Exclusive examples of high-performance thin-film optical filters for fluorescence spectroscopy made by plasma-assisted reactive magnetron sputtering

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ABSTRACT

For more than four decades band-pass filters are important components of microscopes used for the fluorescence spectroscopy. During all the time this special field of application has been one of the main drivers for research and development in thin-film optics, particularly for the thin-film design software and the coating technology. With a shortwave pass filter, a multi-notch filter, and a classical band-pass filter as examples of such filters provided for the latest generation of fluorescence microscopes we present the state-of-the-art in coating design and technology. Manufacturing these filters is a great challenge because the required spectral characteristics need necessarily multilayers with up to 300 layers and overall thicknesses up to 30 µm. In addition, the designs require also 3 to 5 nm as thinnest layers and all the layers are completely of non-quarterwave type. The filters were manufactured in a rapid-prototyping regime by a Leybold Helios plant using plasma-assisted reactive magnetron sputtering of thin films of different metal oxides. Designed and real spectra are compared and differences are discussed. Measurement results of other optical and non-optical characteristics as film stress, total integrated scattering, and micro roughness are presented.

Keywords: Thin film optical filters, optical broadband monitoring, magnetron sputtering

1. INTRODUCTION

Thin film filters utilized in fluorescence spectroscopy or other fields of high-end instrumentation are characterized by closely separated spectral high transmission regions from rejection regions of optical density (OD) 4, 5 or more. Such applications require a variety of shortwave-, longwave pass filters, multi-notch filters, or band-pass filters prepared with steep edges of few nanometers, and also spectrally positioned within less than half a percent of the design wavelength. This in turn enforces a large total thickness of the layer stack connected with a large number of layers. A successful production depends on several aspects. First the deposition plant must be able to form dielectric layers with reproducible optical constants and low losses. Satisfactory deposition rates and easy handling are of further interest regarding machine operation. Additional requirements include the development of robust designs[1] as well as a precise monitoring device and the choice of a suitable monitoring regime. Together with the demand on sufficient yield and economic production it is preferable to refrain from test-runs, which would last for instance in such extensive cases ten to twenty hours.

Therefore, a reliable process concept and the capability to manufacture complex dielectric filters in a rapid-prototyping regime are of extraordinary importance. The following manuscript does also serve as a report on experience gained with the plasma assisted reactive magnetron sputtering (PARMS) machine[2] within two and half years of operation in an industrial environment. During this time more than 500 coating batches were manufactured. More than 200 different coating designs were successfully realized, with most of them exhibiting a total thickness between 10µm and 20µm. Optical broadband monitoring has been applied to all these filters, offering highest flexibility in daily product change[3].

In this introduction basic features of the plant and also the monitoring concept will be shortly summarized. With a certain demand on complex dielectric coatings in mind the given examples therefore represent almost typical production samples. As designed they are immediately realizable with a coating machine capable to reproduce stable coating conditions and a monitoring strategy, which merely considers possible thickness error sensitivity within the growing stack.

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Other than that mentioned also non-optical and mechanical properties can play a limiting factor in achieving high performance and environmental stable coatings. Therefore knowledge of the thin film micro structure plays an important role. This basic film property may be accessible by an experimental analysis of film stress, total integrated scattering, and micro roughness. Such results from single layer characterization of silica, tantala and niobia have already been presented recently[4]. Depending on the individual application these are the metal oxides chosen to meet the specified requirement.

1.1 PARMS

The capability of the Leybold Helios plant using the PARMS process to produce complex thin films of different metal oxides has already been demonstrated previously[5,6]. Excellent results were achieved by applying single wavelength monitoring and also by time control. The later method underlines the basic stability of this deposition process.

The main feature of this magnetron sputter technology is the separation of the material deposition compartment from the oxidation stage. It is possible to run the dual magnetrons in a near metallic transition mode, which prevents arcing and results in high deposition rates of for example 0.6nm/s for high index materials. Complete oxidation will be realized by applying reactive oxygen from a RF-plasma-source to the previously deposited non-stoichiometric or near metallic monolayer. For this reason the substrates are positioned in a fast rotating turntable that allows for the sequential surface treatment for the required few milliseconds. Running the process with the same amount but non-activated oxygen would cause strong under-stoichiometry with k-values of the films higher than $1\cdot10^{-3}$ in the visible spectral range.

Another important plant configuration is the substrate feeding mechanism via load lock. 12 substrate holder positions are available and substrates with a maximum diameter of 4 inch can be loaded. The targets and their surroundings can stay under vacuum condition which prevents early delamination of particles or flaking due to incorporation of atmospheric moisture. Several coatings summed up to a maximum of about 80µm total overall thickness can be done within one maintenance cycle with satisfactory defect or particle densities.

1.2 Optical broadband monitoring

The large rotation speed of the turntable allows for several transmittance measurements per second with a suitable fiber coupled CCD-spectrometer at distinct positions. These spectra are utilized both for online monitoring and controlling the current coating process and for reconstructing and analyzing the already deposited layer stack. In special cases such a reengineering is necessary and also a reoptimization of the remaining layers is required. To a certain degree this procedure can even be done automatically[7]. Having this possibility is of emerging interest especially when the deposition time amounts to more than ten hours.But usually it is preferable to avoid critical situations and to monitor the coating process only as long as error self-compensation effects are applicable[8,9]. By choosing the adequate moment to change the witness sample, an error accumulation will be avoided. This procedure can be realized very comfortable especially at this machine, as mentioned before. Hereby the decision to stop the coating run, insert a new test slide and continue the run depends largely on the individual layer system and has to be decided case by case[10]. Finally interruptions of the coating are to be avoided as often as possible to reduce manual operation and maintain an automated process.This general development of monitoring and online-data handling takes advantage from synergetic effects of actual research and thus is an additional important aspect which enables a deterministic thin film filter production.

2. COATING EXAMPLE

2.1 Near infra-red band-pass filter

The following example of a near infra-red band-pass filter demonstrates both the capability for an immediate production and the evaluation of non-optical properties being essential for a proper function. Such a typical user-defined specification for a sensor application is given in table 1. Though the integrating character of the later device requires mean values in the spectral blocking regions only, the complexity of the design has to be quite high, for the complete UV and visible spectral range have to be damped. This actual design solution consists of 104 layers with 19µm total thickness. With silica always being the low index material tantala had to be the high index material, which in this case has been prescribed by the customer. Both materials were utilized in nearly equal parts.

Table 1. Specified spectral requirement of the NIR-band-pass-filter.

Test criterion	Type	Requirement
300nm – 920nm	Optical density (mean)	OD > 4.5
940nm – 950nm	Transmittance (absolute)	T > 90%
970nm – 1100nm	Optical density (mean)	OD > 3.5
T = 50%	Half-wave-position	935nm / 955nm ± 2nm

The resulting transmittance spectra are depicted in figure 1. They were carried out with a Perkin Elmer Lamda950 spectrophotometer on Ø25mm substrates of thickness 2mm. The good agreement even with any spectral peak position in the blocking band reveals the correspondence of the modeled set of dispersion data utilized to describe the spectral behavior during the broad band monitoring process. Additionally in this respect it may worth mentioning that the applicable spectral range of the utilized online spectrometer ends at 940nm. So none or only a part of the rising pass-band-edge could be used for online-thickness determination during film-growth. Some measurement artifacts can be identified, especially when the NIR detector is applied beyond 850nm. Though some deviations in the pass-band transmittance can be detected, overall losses inside the pass can be deduced to less than one percent. Here the BK7-sample seems to show a slightly higher loss-value, though considering Fresnel backside reflection.

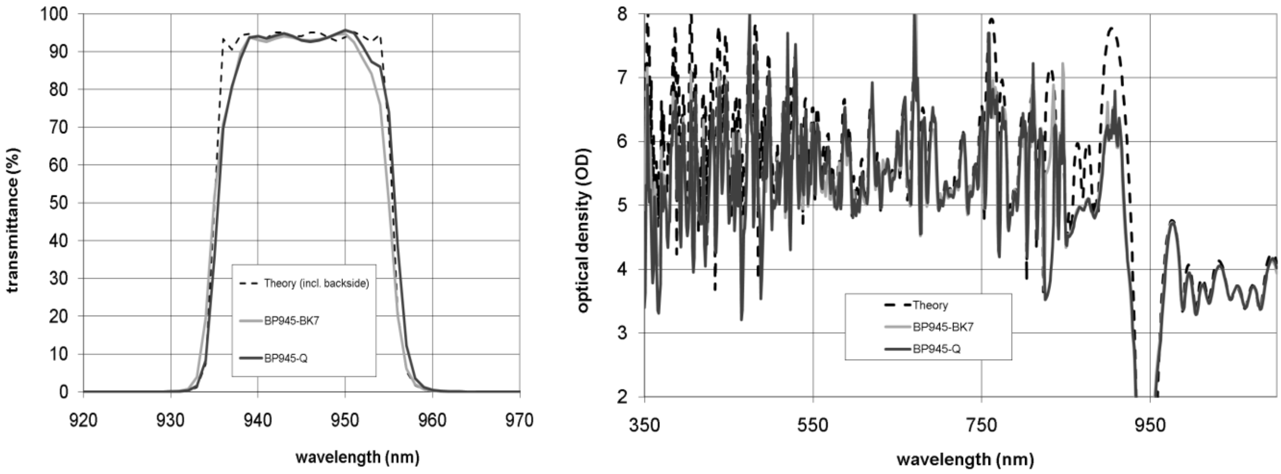


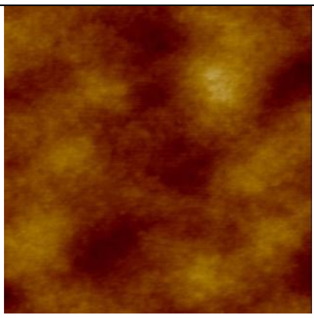
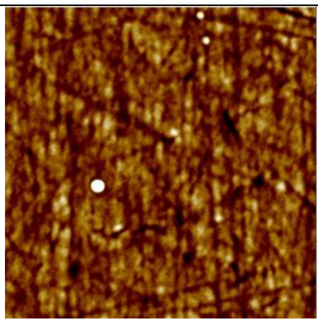
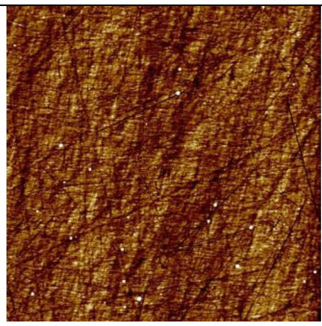
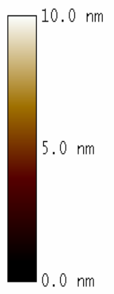
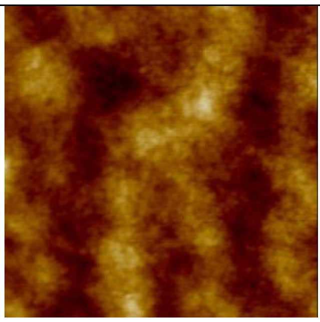
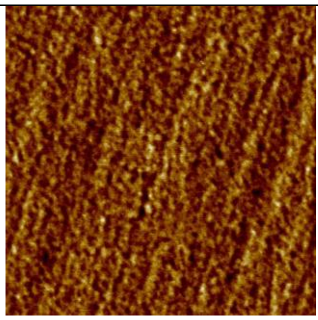
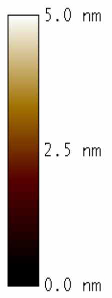
Figure 1. Transmittance (left) and optical density spectra (right) of the NIR-band-pass-filter, including backside reflection. Comparison of measured filters on BK7 and on quartz-substrates with the theoretical values.

Table 2. Results of AFM-roughness measurements at two NIR-band-pass-filters on BK7 and quartz-substrates.

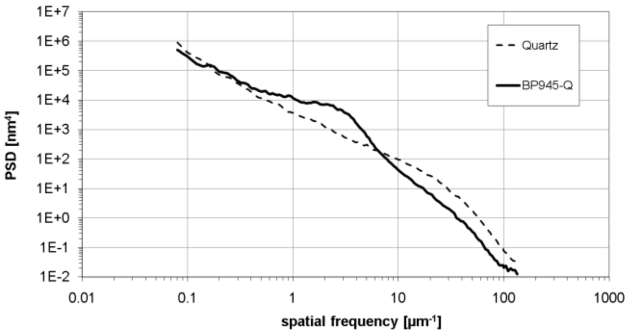
	rms roughness (nm)		
Sample-substrate	**1x1 µm²**	**10x10 µm²**	**50x50 µm²**
BK7	0,75	1,30	1,59
Quartz	0,56	0,58	0,59

Atomic force microscopy (Veeco Dimension 3100 AFM) was applied to the surface of the two samples. Table 2 summarizes the root mean square (rms) roughness results gained at three different sized area-scans. The according topographic plots are presented in table 3. Both types of substrates of the same polishing batch showed very similar tabulated rms-values. Already at this state it can be noted, that despite the fairly large film thickness the applied sputter process does not increase the micro-roughness, at least if the substrate is well polished. Further information can be gained by determining the power spectral density (PSD) function with respect to the spatial frequency.

Table 3. Topografic AFM-pictures of two NIR-band-pass-filters on BK7 and quartz-substrates.

Sample-Substr.	**1x1 µm²**	**10x10 µm²**	**50x50 µm²**	**Heigth (nm)**
BK7				
Quartz				

The PSD-functions (figure 2) are derived from analyzing the height-profile of the AFM-data shown above[11]. For the present the interpretation of the results can lead to the following statements. At least at the quartz substrate no significant granulation can be found, which may be typical for thick coatings, deposited with other magnetron sputter processes. Instead, for certain spatial frequencies even a smoothening effect can be denoted.



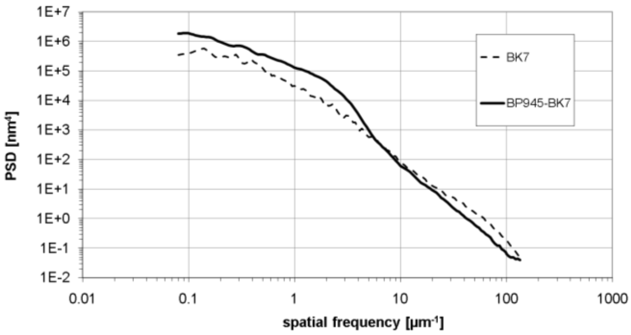


Figure 2. PSD-functions computed from AFM-data of NIR-band-pass-filters and their corresponding substrates, quartz (left) and BK7 (right).

Finally total integrated scattering (TIS) measurements were performed on these samples to link the morphology aspects with optical parameters. TIS-data of the coated filter on the two different substrates are depicted in figure 3. The unit of the ordinate almost represents the integral amount of scattered light in percentage, whereas the contribution from small-angle scattering will be partly excluded from the integration sphere. Nevertheless the difference between the two substrate types seems to be more obvious than one would assume form the roughness measurement results. Increased PSD-values in the low spatial frequency domain indicate a certain accumulation, which may relate to a higher TIS-level at distinct wavelength positions depending on the realized layer stack. If this effect can be deduced from the marginally higher basic roughness of the BK7-substrate alone has still to be clarified. The level of the TIS-data on the quartz-substrates is very low and can almost not be distinguished from the noise-level.

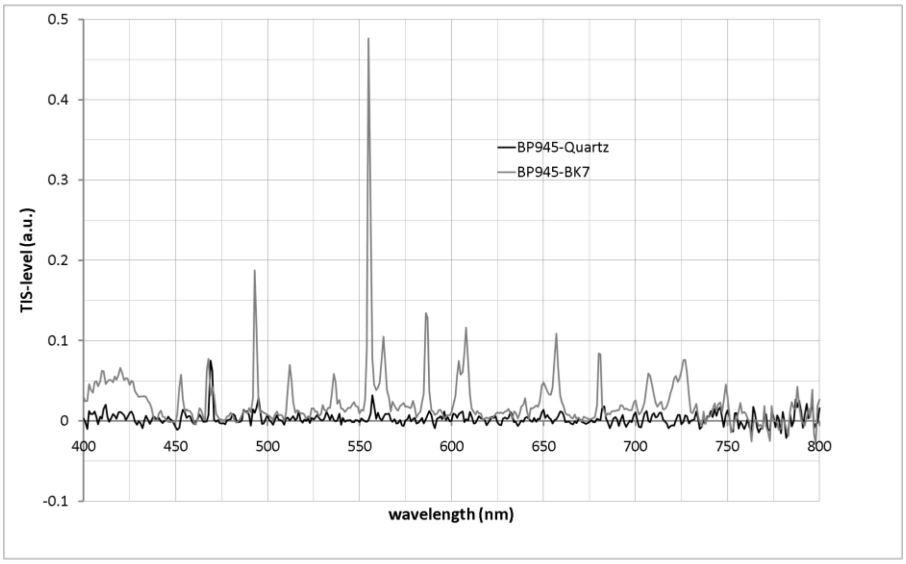
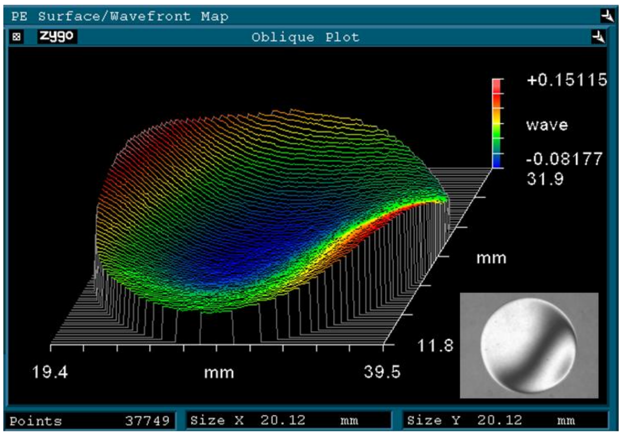


Figure 3. TIS-measurements of two NIR-band-pass-filters on BK7 and quartz-substrates.

An additional mechanical property of interest is the film stress. For this reason the surface form of a backside roughened Ø25mm BK7-substrate of thickness 2mm is pre-measured by an interferometer (Zygo VeriFIRE XP/D) and measured after the coating again (figure 4).



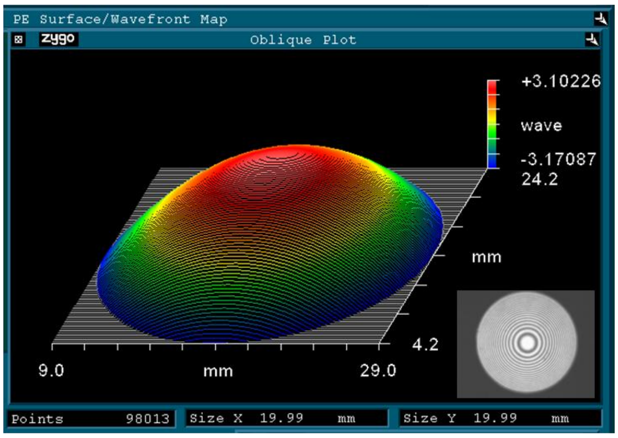


Figure 4. Measured surface deformation of a Ø25mm BK7-test-substrate before (left) and after depositing the 19µm NIR-band-pass-filter (right). The small in-sets show the according interferograms.

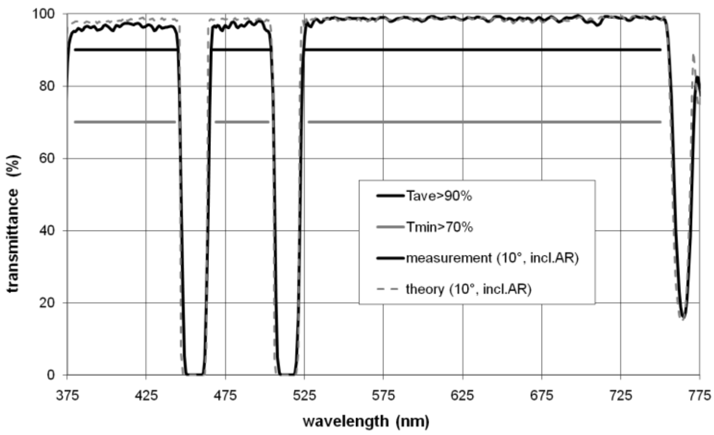
The according difference in curvature can then be directly inserted into Stoney's formula to calculate the film stress of a single layer or in this case of an average value representing the stress of a complete layer stack. With these data one computes compressive stress of 288 MPa. This value is in good accordance with the averaged single layer contributions[4].

2.2 Multiple notch filter

One typical type of fluorescence filters are represented by so called notch filters which reject the laser used to induce fluorescence in the specimen of interest whereas the broadly emitted light will be transmitted for analysis. To avoid signal distortion the laser will be typically blocked to OD4 or more. Such a setup can utilize several laser lines which require an individual blocking of each laser wavelength realized by a single filter surface.

The given example filter blocks the argon-laser-lines 458nm and 514nm. The rejection bands and the pass bands have to be separated by steep edges of 2.5nm from 90% to 0.01% transmittance to allow for a production tolerance of ±2.5nm in wavelength position. To build the reflection zones the 19µm design consists of 199 layers with interlaced detuned thick and thin layer pairs. The individual layer thickness values range from 470nm to 3.5nm. Figure 5 shows the measured compared with the theoretical transmittance spectra. A standard broadband anti reflection coating in the visible spectral range is applied to the substrate backside.

The excellent agreement with the calculated design curve reveals a realized error budget of few tenth of a nanometer only. The result exemplifies that especially this type of layer sequence is extremely sensitive to error self compensation effects when optical broad band monitoring is applied. A certain lack in transmittance can be identified in the shortwave pass region which can be attributed to polishing effects on the BK7 substrates.



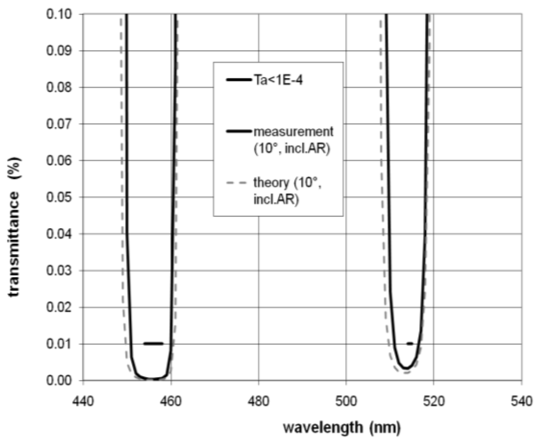


Figure 5. Transmittance (left) and enlargement of the rejection bands (right) of the double-notch-filter.

An important aspect regarding the yield of such a narrow spectral specification is the thickness homogeneity distributed on the substrate holders. A certain degree of variation can be attributed to the growing race-track depth with increasing lifetime of the targets. Online measurements are performed at the centre position of a monitor glass sample located at one of the 12 substrate holder positions inside the turntable. This radial position is therefore fixed for all other loading positions within an accuracy of about a tenth of a percent. As long as the substrates are positioned near to this radial measure a sufficient thickness distribution can be assumed. Then only the geometric distance of the substrates towards the target surfaces influences the gathered thickness. This may vary slightly with the loading position as well as the precision in storing reproducibility during loading the substrate holder into the turntable.

With small substrate dimensions even a large number of samples can be coated within one coating run. Figure 6 demonstrates a typical batch-uniformity determination of such a dual notch filter coating run. For each coated substrate the relative deviation from the target values of the 50%-points at the two rising and falling edges, respectively are plotted. Possible inhomogeneity errors like detuning or layer dispersion mismatch can be interpreted from these data, which are not significant in this case.

Ten groups can be identified, each representing one position of a holder inside the turntable. Such an analysis allows for the height correction of an individual loading position, when such a deviation is systematically. Taking the whole batch into account a small correction of 0.4nm of the design central wavelength would be applicable. Nevertheless the complete batch meets the specification, but a batch-uniformity smaller ±0.2% is achievable.

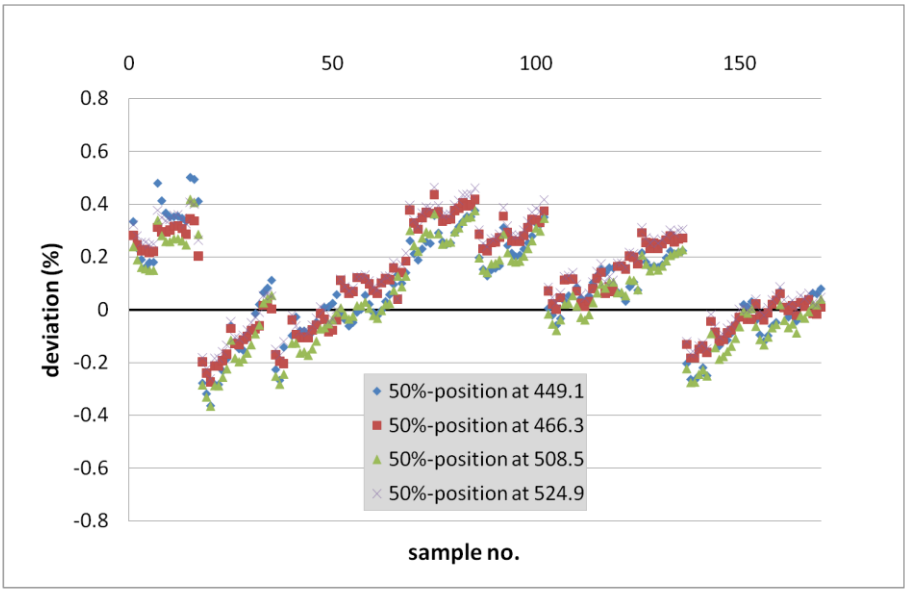


Figure 6. Batch-uniformity calculated at the 50% transmittance values at the 4 edges of the double-notch-filter with respect to the spectral target positions.

2.3 Shortwave pass filter

The final example is given by a shortwave pass filter aimed to block the visible spectral range to a value of OD 5.3 with no exception and exhibiting high transmittance below 390nm.

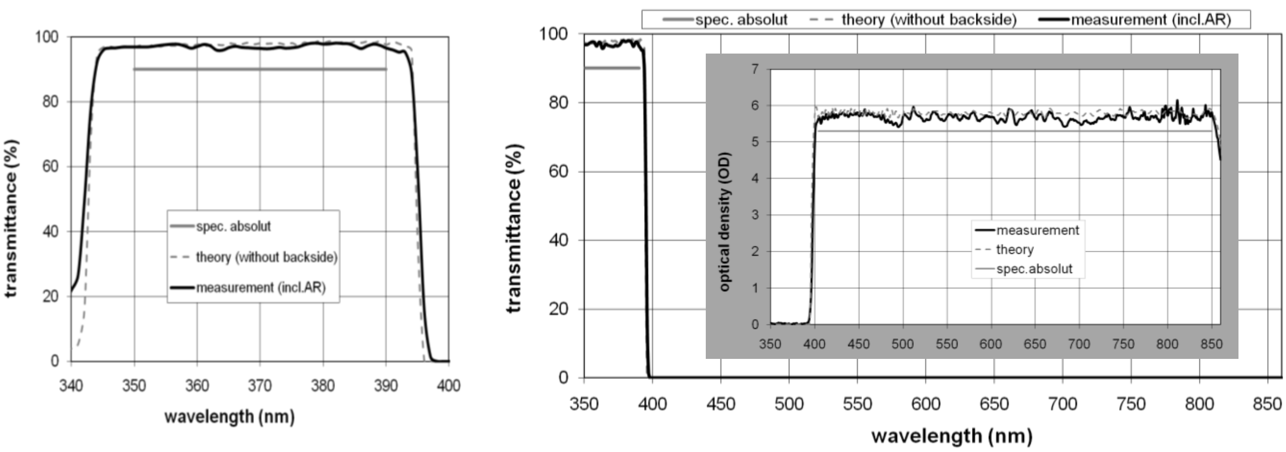


Figure 7. Pass band transmittance (left) and transmittance of the blocking range with optical density (right) of the shortwave pass filter on a quartz substrate.

The design consisted of 171 layers silica/tantala with a total thickness of 13.9µm. This large number of layers seems to be essential to realize a smooth rejection with no arising peaks. The spectral measurement results are presented in figure

7. The blocking range was measured separately with a Perkin Elmer Lamda1050 spectrophotometer, which exhibits an enhanced sensitivity for low intensity levels. High transmittance even near the absorption edge of tantala, exemplify thorough oxidation with low scattering and absorption on quartz substrates. In fact from these measurement k-values in the low 10^{-5} range can be stated for both materials.

3. CONCLUSIONS

This manuscript exemplifies the capability of the PARMS concept to produce exclusive high-performance filters in daily production. The study of the layer morphology of a 19µm NIR-band-pass filters reveals excellent micro-structural properties with no significant additional contribution to the surface roughness - provided the substrate is well polished. Wavelength selective measurements of the total integrated scattering reveal an extraordinary sensitivity to the rms roughness linked with the substrate material. In conjunction with optical broadband monitoring the Leybold HELIOS plant has proved to be an absolutely versatile tool for the rapid-prototyp manufacture of complex optical thin film filters. Applications like filters for fluorescence spectroscopy which require excellent layer properties and narrow spectral specifications can be addressed with batches of sufficient size.

We gratefully thank the Frauenhofer institute of applied optics and fine mechanics (IOF/Jena) for performing the AFM and TIS measurements.

This work was supported by the German Federal Ministry of Economics and Technology, EP090031.

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