

High Stability Optical Coatings by Employing Gradient Index Designs

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ABSTRACT

Optical thin films oftentimes suffer from their poor power handling capability, if further developments of optical systems especially for applications in the deep UV spectral region are requested. Especially, the lifetime of functional coatings on laser and conversions crystals and resonator mirrors are a limiting factor following from laser-induced degradation. During recent years, tremendous process innovations can be recognized in ion beam sputtering technology enabling the efficient production of graded-index profiles with oxide materials. This process variant is accompanied with advanced process monitoring equipment measuring the thickness of the growing mixture layer. The manufacturing possibilities evolving from this innovative combination are demonstrated by reviewing recent results for coatings used for high power cw- and pulsed laser systems.

INTRODUCTION

The process innovation to prepare graded-index coatings by blending oxide materials within the ion beam sputtering process has been developed few years ago [1]. The first graded-index coatings manufactured by the presented technique were produced by co-deposition of titania and silica. During recent years, investigations in the properties of mixture material employing many more oxide materials have been conducted. For the current presentation, selected results are highlighted covering investigations in graded-index coatings for laser applications with cw radiation and ns- and fs-pulses.

MANUFACTURING OF MATERIAL MIXTURES

Coating Principle

At the Laser Zentrum Hannover, the well-established ion beam sputtering process to manufacture of high quality thin films has been expanded to generate materials mixtures by genuine co-sputtering. The scheme shown in Figure 1 describes the basic components utilized for co-sputtering. Charged and accelerated ions are ejected by the ion source showing a high enough kinetic energy to sputter the coating material from the target plate. The sputtered atoms propagate towards the substrates located in the palette forming a thin film under interaction with oxygen as process gas. The blending of coating material is performed by moving the target consisting of two material zones in respect of the ion beam. Because the

ion beam sputters different material amounts depending on its lateral position of the zone target, this technique can be regarded as a genuine co-sputtering coating method for which the material content in coating can be tuned continuously. In former investigations, different high-index materials have been combined with silica to form mixture materials. Among others, mixture materials consisting of zirconia, niobia and tantalum in conjunction with silica have been investigated in regard to refractive indices, band edge and laser damage properties. The current investigation is concentrated on functional coatings employing a gradient index profile by using zirconia and hafnia mixtures. The coatings were manufactured with ion beam sputtering systems at the Institute of Physics, Vilnius, Lithuania and at the Laser Zentrum Hannover, Germany.

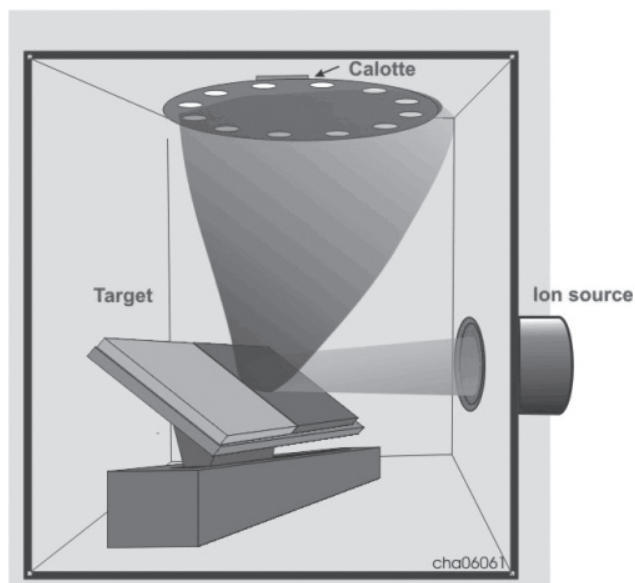


Figure 1: Principle of synthesis of mixture oxide coatings by ion beam sputtering.

Refractive Index Profile

The reliable and automated production of graded-index coatings is based on an exact knowledge of the correlation of lateral zone target position with the corresponding refractive index of the mixture layer. This correlation is represented by a calibration function which can be retrieved by the transmittance of single layer coatings at distinct zone target positions.

As an example, a target calibration for a zirconia/silica zone target is depicted in Figure 2. The graph shows the refractive index at predefined target positions (dots) retrieved from the fitted dispersion curves for the wavelengths 400 and 600 nm. From the displayed data points, a clear propagation of the refractive index along the S-shape curve can be identified which can roughly be structured into three domains. Within an area of about 15 % of the overall travel range at the left and right margins, the refractive index is almost unchanged compared to the extreme target positions. In contrast to that, the refractive index changes rapidly in the crossover range in-between the two target plates.

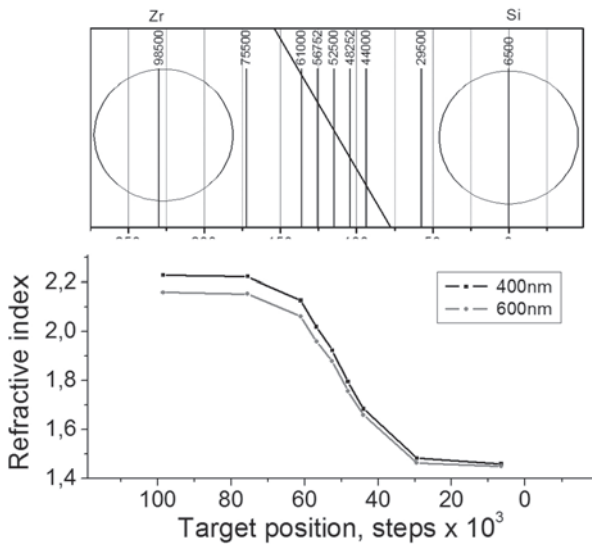


Figure 2: Refractive index of material mixtures (zirconia/silica zone target) as a function of the lateral target position evaluated from the dispersion curves at 400 and 600 nm. In the sketch above the material zones on the target and the approximated erosion area is depicted.

Accordingly, locating the zone target at the extreme target positions, the ion beam predominantly erodes zirconia or silica, respectively, i.e. the material content of the conjugated mixing material can be neglected at these positions. Therefore, classical high- and low-index coatings with the pure coating materials are accessible. But in the crossover range, the refractive index follows continuously the interpolation between the data points. This calibration curve is utilized by the system control as a calibration function. If a distinct refractive index is requested by the thin film design, the system control software derives the corresponding target position from the above curve, in a first order approximation by a linear interpolation.

Wavelength Shift of Absorptance Edge

In previous publications, the absorptance properties of single layer mixture coatings were investigated [2]. Beside the change of refractive indices, the co-deposition of several material

combinations (titania/silica, tantala/silica, hafnia/silica) affects the absorption properties of the synthesized mixtures film drastically. For all of the mentioned material combinations, a strong shift of the band edge can be observed with increasing content of silica in the mixture. Consequently, the usability of coating material can be extended to shorter wavelengths, if the high-index material is blended with silica.

As an example, a guiding mirror for 266 nm was designed employing a tantala/silica mixture for the high index layers and pure silica for the low index layers. Beside the reflecting band at 266nm, the coating shall have high transmittance bands for 532 and 1064 nm. In Figure 3 the measured transmittance of the manufactured coating is displayed. Obviously, the reflecting band is formed in the descending shoulder of the transmittance curve. The corresponding reflectance under s- and p-polarization is shown in the inset graph. Although pure tantala is strongly absorbing below approx. 300 nm, the guiding mirror employing a tantala/silica mixture in the high index layers shows a fair performance at around 266 nm. This example shows clearly that the usability of high-index materials can be extended across the absorption edge of the pure material by blending with silica.

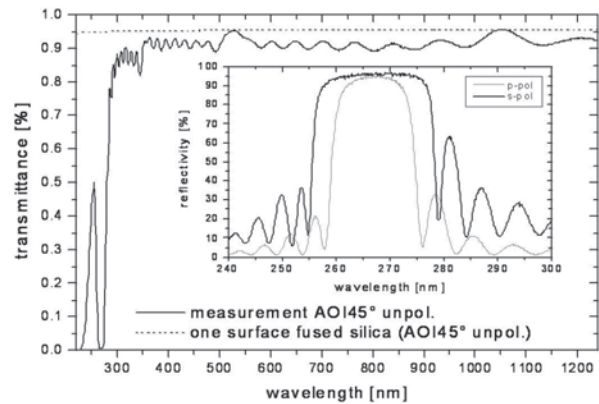


Figure 3: Measured transmittance spectra of guiding mirror (266 nm, 45°), AR 1 ω and 2 ω (1064 nm) employing Ta₂O₅/SiO₂ mixtures; inset: reflectance spectra measured for s- and p-polarization.

Advanced Optical Monitoring

The key component to deposit graded-index coatings at a high precision level is an advanced optical monitoring system. The coating systems used for the current investigation are equipped with a broadband optical monitoring system developed at the Laser Zentrum Hannover e.V. in recent years. This monitoring system is measuring the transmittance of a witness glass placed on a palette position corresponding to the objects to be coated. Hence, the transmittance measurement takes place through the rotating substrate. From the measured transmittance, the thickness of the currently growing layer is determined by comparing the measurement curve with the

theoretical expectation for the ideal layer stack. The technical details of the advanced optical monitoring system can be found in previous publications [3].

As an example, how optical monitoring is used for controlling the thickness of graded-index coatings, a Rugate filter showing high reflectivity at 266 nm has been designed with a quasi-continuously varying refractive index profile established by a sequence of very thin sub-layers. The design consists of close to 400 sub-layers with physical thicknesses between 4 and 6 nm and an overall physical thickness of 2.0 μm . The maximum content of zirconia in the mixture was 50 %. Finally, the design comprises 352 sub-layers in total, because a number of adjacent layers uses the same materials mixture and were merged together.

The resulting in-situ spectrum of the witness sample after termination of the last layer in the stack is presented in Figure 4. It can be seen that the transmittance measured with the broadband monitoring system inside of the coating plant corresponds largely with the theoretical expectations derived from the initial design. This result shows that the broadband optical monitoring system is capable to control materials mixture layers with a thickness down to a few nm.

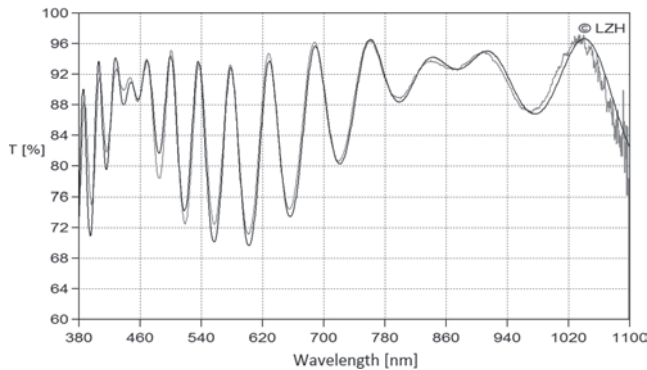


Figure 4: Transmittance spectra of the witness sample after the termination of the last layer, calculated transmittance (without noise), transmittance spectra measured with broadband optical monitor (with noise).

DAMAGE TEST RESULTS

Graded-Index AR-Coating for High Power Lasers

As an example for a graded-index coating for high power cw laser applications, an antireflection coating has been realized on fused silica lenses used inside a focussing system for material processing. The antireflection band has to cover a spectral range from 1030 to 1070 nm, i.e. the wavelengths of high power disk, slab and fiber lasers. Furthermore, the design shall provide high transmittance at the wavelengths 532 and 633 nm for guiding sensor and pilot lasers through the optical system. A graded-index design providing these specifications by using zirconia/silica mixtures has been developed consisting

of 18 sub-layers with a gradually changing refractive index with an overall physical thickness of 0.75 μm . The design and the resulting transmittance curve are presented elsewhere [4].

For the demonstration of the damage handling capability of the realized graded-index AR coating, a test lens has been evaluated by a test procedure close to the industrial application. The AR-coated fused silica lenses were included in the material processing objective inside a high power fiber laser facility. Among other stability tests, the focusing lenses were exposed to continuous irradiation at a wavelength of 1070 nm for more than 10 minutes without any visible surface degradation. The successful pass through this irradiation test proves the power handling capability of zirconia/silica material mixtures for high power cw-radiation in the near infrared spectral range.

Laser Damage of Mixture Materials with fs-Pulses

During former investigations in the fs-laser damage of single layer coatings of pure coating materials, a deeper understanding in the damage mechanisms in the ultra-short pulse damage regime has been achieved [5]. For this experiment, dense and predominantly amorphous IBS single layers of SiO_2 , Al_2O_3 , HfO_2 , Ta_2O_5 and TiO_2 were tested concerning their fs-laser damage. The results show that the damage threshold is clearly depending on the pulse duration and on the coating material. In more detail, the evaluation of the results revealed, that a model function can be found that the threshold is depending on band gap and pulse duration [6]. This behavior can be clearly explained by the dominant electronic interaction: electrons in the lattice are excited by multi-photon ionization and avalanche ionization leading to a critical electron density which leads to catastrophic damage.

In the next step, the investigation was extended to a set of single layer coatings consisting of titania/silica mixtures. For this sample set, the band gap energy and the refractive index have been evaluated according to the composition parameter which is the content of the high index material in the layer. The results of the damage experiment delivered the same clear correlation between threshold and band gap for the mixture coatings. Obviously, laser induced damage in the mixture layers are based on the same mechanisms as for the pure coating materials in the field of ultra-short pulses.

This experiment has been repeated recently with hafnia/silica mixtures [7]. For this experiment, samples with hafnia contents from 0 % to 100 % have been prepared and the band edge energy determined. These samples were tested with different pulse durations in the ultra-short pulse regime. The results of this experiment (Figure 5) also show a clear correspondence with the model function found for pure materials also for hafnia/silica mixtures. Hence, the damage behavior found for the pure coating materials and for titania/silica mixtures has

been identified for hafnia/silica mixtures, too. In other words, the coating layers formed by co-deposition can be regarded as an artificial material constituted on the molecular basis by the combination of three partners and which shows the same physical behavior like a “real” material.

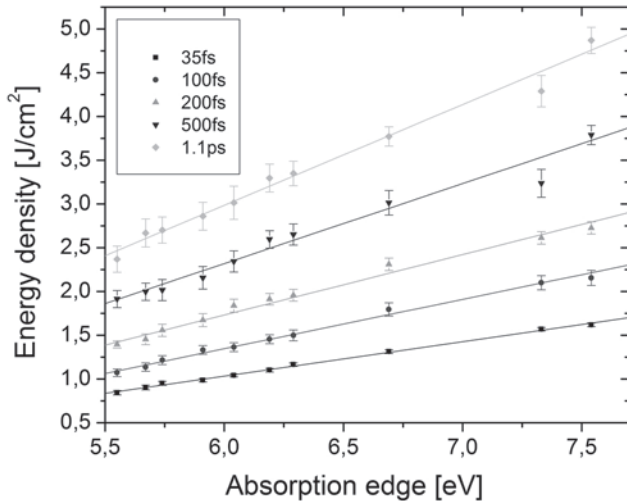


Figure 5: Damage threshold of single layer mixture coatings (hafnia/silica) of different material compositions depending on the pulse duration, fit according to scaling law [6].

Laser Damage of graded-index Mirrors with ns-Pulses

The potential of graded-index coatings has been investigated for a set of mirror coatings for 355 nm [7]. For the mirror coatings, different designs have been selected. As a reference, a standard quarterwave stack consisting of pure hafnia and silica layer was produced. To explore the influence of a certain amount of silica in the high-index material, quarterwave mirrors were manufactured with 95 %, 80 % and 70 % content of hafnia in the high-index hafnia-silica mixture layers. For achieving the same level of reflectivity, few pairs were added to the designs with 80 % and 70 % hafnia content. Finally, one mirror sample was designed with the refractive index step down concept (RISED), for which the refractive index in high-index layers of the quarterwave design is gradually decreasing for the topmost layers. This design concept which is described in detail elsewhere [8] is advantageous in the field for fs-pulsed applications.

The damage test was performed with a laser at 355 nm, 100Hz repetition rate and 6ns pulse duration. The test was evaluated for the 1-on-1 and 10.000-on-1 scenario [9]. In Figure 6, it can be seen that the lowest threshold was determined for the standard QW mirror with 100 % and 95 % content of hafnia in the high-index layer. The RISED mirror showed a slightly higher threshold compared the standard mirrors with 100 % and 95 % hafnia content. For the mirrors with 80 % and 70 % content, the threshold is clearly higher than for the other samples. Obviously, the positive effect of a higher content

of silica in the high-index material, which can be attributed to the lower absorptance in the mixture, overbalances the disadvantage of having more layers in the stack. This result shows the high potential for optimizing the damage resistance of coatings by the use of mixture materials.

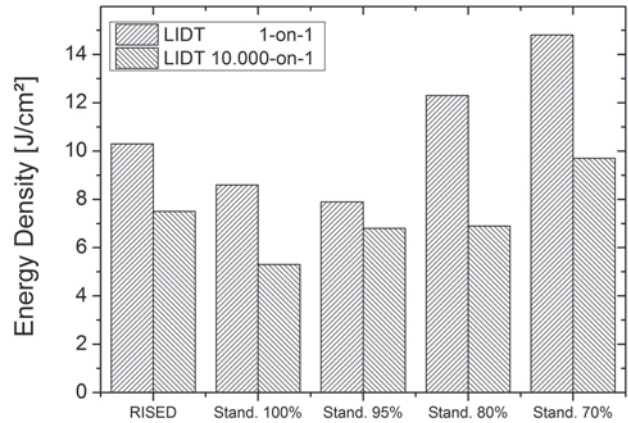


Figure 6: Damage threshold of 355nm mirrors with $Hf_xSi_{1-x}O_2$ mixtures as high-index material in the layer stack; standard QWOT mirrors with different contents of hafnia in the hafnia/silica high-index layers and RISED design variant.

CONCLUSIONS

In this publication, the principle of manufacturing graded-index coatings by using an adapted ion beam sputtering process was described. Several examples for the synthesis of single layer mixture layers or layers systems consisting of different oxide materials were demonstrated. The realization of graded-index coatings on a high level of precision is based on the exact knowledge of the material composition in regard to the lateral position of the zone target and on the implementation of an advanced optical monitoring technique. Moreover, the synthesis of material mixtures opens up new design possibilities due to the absorption edge shift effect. For several high-index materials this significant shift effect has been identified, if those materials were combined with silica. The spectral range, for which high refractive index materials are typically applied, can be distinctly extended to shorter wavelengths. As an example for high precision production of gradient index designs, a Rugate-mirror for 266 nm was manufactured in an automated coating procedure by using zirconia/silica mixtures with approx. 400 sub-layers with typical physical thicknesses of 4 to 6 nm. The power handling capability of oxide mixture coatings has been proven for high power laser applications in the near IR range as well as for fs- and ns-pulsed lasers. In the case of a gradient index AR coating on lenses for a laser processing objective, the material mixture coating withstood a power level of 10 kW at 1070 nm for more than 10 minutes. The damage investigations in single layer coatings using hafnia/silica mixtures with fs-pulses revealed that the damage mechanisms of those mixtures are comparable to pure materials. This leads

to the conclusion, that material mixtures form amorphous layers showing clear damage dependencies from the band gap energy. Finally, an example of a distinct increase of the laser-induced damage threshold for mirror coating at 355 nm was given. Compared to standard quarterwave mirrors produced with hafnia and silica, the blending of the high-index material with silica resulted in a significant increased damage resistance for ns-pulses. The presented studies show the potential of mixture coatings for extending the application bandwidth of typically used coating materials and for opening up new degrees of freedom for designing complex functional coatings, especially in laser technology.

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