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ANTIREFLECTIVE COATING FOR OPTICAL ELEMENTS BASED ON TeO_2

The technology of obtaining of a two-layer MgF_2 and ZnSe antireflective coatings for acousto-optic elements based on TeO_2 single crystals is proposed. The coatings were obtained for wavelengths 1063 nm and 405 nm with reflection coefficient $R_{1063} = 0.05 \div 0.1\%$ and $R_{405} = 0.5 \div 0.6\%$. Subsequent deposition of MgF_2 and ZnSe layers was carried out by the method of thermal evaporation in vacuum at a residual pressure of 10^{-3} Pa. The substrate temperature was maintained at 420K for MgF_2 and at 400K for ZnSe. The deposition rate for each material was ~ 1.2 nm/min. Coating thickness and deposition rate were controlled by the differential quartz crystal microbalance. The coatings have a high long-term stability of optical and mechanical characteristics. It is shown that the discrepancies between the calculated and experimental reflectance spectra are associated with imperfection of the film structures, mechanical stresses presence, inaccuracies in the geometric thicknesses of the layers. A slight shift and change of the R_{min} value in time can be attributed to the relaxation of the mechanical stresses in the films. The technology is based on easy accessible materials, does not require complicated vacuum equipment and provides reflection coefficients that meet modern requirements.

Keywords: thin films, antireflective coating, reflection coefficient.

1. Introduction

Acousto-optic modulators and deflectors based on TeO_2 single crystals are widely used in light beams control devices in modern scientific and applied instruments engineering. However, high refractive index (n = 2.25) of the TeO_2 crystals leads to large optical power losses in optical systems. To reduce the losses antireflective coatings are applied to the working surfaces of the elements. It makes possible in many cases to reduce optical losses significantly, lower the temperature of the working elements and increase their radiation strength. This is especially true in optical aiming (targeting) systems, coherent optics, spectroscopy, super-weak light detectors and so on.

The importance of antireflective coatings is clear from the following. According to the Fresnel's formulas reflection losses of normally incident light for a transparent material with refractive index n ~ 2.25 reaches 15%. It limits use of such materials in laser technologies as far as requires additional increase of radiation power to compensate the losses and leads to overheating of optical elements. Moreover, the reflected light undergoes series of subsequent reflections and creates a diffuse light background, which produces significant masking effect. Modern optical engineers have a large set of technological equipment [1, 2] and techniques to calculate and produce thin-film antireflective coatings. There is a big number of compounds for optical coatings which are mechanically-, chemically- and temperature-stable. However, the spectrum of refractive index values of these materials is limited. Thus, the task of choosing the proper coating material remains complex today. The use of simple in manufacturing single-layer antireflective coatings provides significant reduction of the reflection coefficient (R) of an optical system. It is known [1, 3] that the conditions of zero reflectivity for a single-layer coating formed on a transparent substrate are determined by the expressions:

$$n_f d_f = (2m+1)\frac{\lambda}{4}\ ,\ n_f = \sqrt{n_o n_s} \qquad (1)$$

where n_f – film refractive index, n_0 – falling beam medium refractive index

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(for air $n_0 = 1$), n_s – substrate refractive index, d_f – film geometric thickness, λ – incident light wavelength, m – any integer.

In accordance with (1) to meet the condition of zero reflection the optical thickness of the film must be an odd multiple of $\lambda/4$ and the refractive index should be strictly equal to the geometric mean of the refractive indexes of the surrounding media. Rigid demands for the accuracy of the n values do not allow achieving values of R that meet modern requirements ($R < 0.3\%$). According to [3], the reflection coefficient increases by approximately 1% when the refractive index of the film deviates from the optimal one by 10%. Such reflection losses are too great for modern multicomponent optical systems. The double-layer antireflective coatings do not demand such high accuracy of the films refractive index n values and extends the possibility of obtaining a minimum R for a wide range of wavelengths (λ). Combinations of the thicknesses and refractive indices of materials for two-layer coatings have been discussed in literature many times [1, 3]. From all the diversity of existing approaches we would like to single out the case where the layer with a higher refractive index is located outside and when the minimum of reflection is achieved under the following conditions:

$$n_{f1} \geq \sqrt{n_s}, \quad n_0 \leq n_{f2} \leq \sqrt{n_s} \qquad (2)$$

where n_{f1}, n_{f2} - refractive indices of the outer and inner layers respectively, n_0 - falling beam medium refractive index (for air $n_0 = 1$), n_s - substrate refractive index.

In this case it is not necessary to use materials with strictly defined refractive index values to achieve minimum reflection coefficient. For any pair of materials with refractive indices meeting not-strict conditions (2) it is possible to choose layer thicknesses so that the reflection coefficient will be close to zero for given wavelength. In addition, such coatings have several advantages. The total optical thickness of the two layers is less than a quarter of the wavelength. Such coatings have higher mechanical properties and high radiation strength, which is essential for IR devices.

Fig. 1 represents the comparison of the calculated reflection coefficient dependences of single-layer and two-layer coatings. The dependences are calculated by the FilmStar DESIGN free version program from FTG Software Associates.

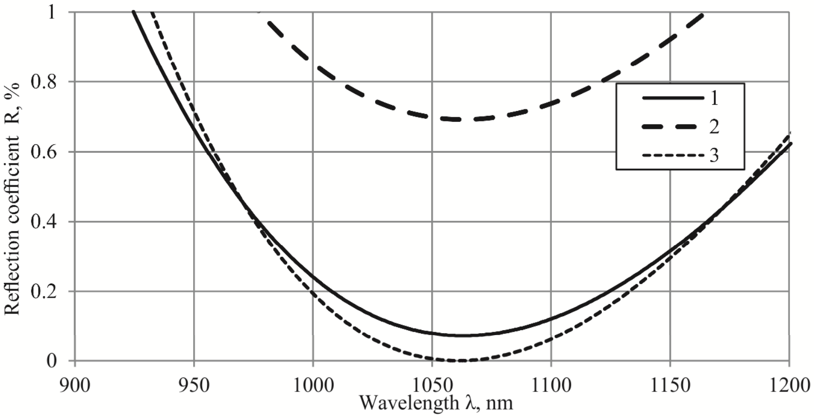

Fig.1. Comparison of the calculated spectral dependences of the reflection coefficient (R) for $\lambda = 1063$ nm for a TeO_2 crystal ($n_s = 2.25$). (1) single-layer coating ($n_f = 1.46$ 182.02 nm of SiO_2), (2) single-layer coating ($n_f = 1.38$ 192.6 nm of MgF_2), (3) two-layer coating ($n_{f1} = 2.3$ 19.8 nm of TiO_2 and $n_{f2} = 1.38$ 118.3 nm of MgF_2).

It can be seen from Fig. 1 that the behaviour of the reflection coefficient spectral dependence curve is similar for both two-layer coating and single-layer coating. The minimum values of the reflection coefficients for single-layer coatings are not zero because the condition (1) is not satisfied exactly.

2. Experimental setup

Oxide films such as, for example, SiO_2, TiO_2, Al_2O_3 are most often used as antireflective coatings. However, these materials have a high evaporation temperature (usually the electron-beam evaporation method is used) and the deposition of similar materials must be carried out in oxygen atmosphere. It greatly complicates the coating production technology. That is why oxygen free compositions were used. MgF_2 was chosen as the material with lower refractive index. It has a refractive index $1.37 \div 1.39$ (accounting dispersion), and has been successfully used for many years to brighten lenses. Powder ZnSe was chosen as the material with a higher refractive index. ZnSe films have refractive index of $2.45 \div 2.6$. The values of evaporation temperatures of these materials in vacuum make possible to use the resistive thermal evaporation method. Presence in VUP-5 of two resistive evaporators makes possible to obtain a two-layer antireflective coating in one vacuum cycle. Additional protection from moisture is not required as far as water solubility coefficient for MgF_2 – 0.0076 g/100 ml., and for ZnSe – 0.001 g/100 ml. Both MgF_2 and ZnSe in crystalline state are sufficiently hard substances (Knoop hardness values 415 kg/mm^2 and 112 kg/mm^2, respectively), that ensures good mechanical strength of the coatings.

MgF_2 and ZnSe powders were preliminarily manually grinded and then dried at 420K for 24 hours in air. It prevented spitting of powders from the tungsten evaporator during film deposition process. Optical element of TeO_2 was fixed in VUP-5 regular films deposition device (Fig. 2). Regular substrate holder was substituted by aluminum disk with a spring-loaded element holder installed. Regular substrate holder was substituted by aluminum disk with a spring-loaded element holder installed. A thin heat-conducting rubber strip was laid between the element and aluminum stoppers - heat exchangers. During films deposition, the element was heated by a regular IR heater (halogen lamps) and the coating thickness was controlled directly with a thickness gauge based on the differential Quartz Crystal Microbalance (QCM), whose operation principle was considered in [4]. The difference of frequencies was registered by a frequency meter.

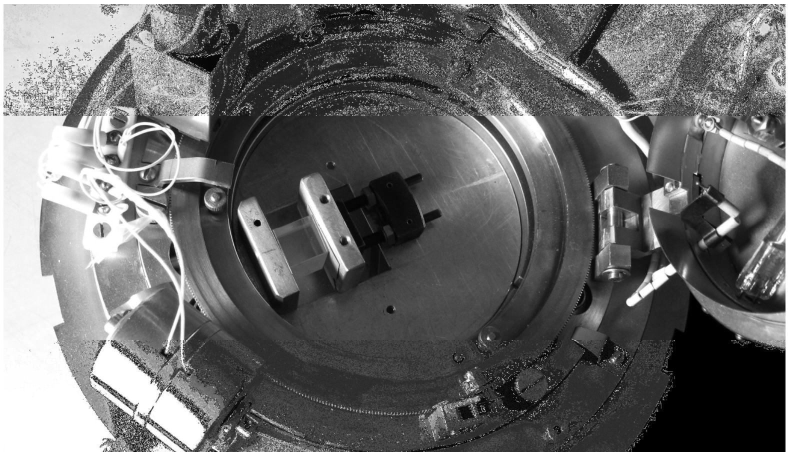

Fig. 2. VUP-5 regular films deposition device with an element holder

The change of frequency allowed controlling the deposition rate while absolute value of the frequency change was proportional to the thickness of deposited film.

Available programs for antireflective coatings calculation (for example, FilmStar Design free version) have a limited set of predefined coating materials and do not take into account the dispersion of the refractive indices. We have developed FilmCalc program which calculates reflection coefficients for multilayer coatings considering spectral dependence of the layers refractive indices. This program allowed us to select the optimal layers thicknesses and to obtain a minimum reflection at the required wavelength.

The chamber and sample were preliminarily ion-plasma treated in argon atmosphere for 20 minutes. Then the chamber was evacuated to residual pressure of 10^{-3} Pa, the holder with the sample was heated to 420K and the MgF_2 layer was deposited. After that the sample was cooled to 400K during 20 minutes and the ZnSe layer was deposited. After the depositions, the sample was cooled in vacuum to room temperature for 120 minutes. The reflection spectra of the TeO_2 elements with deposited coatings were measured using the spectrophotometers Specord M40 and Specord 61 NIR with a 3-mirrors reflection adapter (angle of incidence $< 7°$). Fresh-deposited Al mirror and internal data correction was used to calibrate the spectrophotometers. The measurement error (including errors of the sample position repeatability, aging of the mirror coatings and aging of correction data at the reflection minimum) did not exceed ±0.05%. The surface morphology was monitored by the Carl Zeiss Jenaval microscope.

3. Experimental results

During the trial deposition, it was noted that despite the high accuracy of the deposition process thickness control (±0.3 nm) there was a significant deviation of the obtained reflection coefficient spectra from the theoretical ones. By inverse calculation it was possible to establish that the observed discrepancies are related to the deviation of the refractive indices of the deposited films from the expected values. As noted in [1, 5], the substrate temperature and the deposition rate significantly affect the formation of the film structure. Our further studies showed that the element temperature of 420K was optimal for the deposition of MgF_2 films. The coatings were transparent and strong, with good adhesion and a refractive index close to expected $n = 1.38$ at $\lambda = 1063$ nm. The optimum element temperature for ZnSe deposition was 400K (the refractive index was close to $n = 2.5$). The optimal deposition rate for both materials was about 1.2 nm/min.

According to results of the FilmCalc program calculations the minimum reflection coefficient (R_{min}) at a wavelength of 1063 nm can be achieved when the thickness of the outer ZnSe layer is 15 nm, and the thickness of the inner of MgF_2 layer is 123 nm.

The reflection spectra of the resulting two-layer coating are shown in Fig. 3. Fig. 3 shows that the value of R at the wavelength of 1063 nm is about 0.1%. Some discrepancies between the calculated and experimental reflection spectra can be attributed to imperfection of the film structure and inaccuracies in the geometric thicknesses of deposited layers. Also, the presence of mechanical stresses in deposited films may lead to deviations of the layers refractive indexes. Measurements of reflection spectra of the same element repeated after 6 months showed some shift and decrease of the R_{min} value (Fig. 3, curve 2) which can be attributed to relaxation of mechanical stresses in the films. Above mentioned technique was applied to obtain two-layer antireflective coating on TeO_2 elements for $\lambda = 405$ nm. The calculations showed that for 2.9 nm ZnSe outer and 47.9 nm MgF_2 inner layers the reflection coefficient would reach its minimum value.

The reflection spectrum of such deposited two-layer coating is shown in Fig. 4.

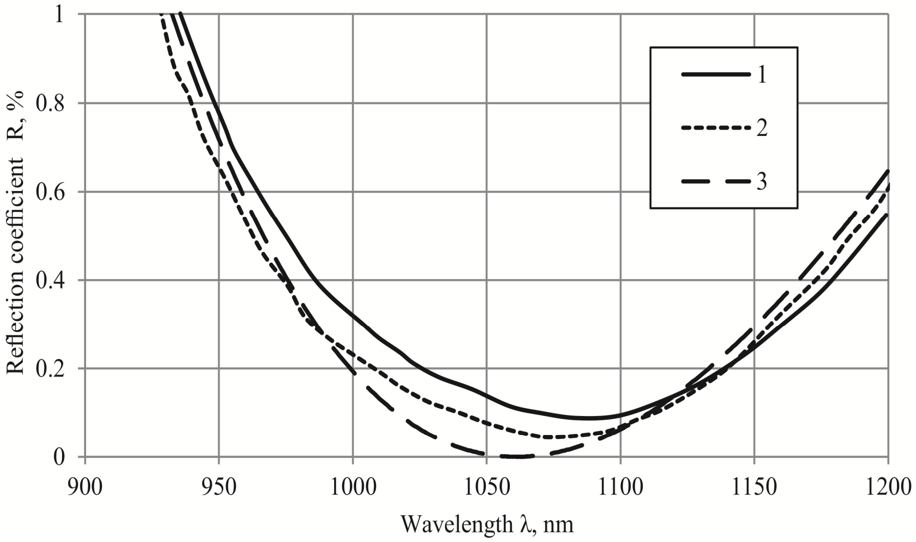

Fig. 3. Reflection spectra of a two-layer ZnSe/MgF_2/TeO_2 coating for λ = 1063 nm. (1) immediately after coating, (2) in 6 months, (3) calculated.

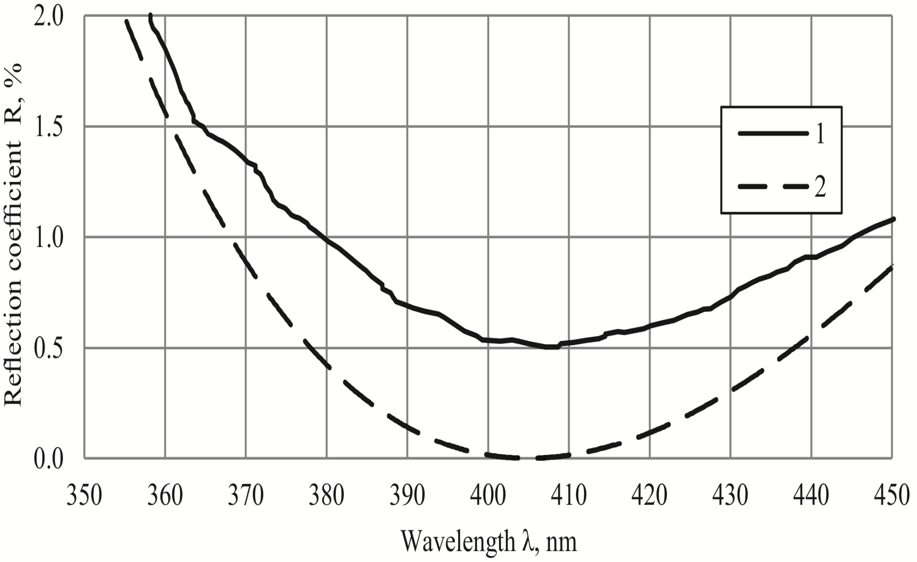

Fig. 4. Reflection spectra of a two-layer ZnSe/MgF_2/TeO_2 coating for λ = 405 nm. (1) fresh-deposited coating, (2) calculated spectrum.

Fig. 4 shows that obtained coating effectively reduces the reflection coefficient of the optical element at the target wavelength. The resulting value of R_{min} ~ 0.5% differs from the calculated one more significantly than for the 1063 nm case. This can be attributed to several reasons. The relative error of the film geometric thickness measurement increases for thinner layers and that leads to more significant deviations of the R values from the calculated ones. With such small ZnSe film thickness it is difficult to expect good continuity of the layer and the decrease of the continuity will lead to R value increase. In addition, the self-absorption edge of ZnSe (400 nm) was quite close to the target wavelength (405 nm) and it could also affect the R value.

Nevertheless, the spectral position of the reflection coefficient minimum (R_{min}) of the coating was close to the calculated one.

4. Conclusions

The calculation and deposition technique for two-layer antireflective coatings for acousto-optic elements is proposed. Optical coatings were obtained for wavelengths $\lambda = 1063$ nm and $\lambda = 405$ nm with reflection coefficients $R_{1063} = 0.05 \div 0.1\%$ and $R_{405} = 0.5 \div 0.6\%$. The oxygen free MgF_2 and ZnSe were used as materials for the antireflective coating.

The FilmCalc program developed by authors was used to preliminary calculations of the layers thicknesses. The calculations considered the spectral dependence of the refractive indices of the layers.

The discrepancies between the calculated and experimental reflection spectra of the coatings are associated with the films structures imperfection, presence of mechanical stresses in them, insufficient continuity of the layers and geometric thicknesses inaccuracies of the deposited layers. The R values can be affected by the proximity of the target wavelength to the coating materials self-absorption edge.

Stability investigations showed that storage of the samples for 6 months under laboratory conditions without special protective measures did not significantly influence the R_{min} values and the λ selectivity. Morphology investigation of the coating surface (magnification up to 1000 times) did not show detachments, opacities and cracks.

Developed technology is not difficult to repeat, is based on easy accessible materials, does not require complicated vacuum equipment and provides a reflection coefficient that meets the modern requirements for acousto-optic elements.

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