_H), n_L(\lambda)(1+h_L); \lambda). \quad (4)$$

After minimization of the $MDF$ function with respect to $h_H$ and $h_L$ parameters, we obtain corrected refractive indices $\bar{n}_H = n_H(1+h_H)$ and $\bar{n}_L = n_L(1+h_L)$, where $h_H = -0.0112\%$, $h_L = 0.0207\%$. As BBM provides self-compensation mechanism [14–16], then relatively large errors in layer thicknesses are possible. Hence, at the second step of the RE algorithm we assume non-correlated random errors in layer thicknesses and present them in the form:

$$T(X_i; \lambda) = T(d_1(1+\delta_1), ..., d_j(1+\delta_j); \bar{n}_H(\lambda), \bar{n}_L(\lambda); \lambda). \quad (5)$$

Determined relative errors in layer thicknesses are shown in Fig. 5. It is seen from Fig. 5(a) that the largest error of about 5.5% is found in the thinnest layer with number 25. Compensation of this error is observed in the subsequent three last layers 26-28. Obtained fitting of measured normal incidence transmittance values by model data is shown in Fig. 4(b).

![Fig. 5. Estimated relative errors in layer thicknesses of the edge filter coating (a). Comparison of oblique incidence experimental transmittance data related to EF-Suprasil sample with model transmittance (b).](image-url)
As design requirements are specified at \( \text{AOI} = 45^\circ \), the correspondence between experimental and model oblique incidence characteristics must be checked. Perkin Elmer Lambda 950 allows one to measure transmittances \( \tilde{T}^{(s)}(\lambda_j) \) and \( \tilde{T}^{(p)}(\lambda_j) \) for \( s \)- and \( p \)-polarized light. In Fig. 5(b) we compare transmittance, calculated for model coating found as a result of applied RE algorithm, and experimental oblique incidence transmittance data \( \tilde{T}(\lambda_j) \) calculated as \( \left( \tilde{T}^{(s)}(\lambda_j) + \tilde{T}^{(p)}(\lambda_j) \right) / 2 \). In Fig. 5(b) one can observe a good correspondence between measured and model transmittance data. This correspondence confirms the reliability of our RE results and, more general, validates our RE approach. It can be checked that transmittance values in the spectral range from 670 to 710 nm are greater than 93.5\%, reflectance values in the range \([585; 643]\) nm, calculated as \( \tilde{R}(\lambda_j) = 100\% - \tilde{T}(\lambda_j) \), are more than 99\%. For the next oblique-incidence coatings, it is not necessary, therefore, to perform oblique-incidence measurements.

4. Reverse engineering of a beam splitter

The third task is post-production characterization of a specialized beam splitter working at the AOI of 45\(^\circ\). The target specifications are the following. Target transmittance of the sample is more than 98\% in the range from 935 nm to 945 nm, target reflectance is more than 98\% in the range from 967 nm to 971 nm. Substrate is Suprasil of 1 mm thickness. In the design process we used \( \text{Nb}_2\text{O}_3 \) as high index material and \( \text{SiO}_2 \) as low index material. Nominal refractive indices of these materials and substrate refractive index are presented in Table 1. The solution to this problem is a 52-layer design for the substrate front side and 10-layer AR coating for the substrate back side [9]. Structures of both designs are presented in Table 2.

The experimental samples were produced with magnetron-sputtering plant (Helios, Leybold Optics GmbH). The plant was equipped with a BBM system [10–12]. As the sample is to be coated from its both sides, two deposition runs were performed.

In the first deposition run, we deposited the 52-layer coating at two samples. We placed two substrates, namely, Suprasil of 1 mm thickness and Glass B260 substrate of 1 mm thickness exactly at the same distance from the rotation center of turntable. At these positions two corresponding samples denoted as BS-Suprasil and BS-Glass were produced. The sample BS-Glass was placed on the BBM measurement position.

In the second run we deposited the AR coating at two samples. We turned up the BS-Suprasil sample and deposited 10-layer AR coating on its back side. We refer to this sample as BS-AR-Suprasil. Also, a new uncoated B260 Glass substrate was placed on the position where the BBM measured transmittance and the sample, denoted as AR-Glass of AR coating on B260 Glass substrate, was produced. As a result of two deposition runs we had three samples: BS-AR-Suprasil (Suprasil substrate coated from its front and back sides), BS-Glass and AR-Glass samples (B260 Glass substrates coated with 52-layer beam splitter and 10-layer AR, respectively). For the latter two samples \textit{in situ} transmittance data were recorded. In the course of both depositions, the BBM device worked in a passive mode for data acquisition only. Layer thicknesses were controlled using well-calibrated time monitoring [17,18].

We perform our RE process on the basis of analysis of BBM measurements. The nominal refractive indices (see Table 1) are known quite reliably because they had typically been used in the course of the deposition of complicated dispersive mirrors which are very sensitive to refractive index variations [17–20]. For this reason, we apply RE algorithm assuming random errors in layer thicknesses only. This algorithm is similar to one described in Section 3. As in this case we used accurate time monitoring, errors in layer thicknesses \( \delta \) are expected to be small [21]. We consider generalized discrepancy function which is obtained by adding Tikhonov’s stabilizing functional [22] to the multiscan discrepancy function defined by Eq. (3):
\[ GMDF^2 = \frac{1}{NL} \sum_{i=1}^{N} \sum_{j=1}^{L} \left[ T^{(i)}(X_i; \lambda_j) - \hat{T}^{(i)}(\lambda_j) \right]^2 + \alpha \frac{1}{N} \sum_{i=1}^{N} \delta_i^2, \tag{6} \]

where \( \alpha \) is a regularization parameter that is selected to provide a compromise between data fitting and stability of the solution. The parameter \( \alpha \) was taken equal to 1. In Eq. (6) \( L = 1356, \lambda_i = 400, \lambda_z = 1000 \), the model transmittance \( T^{(i)}(X_i; \lambda_j) \) is:

\[ T^{(i)}(X_i; \lambda) = T^{(i)}(d_i(1+\delta), \cdots, d_i(1+\delta); n_i(\lambda), n_i(\lambda); \lambda). \tag{7} \]

Fig. 6. Fittings of \textit{in situ} experimental transmittance data (red crosses) by model transmittances (black solid curves) related to sample BS-Glass after the deposition of layer 13, 26, 39, and 52.

Fig. 7. Fittings of normal incidence experimental transmittance data by model transmittance related to the sample AR-Glass (a). Estimated relative errors in layer thicknesses of the samples BS-Glass and AR-Glass (b).
We perform separate RE processes of produced BS-Glass and AR-Glass samples because BBM data correspond to them. Achieved fitting of experimental in situ transmittance data related to BS-Glass sample by model data is shown in Fig. 6. Final fitting of normal incidence ex situ transmittance data related to AR-Glass sample by model transmittance is presented in Fig. 7(a). Estimated errors in layer thicknesses of BS-Glass and AR-Glass coatings are shown in Fig. 7(b). It is seen that the errors do not exceed 2–3% that is in correspondence with estimated accuracy of time monitoring.

For the sample BS-AR-Suprasil, multangle spectral photometric measurements were performed using an advanced automated double beam UV-VIS-NIR multi-angle spectrophotometer, the Cary 7000 Universal Measurement Spectrophotometer (UMS) from Agilent Technologies [23]. In Fig. 8(a) we compare 45 degree transmittance data related to BS-AR-Suprasil sample and transmittance calculated for Suprasil substrate coated with obtained model beam splitter and model AR designs. Experimental transmittance data was calculated as $\left(\tilde{T}_s(\lambda_i) + \tilde{T}_p(\lambda_i)\right)/2$. For additional validation of the obtained results we also compare oblique incidence experimental and model transmittance data for both s- and p-polarizations, AOI = 30° [Fig. 8(b)]. Good correspondence between experimental and model data observed in both Figs. 8(a) and 8(b) validates our RE results. We conclude, therefore, that for further oblique incidence coatings of this type, the RE algorithm described in this section can be applied without additional verification of the results.

![Fig. 8. Comparison of oblique incidence experimental transmittance data of the sample BS-AR-Glass with model transmittance: (a) non-polarized light at 45°, (b) s- and p-polarizations at 30°.](image)

5. Reverse engineering of a 43-layer quarter-wave mirror

In the fourth RE problem we characterize a 43-layer quarter-wave mirror for the central wavelength of 800 nm. AOI is 45°, light is unpolarized. Odd layers are HfO$_2$ layers and even layers are SiO$_2$ layers. Corresponding nominal refractive indices are presented in Table 1 (rows 8 and 9). Physical thicknesses of high and low index layers are 114.56 nm and 156.58 nm, respectively. The experimental samples were produced with e-beam evaporation using SYRUSPro 710 (Leybold Optics GmbH). In the course of the deposition process the substrate temperature was 120°C. Layer thicknesses were controlled with the help of a quartz crystal monitor. SYRUSPro 710 is also equipped with BBM device [10–12]. This device was used for data acquisition and for on-line recalibration of the quartz crystal monitor as well.

We produced two samples of the 43-layer quarter-wave mirror. Fused Silica substrate of 6.35 mm thickness was placed on one of the calotte positions, and B260 Glass substrate of 1 mm thickness was placed on the position where BBM performed transmittance measurements. Two deposited samples are denoted as HR800-FusedSilica and HR800-Glass. In Fig. 9(a) we compare experimental in situ transmittance data measured by BBM device,
experimental \textit{ex situ} transmittance data taken by Cary 7000 UMS and theoretical transmittance data related to HR800-Glass sample. In Fig. 9(a) one can observe quite large deviations between all three transmittance curves. The main deviations are shifts of the experimental curves to the shorter wavelengths with respect to theoretical curve, and differences in widths of the high-reflection zones in measured and theoretical data. The shifts to the shorter wavelengths are explained by underestimation of layer optical thicknesses. This underestimation can be connected with underestimating layer geometrical thicknesses and/or underestimation of nominal refractive indices of layer materials. Underestimation of layer geometrical thicknesses can be attributed to inaccurate calibration of the quartz crystal monitor.

\begin{figure}
\centering
\includegraphics[width=\textwidth]{fig9.png}
\caption{Comparison of \textit{in situ} experimental transmittance data, \textit{ex situ} experimental data and theoretical transmittance related to HR800-Glass sample (a). Fitting of \textit{in situ} experimental transmittance data by model transmittances related to HR800-Glass sample (b).}
\end{figure}

The nominal refractive indices of HfO$_2$ were used only as starting assumption, before we did not have reliable refractive index for the case of deposition at low substrate temperature of 120°C. It is known [24] that the width of high-reflection zone of quarter-wave mirror is dependent on the ratio of high and low refractive indices. Theoretical width of high-reflection zone is about 126 nm, width of high-reflection zone according to \textit{in situ} data is 133 nm and the width of high-reflection zone according to \textit{ex situ} measurements is 143 nm. If we neglect small effect of porosity of SiO$_2$ and assume that SiO$_2$ index is quite stable, then we should expect that refractive index of HfO$_2$ layers in vacuum is larger than the assumed nominal refractive index. Also, the refractive index of HfO$_2$ layers in the atmosphere is higher than refractive index of HfO$_2$ in vacuum. The latter statement can be explained by porous structure of HfO$_2$ layers [25]. After exposition to atmosphere, pores in HfO$_2$ layers are filled with water, and the refractive index of the HfO$_2$ layers increases. This phenomenon is known as a vacuum shift [25,26].

Taking into account the qualitative issues presented above, we propose the following RE algorithm. At the first step of the algorithm we analyze BBM data related to HR800-Glass sample. We choose coating model assuming random errors in layer thicknesses and random offsets of HfO$_2$ refractive indices. The discrepancy function is defined by Eq. (3) with

\begin{equation}
T^{(i)}(X; \lambda) = T^{(i)}(d_1(1+\delta_1),\ldots, d_i(1+\delta_i); n_H(1+h_{H,i}), n_L,\ldots,n_H(1+h_{H,j}); \lambda).
\end{equation}

Here $h_{H,1},\ldots,h_{H,j}$ are offsets of refractive indices of HfO$_2$ layers. In Eq. (3) $L=1370$, $\lambda_1 = 400$ nm, $\lambda_2 = 1000$ nm.

In Fig. 9(b) we present achieved fitting of \textit{in situ} transmittance data by model transmittance data. In Fig. 10(a) we compare average refractive index of HfO$_2$ layers.
calculated as \( \tilde{n}_H = n_H + \sum_{i=1}^{22} \frac{\tilde{h}_{H,i}}{22} \) and their nominal refractive index. As it was expected on the basis of the qualitative analysis, estimated refractive index is higher than nominal refractive index. In Fig. 10(b) estimated relative errors in layer thicknesses are shown. It is seen that all layers, except the first layer, are underdeposited. We call the design defined by the vector \( X = (\tilde{d}_1, \ldots, \tilde{d}_{43}, n_H, n_L) \) with actual thicknesses \( d_i = (1 + \delta_i)d_i, \ldots, d_{43} = (1 + \delta_{43})d_{43} \) as an intermediate design.

![Fig. 10. Refractive indices of HfO\(_2\) layers: nominal, average in situ and ex situ (a). Estimated relative errors in layer thicknesses of 43-layer quarter-wave mirror (b).](image)

In Fig. 10, we compare experimental normal and oblique incidence transmittance data related to HR800-FusedSilica sample with model transmittances calculated for intermediate design. We assume offset of HfO\(_2\) refractive index. We assume also that HfO\(_2\) layers are inhomogeneous, i.e., refractive indices of layers are varying with respect to physical thickness. We search for offset in refractive index of

In Fig. 11 we compare experimental normal and oblique incidence transmittance data related to HR800-FusedSilica sample, and transmittance values, calculated for intermediate design and Fused Silica substrate. In Fig. 11 one can observe that experimental data are shifted to the longer wavelengths with respect to model data. The widths of high-reflection zones of both curves are different which can be explained mainly by differences in refractive indices.

At the second step of the RE process we assume that actual layer thicknesses have been reliably determined at the first step of the RE process. We expect offset of HfO\(_2\) refractive index. We assume also that HfO\(_2\) layers are inhomogeneous, i.e., refractive indices of layers are varying with respect to physical thickness. We search for offset in refractive index of
HfO$_2$ layers $h_i$ and degree of inhomogeneity $\delta_i$ of these layers. Degree of inhomogeneity $\delta$ is defined as

$$\delta = (n^+ - n^-)/n_{av},$$  

where $n^+$ and $n^-$ are refractive indices of a layer at its outer and inner boundaries, and $n_{av}$ is the average layer refractive index $n_{av} = (n^+ + n^-)/2$.

We perform RE on the basis of normal incidence transmittance and reflectance data. The algorithm is based on the minimization of discrepancy function taken in the form:

$$DF^2 = \frac{1}{2L} \sum_{\lambda_j} \left[ T(X; \lambda_j) - \hat{T}(\lambda_j) \right] + \frac{1}{2L} \sum_{\lambda_j} \left[ R(X; \lambda_j) - \hat{R}(\lambda_j) \right],$$  

where $L = 701$, $\lambda_i = 400$ nm, $\lambda_4 = 1500$ nm. In Eq. (10) $T(X; \lambda)$ is

$$T(X; \lambda) = T(\delta_1, \ldots, \delta_k; n_{hi}, n_i, h_i; \lambda).$$  

Corrected refractive index of HfO$_2$ layers is shown in Fig. 10(a) (blue curve). As it was expected, corrected refractive index is higher than nominal refractive index and also higher than refractive index of HfO$_2$ layers in vacuum. The estimated degree of bulk inhomogeneity is about $-8\%$. Such high degree of inhomogeneity may be explained by low deposition temperature.

![Fig. 12. Final fitting of normal (a) and oblique incidence AOI = 45° (b) experimental transmittance data related to HR800-FusedSilica sample by model transmittances.](image)

The final fitting of normal incidence experimental transmittance data by model data is shown in Fig. 12(a). The fitting is not ideal because the applied model is not accurate enough. At the same time, this model takes into account all main features of the deposited coating. More accurate model would assume different degrees of inhomogeneities of each layer as well as possible interlayers caused by interdiffusion of HfO$_2$ and SiO$_2$ materials. Multiple model parameters, however, cannot be found reliably from single ex situ transmittance/reflectance spectra.

For verification of the RE results we compare transmittance data measured at AOI = 45° using Cary 7000 UMS and model transmittance data [Fig. 12(b)]. In Fig. 12(b) one can observe good agreement between experimental and model data. Also, we observed good correspondences for experimental transmittance/reflectance data measured at all incidence angles from 0° to 45° with 5° angular step. Good correspondence between experimental and model oblique incidence data validates our RE approach.
Fig. 13. Comparison of normal incidence experimental data related to the second deposition run and theoretical transmittance data (a). Model reflectance at AOI = 45° after additional RE procedure (b).

We used obtained RE results in order to adjust the deposition process for the next deposition run. Namely, we changed tooling factors of the quartz crystal monitor, specified corrected refractive index of HfO₂ layers in the theoretical design, recalculated corresponding layer thicknesses, and took into account the vacuum shift. In Fig. 13(a) we compare theoretical transmittance values of recalculated 43-layer quarter-wave mirror and transmittance data measured at PerkinElmer Lambda 950. It is seen from Fig. 13(a) that the correspondence between theoretical and experimental data is significantly improved. In order to further improve the deposition process, we performed RE again and calculated model reflectance at 45 degrees [Fig. 13(b)]. We can judge now the performance of our sample on the basis of this data: reflectance in the vicinity of the wavelength 800 nm is larger than 99.95%.

6. Conclusions

We have discussed choices of reliable approaches for the post-production characterization of complicated oblique incidence coatings. Each approach includes the choice of experimental data set, selection of the numerical algorithm, coating model, and verification of the obtained RE results. Since accurate oblique-incidence $R$ and $T$ measurements are more complicated than normal and quasi-normal incidence measurements we propose to lessen the first ones and in the mass-production to rely on the second ones. To achieve this goal we propose to perform careful oblique-incidence measurements for several test coatings only and to use the obtained experimental data for adjusting RE process. The RE process is to be performed on the basis of normal incidence data, oblique incidence measurements are used for verification of the results only. For further oblique incidence coatings produced under the same deposition conditions, the adjusted RE procedure can be used without additional verification. The discussed concept was successfully applied for the post-production characterization of complicated multilayer coatings.

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