Stress compensation with antireflection coatings for ultrafast laser applications: from theory to practice

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Abstract: Each complicated coating, in particular, a dispersive mirror consists of dozens of layers. Thin films layers have mechanical stresses. After summing up stresses from all layers, the resulting stress is high enough to bend even a relatively thick substrate. To avoid this effect we suggest depositing an antireflection coating (AR) at the back-side of the substrate which together with suppression of unwanted reflections from the back side will also compensate this stress. We demonstrate unique, extremely thick and sophisticated AR coating consisting of 71 layers with the total physical thickness of 7.5 µm. This AR coating completely compensates stress from the dispersive mirror coated on the front side and minimizes unwanted reflections.

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1. Introduction

Most of the modern ultrafast laser systems include dispersive mirrors (DM) [1–5] used for accurate phase control required for efficient pulse compression. DMs are multilayer coatings providing high reflectance and specified group delay (GD) or/and group delay dispersion (GDD) in wide spectral ranges. Design and production of these coatings require using effective numerical design algorithms and high precision deposition techniques.

In this paper we consider a multilayer optical element for the multiterawatt few-cycle light wave synthesizer (LWS) based on optical parametric amplification. LWS-20 currently produces sub-5-fs, 80-mJ, 18-TW pulses. The specifications of this optical element are as follows. The DM should have 3″ diameter and an average value of the GDD of +75 fs² of the GDD and reflectance $R > 99\%$ in spectral wavelength range 560-1030 nm.

In addition to these requirements the DM should have the diameter of 75 mm at a substrate with 15 mm thickness. Due to mechanical stresses of DM thin film layers the substrate becomes bended, that distorts wave front quality resulting in the distorting of the beam waveform of laser system. Thicker substrates could be used, but realistic thickness (about 30 mm) will not eliminate the problem. To compensate this stress we coat substrate back side with a coating having approximately the same mechanical stress, similarly to the approach described in [6], Design Problem B. As the result, the surface flatness of the substrate will not be degraded. The back side of the sample is to be covered by an antireflection (AR) coating providing the lowest possible reflection in the working spectral range of the DM, since a small fraction of light penetrating through the DM coating is used to check the laser beam profile. In principle, it is possible to coat the back side with a relatively simple AR having a thick SiO₂ layer next to the substrate. Unfortunately for this approach more detailed knowledge about stress produced by each of layer materials on the DM side is required. Indeed, such AR coating will have a relative contents high-index and low-index layer materials very different from that of DM, and in order to reach good stress compensation the thickness of thick SiO₂ layer should be specially adjusted taking into account detailed stress information. On the other hand, if the back side is coated with AR coating having total physical thicknesses of each layer material close to the total physical thicknesses of the same materials in DM, stress compensation goal will be automatically achieved. Thus we come up to the problem of the design and deposition of extremely thick and complicated AR coatings.

Historically AR coatings are the oldest and the most popular type of optical coatings. Recently, a series of publications on theoretical design of broadband AR and achievable minimum residual reflectance has been published [7–10]. In the present case, AR coating design is different from conventional one because some important conditions must be satisfied. The first important condition is that AR coating thickness is to be consistent with DM coating thickness in order to provide approximately equal stress from both sides of the DM-AR optical element. The second condition is AR coating feasibility. Optimal AR designs contain thin layers (layers with thicknesses less than 10 nm). In [7] we produced two optimal 12-layer AR designs working in the spectral range from 400 to 1200 nm and demonstrated that production of such designs containing layers with 5 and 7 nm thicknesses is realistic. In the present case we expect AR designs consisting of significantly larger number of layers. We expect also many thin layers among them. Actually, these layers can cause a problem in the course of the deposition even in the case of high precision deposition systems.

In this work we report about design and production of AR used in this application. We demonstrate how to reach a balance between theoretical expectations and feasibility demands.
In Section 2 we discuss the theoretical design of AR for our DM application. In Section 3 we present spectral performance of the produced AR and experimental properties of the DM-AR optical element. The final conclusions are presented in Section 4.

2. Design of AR coating for DM application

In optical coatings for our DM-AR optical element we used Nb$_2$O$_5$ as high index material and SiO$_2$ as low index material. The substrate was Suprasil of 15 mm thickness and 75 mm in diameter. Wavelength dependencies of refractive indices of thin-film materials and substrates are described by Cauchy formula:

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

where $A_0$, $A_1$, $A_2$ are dimensionless parameters, $\lambda_0 = 1000$ nm, $\lambda$ is specified in nanometers. The values of these parameters are presented in Table 1.

| Material        | $A_0$     | $A_1$     | $A_2$       |
|-----------------|-----------|-----------|-------------|
| Nb$_2$O$_5$     | 2.218485  | 0.021827  | 3.99968×10$^{-3}$ |
| SiO$_2$         | 1.465294  | 0         | 4.71×10$^{-4}$ |
| Suprasil        | 1.449     | 0.00303   | 4.087×10$^{-4}$ |
| Glass B260      | 1.510027  | 5.2539×10$^{-7}$ | 6.328×10$^{-5}$ |

In the DM-AR optical element, the mirror is to be designed first because meeting target specifications on reflectance and GDD is definitely more challenging than AR designing. Synthesis of DM was performed with the help of needle optimization technique incorporated into OptiLayer software [11,12]. As the result a 74-layer DM was obtained. Theoretical reflectance and GDD in the spectral range of interest are shown in Fig. 1. Physical thickness of the obtained design is $\text{PhT}_{DM} = 7568$ nm and its optical thickness at the wavelength of 800 nm is $\text{TOT}_{DM} = 13435$ nm. Total physical thicknesses of Nb$_2$O$_5$ and SiO$_2$ layers are $\text{PhT}_{Nb_2O_5} = 2979$ nm and $\text{PhT}_{SiO_2} = 4589$ nm, respectively.

![Fig. 1. Theoretical reflectance (a) and GDD (b) of 74-layers DM design (black curves). Green lines indicate target values.](image)

The values $\text{PhT}_{DM}$ and $\text{TOT}_{DM}$ give estimations for physical and optical thicknesses of AR coating compensating DM stresses. According to [10,13] optimal AR designs consist of quasi-periodic groups of layers (clusters). Optical thickness of one cluster $T_c$ is estimated as:
\[
T_r = \frac{\lambda_u}{2} \left[ 1 + \frac{2}{\pi} \arcsin \left( \frac{n_h}{n_l} - 1 \right) \right] = \frac{1030\text{nm}}{2} \left[ 1 + \frac{2}{\pi} \arcsin \left( \frac{2.25/1.46 - 1}{2.25/1.46 + 1} \right) \right] = 585\text{nm}
\]

Eq. (2) and below, \( \lambda_u, \lambda_l \) denote lower and upper boundaries of AR spectral range; \( n_h, n_l \) denote refractive indices of high and low index materials taken at the wavelength of 800 nm. The required number of clusters \( M \) in the AR design is approximately equal to \( \frac{\text{TOT}_{DL}}{22 - 23} \).

According to the estimation of the number of layers in the optimal AR designs [8], the number of layers in one cluster is expected to be:

\[
N_{cl} = 2 \left( \frac{2T_r}{\lambda_l} + 1 \right) = 2 \left( \frac{1170}{560} + 1 \right) = 6
\]

It means that the total expected number of layers \( N \) is about \( N = N_{cl} M = 6 \times 22 = 132 \).

The residual reflectance of AR coatings is defined as

\[
R_{av} = \frac{1}{\lambda_u - \lambda_l} \int R(\lambda) d\lambda
\]

According to the estimation [9] the minimum achievable reflectance is 0.07%.

After the simple theoretical estimations, the numerical results are presented. The evolution of the residual reflectance \( R_{av} \) of optimal AR designs simulated with OptiLayer [12] with respect to the number of clusters \( M \), as well as evolutions of optical thickness TOT, physical thickness PhT and the number of layers \( N \) are shown in Fig. 2. We checked that average increase of optical thickness when adding one cluster is equal to 585 nm that is consistent with theoretical predictions. The number of layers \( N \) is increased by 4–6 layers when adding a new cluster. Average physical thickness of one cluster is 331 nm. Thicknesses of NbO\(_2\) and SiO\(_2\) layers in one cluster are about 150 nm and 210 nm. Then, the number of clusters in AR can be estimated also as: NbO\(_2\) / 150 = 20, and SiO\(_2\) / 210 = 22.

Therefore the number of clusters can be estimated as 20–22 and the number of layers as 120–132. It should be noted that estimation of the number of layers given by Eq. (3) was obtained in [8] for broadband AR with the ratio \( \lambda_u / \lambda_l \geq 2 \). In the present work \( \lambda_u / \lambda_l = 1.8 \), so Eq. (3) can be used only as a rough estimation of the number of layers.

We found that 106-layers AR design solution has the parameters close to the estimated ones: TOT = 13924, PhT = 7748 nm. This design is marked by red asterisks in Fig. 2. Reflectance of this solution is plotted in Fig. 3 by the black curve. The residual reflectance provided by 106-layers AR design is 0.073% in excellent agreement with the theoretically predicted minimum achievable reflectance.
The 106-layer AR design contains seven layers with physical thicknesses less than 10 nm. These layers should be removed in order to avoid problems with deposition of thin layers corresponding to our feasibility requirement about design. We call this procedure as a thin layer removal procedure. We performed this procedure by selecting the thinnest layer in the design and substituting it with the layer of alternate material having the same optical thickness. After this substitution the number of layers in the design is decreased by two, since adjacent layers are formed of the same material. This operation increases the merit function that is partially compensated by the following reoptimization procedure, which changes the design only insignificantly. These iterations are repeated until there are no layers thinner than 10 nm in the AR design. As the result of the thin layer removal procedure, a 71-layer AR design was obtained. Reflectance of this design is shown in Fig. 3 by the red curve. The residual reflectance of 71-layer AR design is 0.08 that is also very close to the minimum achievable reflectance value. It is also important to note, that the total physical thicknesses of layer materials are: $T_{\text{Nb}_2\text{O}_5} = 3077 \text{ nm}$ and $T_{\text{SiO}_2} = 4340 \text{ nm}$, which is quite close to respective values of the DM mentioned above. The thickness difference between DM and AR for $\text{Nb}_2\text{O}_5$ is about 100 nm, and for $\text{SiO}_2$ is about 250 nm. Therefore the goal of the DM stress compensation should be also achieved with sufficient accuracy.

**Fig. 3.** Reflectance of 106-layer optimal AR design and 71-layer modified AR design.

### 3. Experimental results

We produced designed DM and AR coatings using Leybold Optics magnetron sputtering Helios plant, layer thicknesses were controlled using well-calibrated time monitoring [3]. The plant is equipped with two proprietary TwinMags magnetrons and a plasma source for plasma/ion-assisted reactive middle frequency dual magnetron sputtering.

Helios plant is also equipped with broadband monitoring (BBM) system [14]. This system was used in a passive mode for data acquisition only. We performed two deposition runs using Suprasil substrate for the DM-AR optic production and cheaper B260 Glass substrate as a test witness sample. In the first run, the front sides of Suprasil and B260 Glass substrates were covered with DM coatings. In the second run, the back side of Suprasil substrate and front side of another B260 Glass substrate were covered with AR coating. As a result, three samples were obtained: DM coating on B260 Glass substrate (sample DM-B260), AR coating on B260 substrate (sample AR-B260) and DM-AR coatings on Suprasil substrate (DM-AR-Suprasil). The BBM data were recorded for DM-B260 and AR-B260 samples.

Turntable of Helios plant has 16 sample positions located at the same distance from the center of rotation. We placed all samples exactly in the middle of two sample positions of turntable. The B260 Glass substrates were placed on the position where BBM system performed measurements. Figure 4 demonstrates a remarkable correspondence between theoretical and experimental data of AR-B260 sample taken in the course of the deposition by BBM device.

After the deposition, transmittance data of produced DM-AR-Suprasil sample was measured by Perkin Elmer Lambda 950 spectrophotometer in the range from 400 nm to 1300 nm.
nm. In Fig. 5(a) we compare theoretical and measured transmittance data related to DM-AR-Suprasil sample and observe very good correspondence between the data. GD and GDD of the sample DM-AR-Suprasil were extracted from the measurements provided by a white light interferometer and processed with specially elaborated algorithm [15]. In Fig. 5(b) we observe that theoretical and measured GD values are consistent.

We also measured the surface flatness of the produced samples with homemade Fizeau interferometer (the accuracy about $\lambda/20$). The measurements were performed at He-Ne laser wavelength of 632.8 nm. In the case of DM samples without compensating AR coating the flatness of the surface degraded from $\lambda/10$ to $\lambda/4$. The deposition of the AR coating at the backside of the sample improved surface flatness from $\lambda/4$ to the original value $\lambda/10$, corresponding to the uncoated substrate. Therefore full stress compensation with AR coating is confirmed.

![Fig. 4. Comparison of theoretical and experimental BBM transmittance data related to AR-B260 sample.](image)

![Fig. 5. Comparison of theoretical and experimental spectral characteristics of DM-AR-Suprasil sample: transmittance (a) and GD (b).](image)

**4. Conclusions**

We designed and produced a complex multilayer element consisting of DM and back-side 71-layer AR coating for reducing the total stress and back reflections. Deposition of such AR coatings is a challenge for optical coating engineers because even simple AR coatings are very sensitive to errors in layer thicknesses. The deposition of many-layers AR coatings requires a deep understanding of the physical processes in deposition and influence of various factors on thickness control, experience in extracting information from *in situ* measurements.
and making (if necessary) on-line corrections to the deposition process. The goals of stress compensation and suppression of back reflections were successfully achieved.

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