Comparison of algorithms used for optical characterization of multilayer optical coatings

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Two algorithms used for the on-line and off-line characterization of multilayer optical coatings are experimentally compared using test samples produced by two different deposition processes and different monitoring approaches. One of these algorithms, called the triangular algorithm, demonstrates its superiority in all considered situations. We performed experiments with multilayer samples formed by high-density thin films, which allowed us to neglect possible errors in the film refractive indices and concentrate only on errors in the thicknesses of the layers of the produced coatings. © 2011 Optical Society of America

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1. Introduction

At the current state of the art in production of optical coatings, the reliable determination of thicknesses of layers of multilayer coatings during and after the manufacturing process is one of the key issues in advancing the quality of optical coating production. Determination of actual thicknesses of layers of manufactured coatings (off-line characterization) is required for adjusting deposition parameters, recalibrating monitoring devices, and improving the control of the thicknesses of individual layers [1–5]. Determination of layer thicknesses in the course of the manufacturing process (on-line characterization) is important in connection with the growing implementation of broadband monitoring (BBM) devices in deposition environments. The first BBM systems have been used since the late 1970s [1,2,6–8], but in recent years, new effective BBM systems have been elaborated and successfully used [5,9–12].

In the case of BBM systems involving reoptimization procedures, reliable values of thicknesses of already-deposited layers are required for reoptimization of the remaining layer thicknesses [1–4,13]. In the case of BBM systems without reoptimization procedures, reliable values of thicknesses of already-deposited layers are also useful, because these values may be required for the implementation of monitoring algorithms.

From a mathematical point of view, the determination of actual layer thicknesses is a typical inverse problem with all the difficulties associated with the solving of inverse problems [14,15]. Mathematical

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algorithms and corresponding software for off-line characterization have been in use for many years [4,16,17]. These algorithms allow for the determination of layer thicknesses from measurement data obtained after a coating deposition. However, if a number of unknown parameters (layer thicknesses) is large, the instability of the inverse problem solution grows, and this may lead to the wrong characterization results.

A basic approach to reduce the instability of the inverse problem solution is to involve more measurement data in a characterization procedure [14]. In the case of optical coating production controlled by BBM systems, measurement data are recorded at multiple intermediate steps of a deposition process. In principle, all these data can be used by a characterization procedure, and, indeed, they have been used for the characterization of special types of rugate coatings [12,18].

For the characterization of conventional multilayer stacks, it was proposed to use measurement data recorded after deposition of each coating layer [3,4,12,15] (measurement scans). Generally speaking, various combinations of measurement scans can be used by a characterization procedure, and various combinations of layer thicknesses can be determined at different steps of this procedure. Thus, potentially a large variety of characterization algorithms exists. But so far, two such algorithms, triangular and sequential, were distinguishingly formulated and presented [3,4,12,15]. In the course of on-line characterization based on the triangular algorithm, all already-recorded measurement scans are used, and thicknesses of all deposited layers are determined. In the case of the sequential algorithm, only the last measurement scan is used, and only the thickness of the last deposited layer is determined, while thicknesses of previously deposited layers are fixed. Both algorithms can be used also in the regime of off-line characterization.

Obviously the triangular algorithm utilizes more experimental information, and one may hope that it provides more reliable and stable results. However, all measurement data contain inevitable random errors (random noise) and systematic errors (offset and drift errors) [19]. These two types of errors influence an accuracy of optical parameters determination [20]. Thus, utilizing more measurement scans leads to involving of more measurement errors in the characterization process, and this may degrade the accuracy of the triangular algorithm.

Currently, there is no rigorous theoretical proof that the triangular algorithm provides more reliable characterization results than the sequential algorithm. The theoretical explanation of this fact is not a straightforward task and requires application of a complicated mathematical apparatus. So, in this study, we concentrate our attention on an experimental comparison of the triangular and sequential algorithms.

In Section 2 we briefly describe both algorithms and the respective characterization procedures. In Section 3 we compare the algorithms based on the off-line characterization of two samples produced using the BBM technique. In Section 4 we apply the algorithms to off-line characterization of test coatings produces by magnetron sputtering with accurately calibrated time monitoring. One of these test coatings contains intentionally introduced thickness errors, which allows further comparison of the algorithm accuracies. In our experiments, we use stable deposition processes that produce high-density films. This allows us to neglect possible errors in the film refractive indices and concentrate our attention only on errors in the thicknesses of the coating layers. The final conclusions of our study are provided in Section 5.

2. Description of the Algorithms

In this paper we consider BBM data with transmittance measurement data that are available on a set of wavelengths denoted as $\{\lambda_j\}, j = 1, \ldots, L$, where L is the number of wavelength points. Denote $\hat{T}^{(i)} = \hat{T}^{(i)}(\lambda_j)$ as a measurement scan acquired after the deposition of a layer number i. The total number of design layers is m and $1 \le i \le m$. Denote $T(d_1, \ldots, d_i; \lambda_j)$ as the model transmittance data for the first i deposited coating layers.

The triangular algorithm at its ith step determines thicknesses of all already-deposited layers $d_1, \ldots, d_i$. The algorithm is based on the minimization of the discrepancy functions [4,15]:

$$\mathrm{DF}(i) = \left(\frac{1}{i}\sum_{k=1}^{i}\frac{1}{L}\sum_{j=1}^{L}\left[\frac{T(d_1, \ldots, d_k; \lambda_j) - \hat{T}^{(k)}(\lambda_j)}{\Delta T_j}\right]^2\right)^{1/2}, \quad 1 \le i \le m, \tag{1}$$

where ΔT_j are measurement tolerances. In computational experiments of the following sections, we exclude from consideration wavelength regions with high errors in transmittance data. Because of this fact, a specification of wavelength-dependent tolerances does not affect basic results related to the discrepancy function minimization. It is convenient to assume all tolerances equal 1%, because in this case, the discrepancy function values represent the mean square deviation of the model transmittance data from measurement data.

In the course of on-line characterization based on the triangular algorithm after deposition of each new coating layer, we obtain a new set of layer thicknesses: $\left\{d_1^{(1)}\right\}, \left\{d_1^{(2)}, d_2^{(2)}\right\}, \ldots, \left\{d_1^{(m)}, \ldots, d_m^{(m)}\right\}$. In the case of off-line characterization, we consider the last set $\{d_1^{(m)}, \ldots, d_m^{(m)}\}$ as the characterization result delivered by the triangular algorithm.

The sequential algorithm at each step determines only the thickness of the last deposited layer d_i. This thickness is determined by minimizing with respect to d_i the discrepancy function [15]:

$$\mathrm{DF}(i) = \left(\frac{1}{L} \sum_{j=1}^{L} \left[\frac{T(\bar{d}_1, ..., \bar{d}_{i-1}, d_i; \lambda_j) - \hat{T}^{(i)}(\lambda_j)}{\Delta T_j} \right]^2 \right)^{1/2}, \quad (2)$$

where $\bar{d}_1, ..., \bar{d}_{i-1}$ are the thicknesses of the previously deposited layers that were determined at the previous steps of the algorithm.

One can see that in the case of the sequential algorithm, the discrepancy functions are minimized with respect to only one variable. Because of this fact, this algorithm usually works faster than the triangular algorithm. This is of course an attractive feature of the sequential algorithm. At the same time, as discussed in Section 3, this algorithm may suffer from the effect of accumulation of thickness errors.

3. Comparison of the Triangular and Sequential Algorithms

In this section, we compare the algorithms using samples produced with broadband optical monitoring control of thicknesses during deposition. Recently it was proved that BBM provides an error self-compensation mechanism in the course of optical coating production [21]. This mechanism reveals itself as follows. Despite a rather large level of errors in the thicknesses of the individual layers, these errors are arranged by the BBM technique so that they partly compensate each other, and the final spectral characteristic of manufactured coating is good enough. Thus for coatings produced with BBM techniques, one can expect noticeable thickness errors, and it is interesting to investigate how these errors are determined by different characterization algorithms.

It was shown in [21] that narrow bandpass filters exhibit a strong error self-compensation mechanism. For our research, we use narrow bandpass filters with the following target specifications. The target transmittance is zero in the spectral ranges from 430 to 520 nm and from 545 to 630 nm and is 100% in the spectral range from 522 to 542 nm, respectively. We designed two filters denoted below as NBPF1 and NBPF2. These filters contain 26 and 25 layers, respectively. In the design process we used titanium dioxide as a high-index material and silicon dioxide as a low-index material. The refractive indices of these materials are specified by the Sellmeier equation:

$$n^2(\lambda) = 1 + \frac{A_1\lambda^2}{\lambda^2 - A_2} + \frac{A_3\lambda^2}{\lambda^2 - A_4}, \quad (3)$$

where $A_1 = 4.017$, $A_2 = 0.0525$, $A_3 = 1.402$, $A_4 = 80$ for TiO_2 and $A_1 = 1.159$, $A_2 = 0.0015$, $A_3 = 0.5$, $A_4 = 61$ for SiO_2; λ in this formula should be expressed in micrometers. Two designs were obtained using OptiLayer software [16].

The structures of the designs NBPF1 and NBPF2 are presented in Table 1. One can see that the designs are structurally different. In the case of the NBPF1 design, nearly all layers are quarter-wave or multiple quarter-wave layers. Two structurally different designs are chosen in order to compare the algorithms for quarter-wave and non-quarter-wave designs. In both cases, all odd layers are high-index layers and all even layers are low-index layers. The theoretical transmittances of the NBPF1 and NBPF2 designs are shown in Figs. 1 and 2 by solid curves in comparison to the measured spectra.

Table 1. Structures of the Designs NBPF1 and NBPF2

Layer Number	Optical Thickness, QWOT at 532 nm	Optical Thickness, QWOT at 532 nm
1	1.00	1.064
2	1.00	1.485
3	1.00	1.066
4	2.00	1.322
5	1.00	3.356
6	1.00	1.289
7	1.00	0.936
8	1.00	1.277
9	1.00	1.108
10	1.00	1.280
11	1.00	0.891
12	4.00	1.306
13	1.00	0.628
14	1.00	2.098
15	1.00	0.936
16	1.00	1.040
17	1.00	0.933
18	1.00	1.009
19	1.00	0.934
20	2.00	1.022
21	1.00	0.937
22	1.00	2.337
23	1.00	0.426
24	1.00	1.239
25	0.239	0.952
26	1.372	

The NBPF1 and NBPF2 filters were produced using the ion assisted deposition process based on a Leybold SYRUSpro 1100 deposition plant equipped with an APSpro. For the monitoring of layer thickness, the BBM system developed at the Laser Zentrum Hannover was used. This BBM system provides a fully automated process control based on

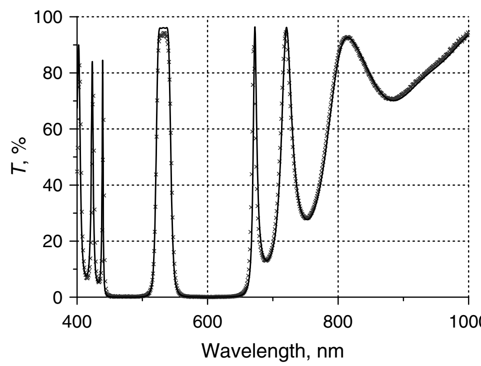

Fig. 1. Comparison of measurement transmittance data of the coating NBPF1 (crosses) and theoretical transmittance of the design NBPF1 (solid curve).

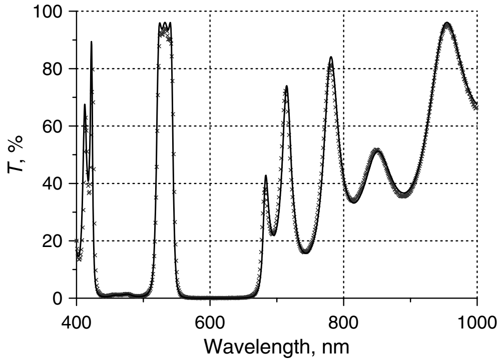

Fig. 2. Comparison of measurement transmittance data of the coating NBPF2 (crosses) and theoretical transmittance of the design NBPF2 (solid curve).

in situ broadband transmittance measurements on moving substrates [9]. Transmittance scans after deposition of each layer were recorded in the spectral range from 360 to 1080 nm.

Figures 1 and 2 compare transmittance spectra of the produced samples with theoretical transmittances of the NBPF1 and NBPF2 designs. The most noticeable deviations between measured and theoretical transmittance data are observed in the high transmission zones. These deviations may be attributed to errors in layer thicknesses.

To determine errors in layer thicknesses with the help of the triangular and sequential algorithms, we applied the OptiRE module of OptiLayer software [16]. For both algorithms, the discrepancy functions [Eqs. (1) and (2)] were calculated on the wavelength grid in the spectral range from 400 to 1000 nm containing 1354 wavelength points.

Relative thickness errors found by the triangular and sequential algorithms in the case of the NBPF1 filter are shown in Fig. 3. It is clearly seen from Fig. 3 that errors found by the algorithms are completely different. Unfortunately we do not have true error data in order to compare accuracies of the triangular and sequential algorithms in a direct way. For this reason, in this section we provide some indirect reasonings indicating that the triangular algorithm results are more reliable. A direct proof of this fact is provided in Section 4. First of all, consider the general patterns of errors found by the triangular and sequential algorithms. In the case of the sequential algorithm (see Fig. 3), an error pattern typical for a cumulative effect of thickness errors is clearly observed. This effect was investigated in [21] for the broadband optical monitoring with layer thicknesses determined by the algorithm that may be considered as a version of the sequential algorithm. In the case of optical characterization based on the sequential algorithm, all considerations of [21] are also applicable, and thus one should expect that cumulative effect leads to a wrong determination of thickness errors at least for the last filter layers. In the case of the triangular algorithm, thicknesses of previously deposited layers are redetermined, and this may prevent a growth of thickness errors.

In Fig. 4 we compare discrepancy function values corresponding to the triangular and sequential algorithms at all steps of the characterization procedures. One can see that in the case of the triangular algorithm discrepancy function, values are increasing from layer to layer much slower than in the case of the sequential algorithm. This is again an indirect evidence of better accuracy of the triangular algorithm.

To provide another evidence of better reliability of the triangular algorithm, we performed an analysis of the model measurement data. These data were generated for the filter NBPF1 in the following way. We took the design without errors in layer thicknesses and specified errors only in the model transmittance scans: random noise of the 1% level was added to each scan, and then the measurement scans were shifted as a whole for constant values ΔT_i, $i = 1, \ldots, m$. The values ΔT_i were normally distributed with a standard deviation of 0.5%.

Errors in the layer thicknesses of the model sample found by the triangular and sequential algorithms are shown in Fig. 5. In the case of the sequential algorithm, the errors are three times

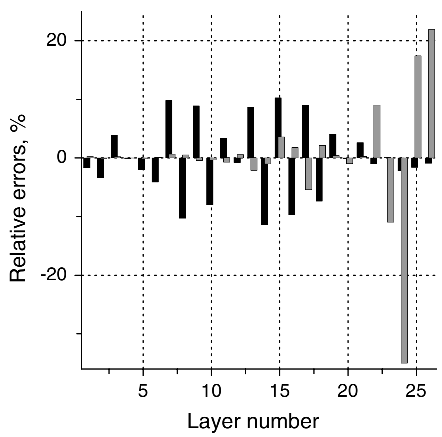

Fig. 3. Comparison of relative errors in layer thicknesses in the coating NBPF1 obtained in the course of triangular algorithm (black bars) and sequential algorithm (gray bars).

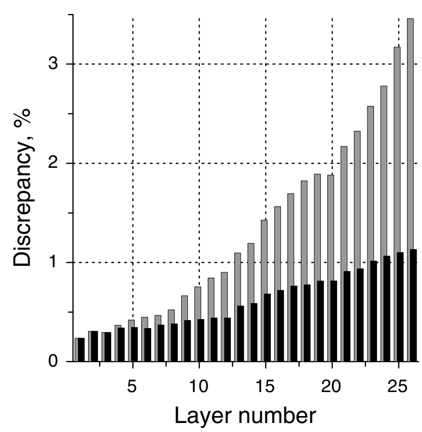

Fig. 4. Comparison of discrepancy function values of the coating NBPF1 obtained in the course of triangular algorithm (black bars) and sequential algorithm (gray bars).

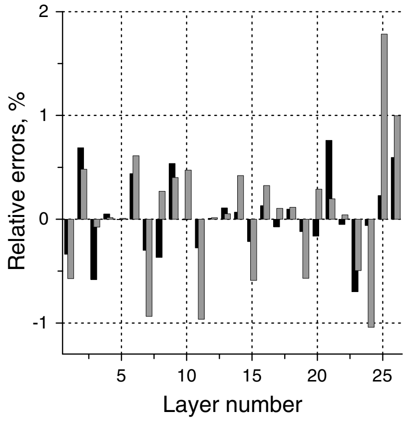

Fig. 5. Comparison of relative errors in layer thicknesses in the coating NBPF1 found by the triangular algorithm (black bars) and sequential algorithm (gray bars) in the case of model data (see the text for details).

higher than in the case of the triangular algorithm: the rms levels of these errors are 0.37% and 0.12%, respectively.

Relative thickness errors found by the triangular and sequential algorithms in the case of the NBPF2 filter (non-quarter-wave design) are shown in Fig. 6. As in the case of NBPF1, the errors found by these algorithms are completely different. In general, errors in layer thicknesses are lower compared to the quarter-wave filter design, but as before in the case of the sequential algorithm, an error pattern typical for the cumulative effect of thickness errors is observed (see Fig. 6). Along with narrow bandpass filters, we performed analogous studies for coatings of different types: mirrors, filters, and polarizers. In all cases, we obtained basically the same results testifying to a better accuracy of the triangular algorithm. However, all these results can be considered only as an indirect confirmation of the statement that the triangular algorithm provides more reliable characterization data. In Section 4 we consider special experiments performed for proving this statement.

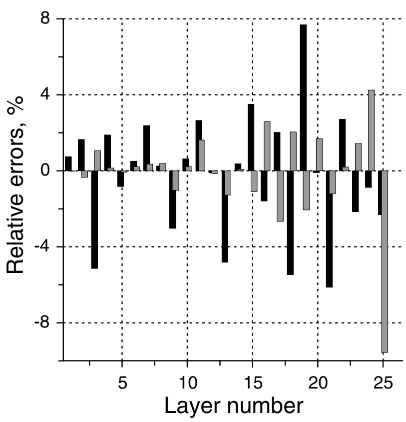

Fig. 6. Comparison of relative errors in layer thicknesses in the coating NBPF2 obtained in the course of the triangular algorithm (black bars) and sequential algorithm (gray bars).

4. Special Experiments Aimed at the Verification of the Algorithms

To check the reliability of thickness error determination by the triangular and sequential algorithms, we performed experiments with special test samples produced in the Leybold Optics magnetron-sputtering Helios plant using time monitoring of the layer thicknesses [22]. In this plant, high-quality dispersive mirrors with many dozens of layers were successfully deposited [22–25], and based on these experiments we estimate that the level of relative thickness errors does not exceed 1%[24,25].

In this section, we consider two test samples of the 15-layer quarter-wave mirrors (QWMs) with a central wavelength of 600 nm. For both samples, the first layer is of the high-index material. We used tantalum pentoxide as a high-index material and silicon dioxide as a low-index material. The refractive indices of these materials were specified by the Cauchy formula:

$$n(\lambda) = A_0 + \frac{A_1}{\lambda^2} + \frac{A_2}{\lambda^4}, \quad (4)$$

where $A_0 = 2.06572$, $A_1 = 0.016830$, $A_2 = 0.001686$ for Ta_2O_5 and $A_0 = 1.46529$, $A_1 = 0$, $A_2 = 0.0004708$ for SiO_2, λ in this formula should be expressed in micrometers. Deposition experiments discussed in Section 3 and Section 4 were done in two different labs where, for historical reasons, different formulas for representing the SiO_2 refractive index wavelength dependencies are used. In the considered spectral range there is no obvious preference in using any of these formulas (Cauchy or Sellmeier). So we keep in the text the original representations.

The first considered sample, QWM-1, is an ideal QWM, in which all errors in layer thicknesses are connected only with monitoring inaccuracies. During the deposition of the second sample, QWM-2, intentional relative thicknesses equal to +5%, −7%, −5%, and +5% were introduced to the 3rd, 8th, 14th, and 15th layers, respectively. These errors are several times higher than the estimated level of errors associated with the monitoring procedure, and we expect that characterization algorithms will be able to detect these intentional thickness errors.

During the depositions of both samples, broadband transmittance scans were recorded after the deposition of each layer with the help of BBM system developed at the Laser Zentrum Hannover [9]. Terminations of the layer deposition were done by well-calibrated time monitoring, and the BBM system was working in a passive mode for data acquisition only.

Relative errors in the layer thicknesses of the QWM-1 sample found by the triangular and sequential algorithms are shown in Fig. 7. In the case of the triangular algorithm, relative errors in layer thicknesses typically do not exceed 1%, which corresponds to the estimated accuracy of layer thickness monitoring. In the case of the sequential algorithm,

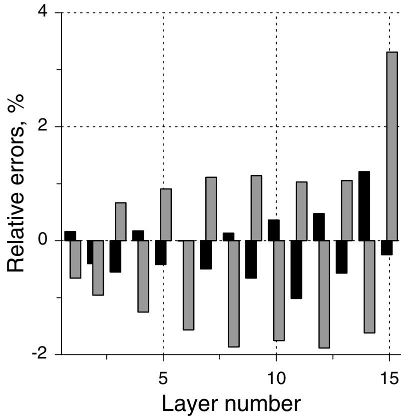

Fig. 7. Comparison of relative errors in layer thicknesses in the coating QWM-1 obtained in the course of the triangular algorithm (black bars) and sequential algorithm (gray bars).

errors are noticeably higher; in particular, the relative thickness error in the last layer equals 3.3%. This is an obvious evidence of an insufficient accuracy of the sequential algorithm.

Further comparison of the triangular and sequential algorithm is provided by the analysis of results obtained for the QWM-2 sample. Relative thickness errors found by the triangular and sequential algorithms are presented in Fig. 8. Errors determined by the triangular algorithm (black bars) are in a good agreement with the intended errors in thicknesses of the layers numbered 3, 8, 14, and 15 (empty bars). The differences between the determined and intended errors do not exceed the estimated accuracy of thickness monitoring.

Thickness errors determined by the sequential algorithm differ from the intended values of errors essentially. For the 8th and 15th layers, the differences between the determined and intended errors exceed 3%, which is much higher than the estimated accuracy of thickness monitoring. One can also see that for many QWM-2 layers that were deposited without intended errors, the sequential algorithm indicates a presence of thickness errors that exceed significantly the estimated level of errors associated with our monitoring procedure. These results show that the sequential algorithm determines thickness errors less accurately than the triangular algorithm.

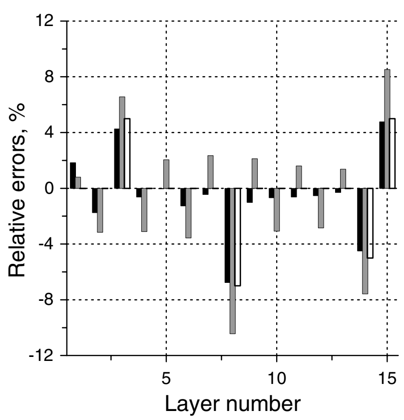

Fig. 8. Comparison of relative errors in layer thicknesses in the coating QWM-2 obtained in the course of the triangular algorithm (black bars), sequential algorithm (gray bars), and expected errors (empty bars).

5. Conclusions

In this study, two previously proposed algorithms for on-line and off-line characterization of multilayer optical coatings (the triangular and sequential algorithms) were compared based on the analysis of experimental data obtained for test samples using different deposition and monitoring techniques. It was shown that the triangular algorithm provides more reliable information about errors in layer thicknesses than the sequential algorithm. The considered algorithms are only two from a large variety of algorithms that can be potentially used for on-line and off-line characterization of multilayer optical coatings. Further theoretical studies are required for estimating potential accuracies of these algorithms. Elaboration of new reliable characterization algorithms may turn out to be a key issue in improving the quality of optical coating production with broadband optical monitoring.

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