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## СТАБИЛИЗАЦИЯ ВЫЧИСЛИТЕЛЬНЫХ АЛГОРИТМОВ ОПРЕДЕЛЕНИЯ СВОЙСТВ ТОНКОПЛЕНОЧНЫХ ОПТИЧЕСКИХ ПОКРЫТИЙ

## А.В. Тихонравов $^1$ , М.К. Трубецков $^1$

Разработка устойчивых вычислительных алгоритмов для определения свойств тонкопленочных оптических покрытий играет ключевую роль для успешного их применения во многих передовых областях технологии. Настоящая статья представляет общую идею, позволяющую разрабатывать эффективные с вычислительной точки зрения и устойчивые алгоритмы определения свойств покрытий для практически всех современных производственных процессов, используемых для изготовления оптических покрытий. Эффективность одного из таких алгоритмов демонстрируется с использованием новой исследовательской методологии, названной вычислительным производством.

Ключевые слова: многослойные покрытия, разработка тонкопленочных оптических покрытий, вычислительное производство, тонкие пленки, стабилизация.

1. Introduction. Computational methods play an increasingly important role in thin film optics, which is a research area with many technological applications. Thin film optical coatings are core elements of nearly all modern opto-electronic devices [1] and the demand for various types of these elements is permanently growing. Production of high-quality optical coatings for modern challenging applications requires the computational design of coatings with complicated structures and the successful practical implementation of the theoretical designs obtained.

Computational design of optical coatings is a complex multiextremal optimization problem. In recent years, a tremendous progress has been observed in the development of optimization techniques for solving this problem [2-5]. As a result, the main attention of researchers is now switched to the successful manufacturing of coatings with complicated design structures. Optical coatings are usually produced in special vacuum chambers by successive depositions of thin films of various dielectric materials [6]. These films form the layers of an optical coating. Thicknesses of coating layers are specified by a theoretical design of this coating and their reliable control is one of the main problems for all coating deposition techniques. A successful control of layer thicknesses requires accurate monitoring of their values in time.

There are two major approaches to monitoring the thicknesses of coating layers during their depositions. The first one is called quartz crystal monitoring. This approach enables measuring the deposition rates of thin film materials in time. The deposition rate is defined as a rate of increase of thin film thickness. It is usually measured in angstroms (Å) per second or in nanometers (nm) per second. Recall that Å is  $10^{-10}$  m, while nm is  $10^{-9}$  m. Typical values of deposition rates vary from several Å/sec to several nm/sec, depending on a deposition process. When quartz crystal monitoring is used, layer thicknesses are monitored by integrating the measured deposition rates.

Another approach to thickness monitoring is called optical monitoring. In fact, this is not a single approach but a wide set of different approaches whose common feature is measuring of optical response from a deposited coating inside a deposition chamber. Usually, reflectance R or transmittance T of a deposited coating is measured either at a single monitoring wavelength  $\lambda_0$  (single wavelength optical monitoring) or at a set of wavelengths in a broad spectral band (broadband optical monitoring). Thicknesses of deposited layers are determined on the basis of R or T measurement data each time when measurements are performed. Obviously, this determination is a typical inverse recognition problem.

Currently, there exists a large variety of different optical monitoring schemes and an even larger variety of different strategies for optical data acquisition. Respectively, there is a large variety of computational algorithms for solving the above mentioned inverse recognition problem. The general term "optical characterization" is applied to the process of determination of optical coating parameters with the aid of optical measurement data [1]. In the following, we shall often use the term "characterization algorithm" to specify an algorithm for determining optical coating parameters, in particular, coating layer thicknesses.

<sup>&</sup>lt;sup>1</sup> Научно-исследовательский вычислительный центр, Московский государственный университет им. М. В. Ломоносова, 119992, Москва; e-mail: tikh@srcc.msu.su, trub@srcc.msu.su

<sup>©</sup> Научно-исследовательский вычислительный центр МГУ им. М. В. Ломоносова

A control of optical coating manufacturing is based on results provided by a characterization algorithm used to process on-line monitoring data. Deposition of a coating layer is terminated when, according to these results, the layer thickness reaches a value prescribed by a theoretical coating design. Errors in characterization results cause errors in the layer thicknesses of a manufactured optical coating. As a result, its properties differ from the properties predicted by a theoretical coating design.

Errors in layer thicknesses are the main reason for the degradation of optical properties of a manufactured optical coating. Some coatings are especially sensitive to these errors. Complicated coatings for modern applications may have many dozens or sometimes hundreds of layers and even small relative errors in thicknesses of their layers may be fatal for these coatings. In this respect, high accuracy of on-line characterization algorithms is extremely important.

Quite often, inaccurate on-line characterization results are connected with the practical instability of the online characterization algorithm being used. We employ the term "practical instability" to designate the situations when errors in layer thicknesses become many times higher than their average value and when the successive on-line determinations of a growing layer thickness give entirely different or physically absurd results. The main goal of this paper is to present a simple, yet general idea for avoiding the practical instability of characterization algorithms and, accordingly, for increasing the accuracy of on-line characterization results. This idea and its practical implementation are discussed in Section 3 of this paper.

Unfortunately, it is very difficult to provide a detailed practical testing of any new computational algorithm for the on-line characterization of optical coatings. First of all, it is too time-consuming, because a single run of a deposition chamber for a manufacturing experiment may take many hours. Secondly, it is too expensive, because this run may cost many thousand dollars. In connection with this fact, a special significance has a new research area that we have named computational manufacturing [7]. Investigations in this area were started several years ago [8, 9], but peculiarities of its research methods and its main research goals were outlined only recently [7]. Computational manufacturing can be used, in particular, to test characterization algorithms. Because the basic ideas of computational manufacturing are not yet widely known, we discuss them in Section 2 in connection with the main goal of this paper.

Final conclusions are given in Section 4, where we also discuss new horizons for further applications of the general idea presented here.

2. Computational manufacturing as a tool for testing on-line characterization algorithms. Computational manufacturing takes the same place between theoretical designing and practical manufacturing as computational physics takes between theoretical and experimental physics. Its practical importance is connected with the fact that computational manufacturing experiments are much cheaper and faster than real deposition experiments. This fact is important, in particular, for testing on-line characterization algorithms, because multiple experiments with different coatings and under different deposition and monitoring conditions can easily be performed.



Fig. 1. General structure of the software for computational manufacturing: arrows indicate the directions of information flow

A general scheme of experiments on computational manufacturing is shown in Figure 1. All blocks presented in this figure are realized as separate software modules. The two left blocks simulate coating deposition in a real deposition chamber and data acquisition in a real monitoring device. The control software simulates processes in a control unit of a real deposition plant; in particular, it supplies signals for starting and terminating the layer depositions and for acquiring optical monitoring data. It is an intellectual center of the computational manufacturing software, because it takes decisions about terminating the layer depositions on the basis of an analysis of on-line monitoring data. This analysis is performed using the on-line characterization algorithm.

The above-presented scheme of experiments on computational manufacturing has been already implemented within the OptiLayer software [10] for the case of broadband optical monitoring. Below we use some results

obtained with the aid of this software to demonstrate the main steps of computational manufacturing experiments. We also present a number of results illustrating the situation with the practical instability of on-line characterization algorithms.



Fig 2. Theoretical transmittance of the 42-layer hot mirror

For our computational experiments we use a theoretical design obtained by the OptiLayer software for the so-called hot mirror design problem. The goal of this problem is to design a multilayer optical coating which transmits almost all light in the visible spectral region from 400 nm to 700 nm and reflects almost all infrared radiation in the spectral region from 700 nm to 1200 nm. The thus-obtained design has 42 layers of two alternating thin film materials with refractive indices equal to 2.35 and 1.45 (typical values for titanium dioxide and silica dioxide used widely as thin film materials). These materials all called high and low index materials, respectively. Spectral transmittances of the theoretical design obtained is shown by the solid curve in Figure 2.



Fig. 3. Simulated deposition rate of one of the thin film materials

Computational manufacturing experiments require a simulation of principal factors causing production errors. One of the main factors related to the coating deposition process is instability of deposition rates of thin film materials. Time dependencies of these rates can be represented as random processes with rather small correlation times. Figure 3 illustrates the simulated deposition rate of the high index material with a mean rate value of 4 Å/sec, rate fluctuations of 1 Å/sec, and a correlation time of 3 sec. The simulated deposition rate of the low index material has a mean rate value of 8 Å/sec, rate fluctuations of 2 Å/sec, and a correlation time of 3 sec. These rates are used in the computational experiments discussed below. The specified parameters are close to those of deposition rates of modern deposition processes.

The total thickness of all layers of the 42-layer design obtained is more than 4000 nm. With the above

rate values, the net time required for depositing such a coating is close to 2.5 hours. In fact, a production time is much larger, because a considerable time is spent for the deposition chamber preparation, there are time intervals between layer depositions, etc. Computational experiments are performed in an internal time scale in which time runs hundred times faster than in reality and results can be obtained in a few minutes.



Fig. 4. One of the arrays of simulated measurement data acquired during the deposition of the ninth layer of the hot mirror (solid curve) and theoretical transmittance at the end of this layer deposition (dashed curve)

Modern broadband optical monitoring devices enable measuring of coating transmittance/reflectance at hundreds of spectral points in a few milliseconds. In the computational experiments presented here, we simulate optical monitoring of coating transmittance at 501 spectral points equally distributed in the wavelength region from 400 nm to 900 nm. There are different types of errors associated with transmittance measurements. In the examples considered below, we simulate random errors in measurement data, which means that the measured transmittance value at the spectral point  $\lambda_j$  differs from the true transmittance value at this point for a random value  $\delta T_j$ . Random errors in transmittance data are normally distributed and their standard deviation is called the level of random errors. The solid curve in Figure 4 presents one of the arrays of transmittance data acquired during the computational deposition of the ninth layer of the hot mirror. This curve connects 501 measured transmittance values. The level of random errors in Figure 4 is equal to 1%.

By  $\{T_{\text{meas}}(t; \lambda_j)\}$  we denote the array of measured transmittance data acquired by the optical monitoring device at the time instant t. The control software is used to prescribe how often transmittance measurements should be performed. In practice, time intervals between measurement data acquisitions may vary from fractions of seconds to several seconds. The main purpose of the control software is to find the appropriate time instants for terminating layer depositions. For this purpose, the arrays of measured transmittance data are analyzed immediately after their acquisitions. It is clear that the analysis of measured data should be as fast as possible. Accordingly, the on-line characterization algorithms used by the control software should work very fast.

All on-line characterization algorithms are based on the comparison of measured transmittance data with theoretically calculated transmittance data for an optical coating with growing layer thicknesses. By  $\{d_1^t, \ldots, d_m^t\}$ we denote the thicknesses of layers of a theoretical coating design. Here *m* is the total number of coating layers. Suppose that *k* coating layers have been already deposited and the (k + 1)th layer is currently deposited. The thicknesses of deposited layers are not equal to the theoretical values  $d_1^t, \ldots, d_k^t$  because of production errors associated with previous layer depositions. Denote the actual thicknesses of these layers by  $d_1^a, \ldots, d_k^a$ . The actual thicknesses of deposited layers are not known precisely; instead of them, some estimates for these thicknesses were obtained by the control software at the previous deposition steps. We denote these estimated thickness values by  $d_1^a, \ldots, d_k^a$ . Let *d* be the growing thickness of the (k+1)th layer. An on-line characterization algorithm should be able to determine this value at any time instant when measurements are done.

Let us consider a basic scheme of the on-line characterization algorithm that got the name "sequential algorithm" in one of our previous publications [11]. Denote by  $T(d_1^e, \ldots, d_k^e, d; \lambda)$  the theoretical transmittance of the (k + 1)-layer system with the fixed thicknesses  $d_1^e, \ldots, d_k^e$  of the first k layers and a variable thickness d of the (k + 1)th layer. This theoretical transmittance depends also on the incident light wavelength  $\lambda$ . Methods for calculating the theoretical transmittance of any multilayer system are well known and their description can be found on the Internet [12].

Let us introduce the discrepancy function

$$F_t(d) = \sum_{j=1}^J \left[ \frac{T(d_1^e, \dots, d_k^e, d; \lambda_j) - T_{\text{meas}}(t, \lambda_j)}{\Delta T_j} \right]^2, \tag{1}$$

where t is the time instant at which the array of measured transmittance data was acquired,  $\Delta T_j$  are measurement data tolerances which account for measurement data accuracy at various spectral points, and the summation is performed over all spectral points at which measurements were made (J is the total number of data in the array of measured transmittance data).

A sequential characterization algorithm is basically an algorithm of a one-dimensional minimization of the discrepancy function with respect to the unknown thickness of the (k + 1)th layer. There could be numerous versions of the sequential algorithm connected with different minimization approaches or modifications of the discrepancy function. For example, some preliminary integration or smoothing of experimental data can be performed in the situations when arrays of measurement data are acquired in short (fractions of seconds) time intervals.

A version of the sequential algorithm is realized in our OptiReOpt software that can be used for the on-line determination of a growing layer thickness [13]. The same algorithm is used by the computational manufacturing option of the OptiLayer software. Computational experiments whose results are shown in Figures 5-7 were performed using this characterization algorithm.



Fig. 5. Comparison of the transmittance of computationally manufactured coating (dashed curve) with the transmittance of theoretical design (solid curve): 0.2% random errors in transmittance data, the two-second time intervals between data measurements

Random errors of modern on-line monitoring devices are usually lower than 1%. Figures 5 and 6 illustrate some final results of one of the computational experiments with the 0.2% level of random errors in measured transmittance data. In these experiments, the time intervals between data measurements were specified equal to two seconds. In Figure 5 the dashed curve presents the transmittance of computationally manufactured coating, while the solid curve is the transmittance of the theoretical design (the same curve as in Figure 2). Figure 6 shows relative errors in the layer thicknesses of the computationally manufactured coating. These errors are defined as relative deviations of the actual thickness values  $d_k^a$  from the theoretical thickness values  $d_k^t$ .

The computational experiment illustrated by Figures 5 and 6 should be considered as a successful one. Deviations of the coating transmittance from the theoretical transmittance are inevitable in practice and the level of deviations observed in Figure 5 is quite acceptable. Other computational experiments with the same parameters of deposition rates and on-line measurements give qualitatively the same results as depicted in Figures 5 and 6.

Obviously, an increase in levels of error factors leads, in general, to a decrease in accuracy of thickness monitoring. One should expect the same consequence from increasing the time intervals between data measurements. Multiple computational experiments confirm this expectation. An increase of production and measurement errors also results in a loss of stability of computational manufacturing, which means that some experiments give practically acceptable results, while other experiments demonstrate a total failure of manufacturing. This loss



Fig. 6. Relative errors in layer thicknesses of the computationally manufactured coating with the transmittance shown by the dashed curve in Figure 5

of stability is usually connected with a failure of the characterization algorithm to determine thicknesses of some coating layers correctly. We refer to such situations as to the practical instability of the on-line characterization algorithm.



Fig. 7. Relative errors in thicknesses of the deposited hot mirror layers causing a failure of manufacturing: manufacturing was interrupted after the deposition of 32 layers

Figure 7 illustrates a situation with the practical instability of the on-line characterization procedure. This figure presents the results of one of the computational experiments with the 0.5% level of random errors in measured transmittance data and the four-second time intervals between data measurements. The computational experiment was interrupted after the deposition of 32 hot mirror layers because of too high errors in the thicknesses of several deposited layers. Such errors entirely destroy spectral performance of the hot mirror.

Multiple computational experiments with the 0.5% transmittance errors and the four-second time intervals between data measurements show a failure of manufacturing in nearly one third of the experiments performed. In all the cases studied, this failure is caused by the instability of the on-line characterization algorithm.

3. Stabilization of the on-line characterization algorithms. The instability of the on-line characterization algorithms is connected not only with the errors in measurement data  $T_{\text{meas}}(t, \lambda_j)$  in equation (1) but also with

the inaccuracies in the determined layer thicknesses  $d_1^e, \ldots, d_k^e$  of the previously deposited layers. It was shown in our previous work [11] that it is possible to obtain more stable on-line characterization results by using the algorithms that enable redeterminations of thicknesses of previously deposited layers at each new step of the on-line characterization procedure. In [11] such algorithms were named the triangular algorithms. They require introducing discrepancy functions that utilize not only the last array of measured transmittance data as in equation (1) but all previously acquired measurement data.

Unfortunately, the triangular algorithms are much more time-consuming than the sequential ones. Their computational speed slows down significantly with the growing number of coating layers and with decreasing the time intervals between data measurements. For these reasons, they are not suitable for the on-line control of layer thicknesses, especially when complicated coatings with many layers are deposited.

In this paper we present a simple idea that can be used to improve the stability of characterization algorithms in any modern production environment. This idea consists in stabilizing the discrepancy function minimization by taking into account material deposition rates.

Optical monitoring is sometimes combined with quartz crystal monitoring based on measuring material deposition rates. In such situations, on-line records of time dependencies of deposition rates are available in parallel with arrays of optical monitoring data. Some estimates for deposition rates are also available in the situations when quartz crystal monitoring is not used. Modern deposition processes are usually rather stable, which means that mean deposition rates are well reproduced from one deposition run to the next deposition run. Hence, the deposition rates can be estimated on the basis of the results of previous deposition experiments and, quite often, with an accuracy up to a few percent. Such a good estimate for a growing layer thickness gives the value rt, where r is the deposition rate of a current layer and t is the time passed from the beginning of its deposition.

Instead of the discrepancy function (1), we consider the new function

$$\Phi_t(d) = F_t(d) + \alpha \,\Omega(d - rt) \,, \tag{2}$$

where  $\Omega$  is a stabilizer and  $\alpha$  is a control parameter which will be traditionally referred to as a regularization parameter.

The simplest form of a stabilizer is as follows:

$$\Omega(d - rt) = (d - rt)^2.$$
(3)

Other even power values may be used instead of the power 2 in equation (3). If the deposition rate r varies in time, then one should substitute rt in equation (3) for  $\int_0^t r(t) dt$ . The choice of the parameter  $\alpha$  in equation (2) is discussed below.

To improve the stability of characterization algorithms, we propose to minimize the function (2) instead of the discrepancy function (1). The essence of this idea is schematically illustrated in Figure 8. Due to the above-discussed inaccuracy of the discrepancy function (1), its minima can be located far from the true layer thickness value at the time instant t. Adding a stabilizer to the discrepancy function (1) enables one to exclude the certainly wrong solutions for d that are located far away from the estimate of rt provided by the material deposition rate.

Consider one of the possible approaches to choosing a value of the parameter  $\alpha$  in equation (2). Suppose that the (k + 1)th coating layer is currently deposited. An inaccuracy of the discrepancy function (1) is associated with errors in transmittance data and with inaccuracies of the estimated thickness values  $d_1^e, \ldots, d_k^e$ . Let us estimate the inaccuracy of  $F_t(d)$  associated with transmittance errors by the value

$$\delta^2 = \sum_{j=1}^J \frac{\langle \delta T_j \rangle^2}{(\Delta T_j)^2},\tag{4}$$

where  $\langle \delta T_j \rangle$  is the estimated root mean square (rms) level of errors in transmittance data and  $\Delta T_j$  are the same tolerance values as in equation (1).

By  $\mu_k^2$  we denote the value of the discrepancy function at the end of the deposition of the kth layer. We use this value as an estimate of the discrepancy function inaccuracy associated with the inaccuracies of  $d_1^e, \ldots, d_k^e$ . The total inaccuracy of  $F_t(d)$  is estimated by  $\delta^2 + \mu_k^2$ . We will also use the last value as an estimate for the minimal values of  $F_t(d)$  during the deposition of the (k + 1)th layer (see Figure 8).

Suppose that we can estimate the deposition rate r with an accuracy of m %. This means that the simplest scheme of monitoring the (k+1)th layer by the deposition time  $t = d_{k+1}^t/r$  enables one to obtain the thickness



Fig. 8. Schematic illustration of the idea of discrepancy function stabilization: solid curve — the discrepancy function at the time instant t, dashed curve — the stabilizer at the time instant t, squares — possible solutions for d without stabilization

of this layer with the same accuracy of m %. Let us choose a value of  $\alpha$  such that

$$\alpha \Omega \left( \frac{m \%}{100 \%} d_{k+1}^t \right) = \delta^2 + \mu_k^2.$$

$$\tag{5}$$

Recall that the sum in the right-hand side of equation (4) is an estimate for the minimal values of  $F_t(d)$ . Choosing  $\alpha$  in accordance with equation (4) provides a fast increase of  $\alpha \Omega$  when d deviates from rt more than  $\Delta d = (m \%/100 \%) d_{k+1}^t$  and thus prevents existing of  $\Phi_t(d)$  minima far away from rt.

The idea we propose was checked by computational experiments with the hot mirror. First we performed 20 computational manufacturing experiments with 1 % transmittance errors, the four-second time intervals between measurements, and without a stabilization of the on-line characterization algorithm. More than 50 % of those experiments were unsuccessful because of high production errors connected with the instability of the on-line characterization algorithm. After that, another series of 20 experiments with the algorithm stabilization was performed. In these experiments we specified the same level of errors in transmittance data and the same time intervals between measurements as before.

A stabilizer was taken in the form of equation (3) and the deposition rates for both the thin film materials were specified with 5 % errors as compared to the mean rate values set in the software module simulating the coating deposition (see Section 2). Such a specification models a practical situation, because material deposition rates are never known precisely. In equation (4) for the regularization parameter  $\alpha$ , we put *m* equal to 20 %. This means that rather large deviations of *d* from *rt* were allowed by the stabilizer. In spite of this fact, all 20 deposition experiments were successful, which confirms the efficiency of the stabilization approach proposed.

4. Conclusions. We believe that the idea presented in this paper has a promising future. It enables developing computationally efficient and stable characterization algorithms for practically all modern production environments, because material deposition rates can usually be estimated with a high accuracy. Algorithms based on this idea can be very flexible. It is possible to apply the idea of stabilization only to those layers that are not reliably controlled by optical means. It is possible to reveal such layers with the use of computational manufacturing experiments. For example, numerous computational experiments show that in the case of the above-considered hot mirror such layers are the layers labeled with the numbers 34, 39, and 40.

Because of a high stability of modern deposition processes, it has become possible to control depositions of some layers by measuring their deposition times. This approach is now often applied to the control of very thin layers or those layers whose thickness variations produce only insignificant variations of transmittance and reflectance. Monitoring of some coating layers by time can be included into the general algorithmic scheme considered in this paper by specifying appropriate values of the parameter  $\alpha$  for these layers.

The idea proposed is the most natural one for combining optical monitoring data and quartz crystal monitoring data in a single characterization scheme. It can be used not only for the characterization of multilayer optical coatings but also for the on-line and off-line characterization of coatings with variable refractive index profiles (also referred to as rugate coatings) when optical monitoring data and quartz crystal monitoring data are available simultaneously.

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