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The GdF_3/MgF_2 bilayer as an antireflective narrow-band ultraviolet filter

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The effective work of optical systems, applied in advanced optic and optoelectronic devices, requires antireflective coatings. In this work we focus on designing the bilayer system dedicated to work as a narrow-band filter within UV range of electromagnetic spectrum. Such coatings are applied in lasers where reduction in reflection to a small value at any single wavelength is needed. The bilayer system is based on GdF_3 and MgF_2 used as materials with middle and low refractive index. The multilayers were obtained by thermal evaporation on a highly purified CaF_2 substrates. The spectral dispersion of the refractive index of single layers has been determined by ellipsometric measurements. Thicknesses of single layers included in the bilayer system, aimed to work at specific wavelength, have been optimized based on optical characteristics simulation, including experimentally measured values of the refractive indices. During the deposition, layer thickness and deposition rate were controlled with Inficon XTC/2 thickness measuring system. Optical properties of obtained GdF_3/MgF_2 bilayer systems have been determined based on spectral dependences of reflectance and transmittance measured with the application of a spectrophotometer. The crystal structure and phase composition of the films have been examined by X-ray diffraction. The result of studies revealed that proper optimization of thicknesses of individual layers creating GdF_3/MgF_2 bilayer systems makes it possible to obtain the antireflective coating for desired wavelength of electromagnetic spectrum. The GdF_3/MgF_2 antireflective bilayer will be applied as a narrow-band filter for 238 nm irradiation produced by gas-ion/BBO crystal laser.

Keywords: thin antireflective films, GdF_3, MgF_2, UV filters.

1. Introduction

The application of antireflective coatings increases strongly nowadays [1]. They are used in different optical devices like monitors, mobile phones, cameras and other optical systems working mainly in a visible range of electromagnetic wave spectrum [2–4]. Due to rapid development of ultraviolet sources also filters and others optical elements for that range become more popular [5–8]. Especially they are used in technological applications like UV lithography in semiconductor industry [9–11]. Because the lasers generate the radiation with specific wavelength, the optical lenses used in optical systems require antireflective coatings dedicated to work as narrow-band filters. In such case, the bilayer systems formed from material with medium and low values of refractive indices are used. The reflectance characteristics form V-type shape for this type of filters.

The aim of our studies was to design a filter ensuring the lowest possible reflection at 238 nm, it means for gas-ion/BBO crystal laser. Therefore we had to choose materials which reveal high transmission in deep UV, however there are not many materials that meet these requirements. Usually as a material with medium refractive index the LaF_3 is used [12, 13]. The alternative choice could be the GdF_3 [5, 14, 15]. As a material with low refractive index, MgF_2 is commonly applied for the whole range of electromagnetic spectrum from UV to IR [13].

In this work we have developed GdF_3/MgF_2 bilayer system dedicated to work as a narrow-band filter for UV radiation with wavelength of 238 nm. Because the crystal structure of a material is dependent on the condition depositions [16], in our studies we have considered the dependence between deposition temperature, crystal structure and optical properties of obtained layers.

2. Experiment

The optical characteristics of antireflective multilayer coatings are dependent on the thicknesses and refractive indices of single layers creating the multilayer system. In turn, refractive indices of materials vary when the substrate temperature during the deposition is changed and hereby the optimal thicknesses of single are different. Therefore, theoretical simulations of optical characteristics of GdF_3/MgF_2 systems were carried out. During the simulation, experimentally determined refractive indices of applied materials have been taken into account.

For that reason, firstly the single layers: GdF_3 and MgF_2 were deposited. Both fluoride films were prepared by thermal source evaporation from molybdenum boats. Before the process, a deposition chamber was pumped down to a base pressure less than 5×10^{-5} mbar by a turbomolecular pump Ebara EBT1400. As a substrate of deposition the quartz glass, commercially available as Corning HPFS (commercial code for Fused Silica glass 7980) was used. Each type of layers was deposited three times. During the processes, all deposition conditions, except substrate temperature, were the same. The difference was that during the following depositions, the substrates were heated to 100, 200 and 300°C, respectively. The rate of deposition and thickness of the films were monitored and controlled by the quartz deposition controller Inficon XTC/2 [7]. The rate of the deposition was 0.4 and 0.5 nm/s for GdF_3 and MgF_2, respectively.

In order to find out refractive indices of the coating and determine dependences between their values and deposition temperature, the obtained single coatings were subjected to studies by means of spectroscopic ellipsometry. The measurements were performed with M-2000 ellipsometer produced by J.A. Woollam Co. Inc., in the spectral range of 190–1700 nm. The ellipsometric spectra have been analyzed using Complete EASE 5.0 software. Based on the optical characteristics simulations, the optimal thicknesses of the coatings belonging to GdF_3/MgF_2 bilayer, ensuring the lowest possible reflection and the highest transmission of a multilayer, were defined.

Then, the deposition of GdF_3/MgF_2 coatings was carried. The bilayer filters were deposited onto three types of the substrates: quartz glass (Corning HPFS), CaF_2 crystal and B270 glass. Samples obtained on B270 glass were used to the examination of crystalline structure of particular layers included in the bilayer system by means of X-ray diffraction. X-ray diffractometry (XRD) patterns were measured with a PANalytical X'pert Pro MPD X-ray diffractometer. The performance of the bilayer systems was evaluated by the measurement of the optical transmittance and reflection in the range of 200 to 400 nm. Optical characteristics were measured with the application of a double beam spectrophotometer Shimadzu UV1601.

3. Result and discussion

3.1. Designing of antireflective filters

In order to design high quality antireflective coatings for specific wavelength, bilayer dielectric filters are applied. Performance of such filters is determined by the refractive indices of applied materials and layers thicknesses. However, the refractive index of the material is not a constant value. Its values are different when the crystalline structure of material is varying. In turn, crystalline structure of material depends on the deposition condition.

In carried out research we studied the influence of substrate temperature on the structural and optical layer properties. Therefore, the unmodified HPFS glass and the single layers of GdF_3 and MgF_2 deposited on the HPFS glass, prepared at three different substrate temperatures, were subjected to measurement by means of spectroscopic ellipsometry (SE). The ellipsometry is a well-known and very useable technique for examination of thin films allowing to determine the thickness of the film d, its refractive index n, and extinction coefficient k. Layer properties are determined based on a change in polarization when light interacts with layered materials [17, 18]. The measurement is recorded as two values related to the reflectance ratio of p- and s-polarized light, given as an amplitude ratio $\tan(\Psi)$ and phase difference Δ, that are linked as follows [17]:

$$\rho = \tan(\Psi)\exp(i\Delta) = \frac{R_p}{R_s} \tag{1}$$

To know the physical properties of studied layers, the theoretical model of dispersion of the refractive index has to be fitted to experimental data, Ψ and Δ. First, we fitted the theoretical model to HPFS glass substrate alone. The dispersion of the refractive index of HPFS quartz glass is shown in Fig. 1. Then, for single layers: GdF_3 and MgF_2, the theoretical models of dispersion were fitted to spectral dependences of

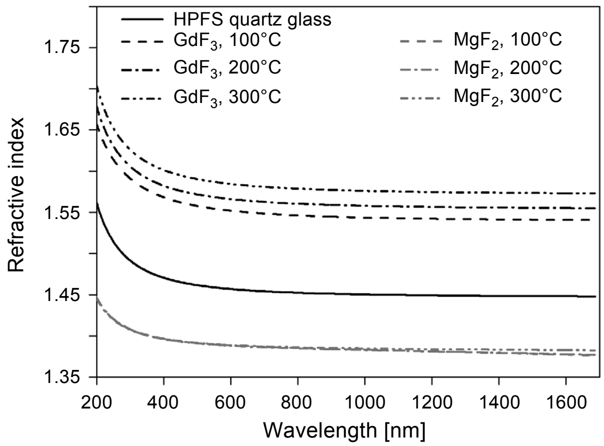

Fig. 1. The dispersion of refractive indices of HPFS quartz glass and single layer GdF_3 and MgF_2 obtained at different substrate temperatures: 100, 200 and 300°C.

ellipsometric angles. To analyze the data, all angular spectra have been fitted simultaneously. For both fluoride layers, Cauchy model dispersion, described by the following equation was applied:

$$n^2(\lambda) = B + \frac{C}{\lambda^2} + \frac{D}{\lambda^2} \tag{2}$$

where B, C, D are material-dependent specific constants.

In such a way, the dispersion of refractive indices of single layers obtained at different substrate temperatures was determined (Fig. 1). Comparing refractive indices of studied layers, it turned out that changing of substrate temperature during the deposition only slightly influences the properties of MgF_2, but in the case of GdF_3 the increase in substrate temperature causes a significant improvement of its refractive index.

The determined values of refractive indices were applied to optical characteristics simulations in order to designate optimal thicknesses of single layers, ensuring the lower reflectance and the highest transmittance of the bilayer system at specific wavelength of electromagnetic spectrum. Optimization process was carried out in two stages, separately for each material and each substrate temperature. During simulation it was assumed that the thickness of MgF_2 layer was constant and the thickness of GdF_3 layer was varied within the range of 30–45 nm (Fig. 2). Comparing simulated optical characteristics, it was turned out that the lowest reflectance equal to 0.65% could be achieved for GdF_3 layer thickness of 36 nm.

Similar simulations were carried out to adjust the optimal value of thickness of MgF_2 layers (Fig. 3). The best result was achieved for MgF_2 layer thickness of 42 nm. All obtained spectral reflectance characteristics have V-shape, typical for bilayer systems (Figs. 2 and 3). Based on the performed simulations, it was found that the stack

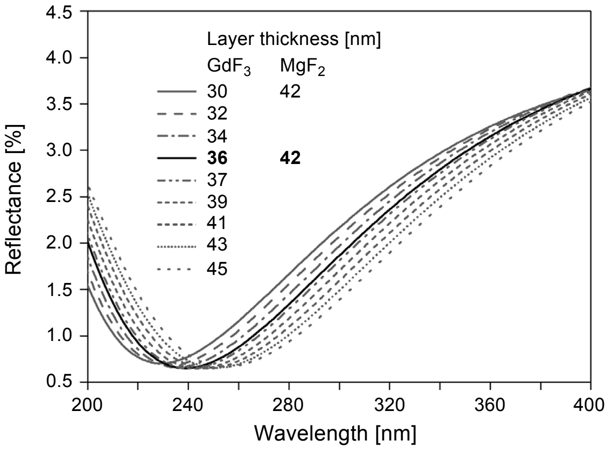

Fig. 2. The simulated reflectance spectra of HPFS glass/GdF_3/MgF_2 bilayer system consisting of MgF_2 layer of 42 nm, and GdF_3 layer with varied thickness from 30 to 45 nm.

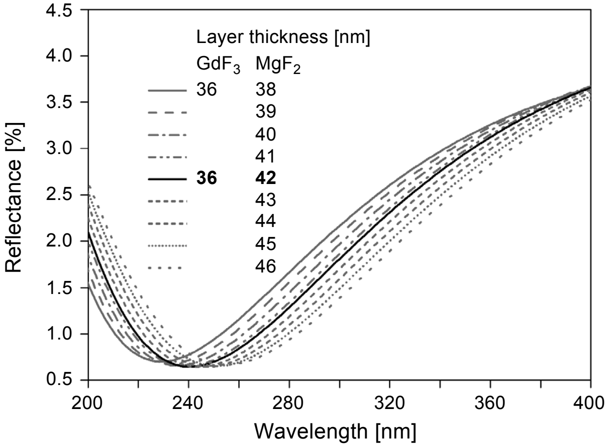

Fig. 3. The simulated reflectance spectra of HPFS glass/GdF_3/MgF_2 bilayer system consisting of GdF_3 layer of 36 nm, and MgF_2 layer with varied thickness from 38 to 46 nm.

of the above materials deposited onto the substrate heated to temperature of 100°C should have physical thickness of 36 and 42 nm, for GdF_3 and MgF_2, respectively.

In the same way, optimal thicknesses of layers deposited on HPFS glass substrate heated to 200 and 300°C were determined. For MgF_2 layer the optimal value of thickness was the same (42 nm) for each substrate temperature, what is clear due to the fact that refractive indices of MgF_2 also were very close for different substrate temperatures. In the case of GdF_3, obtained at a substrate heated to 200 and 300°C, the optimal value of thickness should be lower and equal to 35 nm. However, by comparison of calculated optical characteristics of bilayer stacks with optimal values of thicknesses,

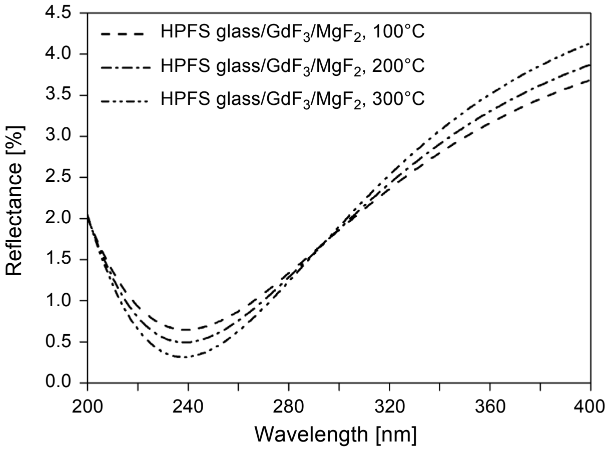

Fig. 4. The comparison of simulated optical characteristics of HPFS glass/GdF_3/MgF_2 bilayer systems with optimal values of thicknesses of particular layers obtained at a substrate heated to 100, 200 and 300°C.

it can be seen that layers obtained at highest substrate temperature reveal the best optical properties (Fig. 4). An increase in the substrate temperature from 100 to 300°C reduces the reflectance at 238 nm from 0.65% to 0.32%.

3.2. Structural properties of GdF_3/MgF_2 bilayer systems

Structural analysis and phase composition of bilayers were determined by XRD. XRD patterns (Fig. 5) show that the GdF_3/MgF_2 layers are poorly crystallized regardless of the substrate temperature. Nevertheless, low intensity signals occurring in

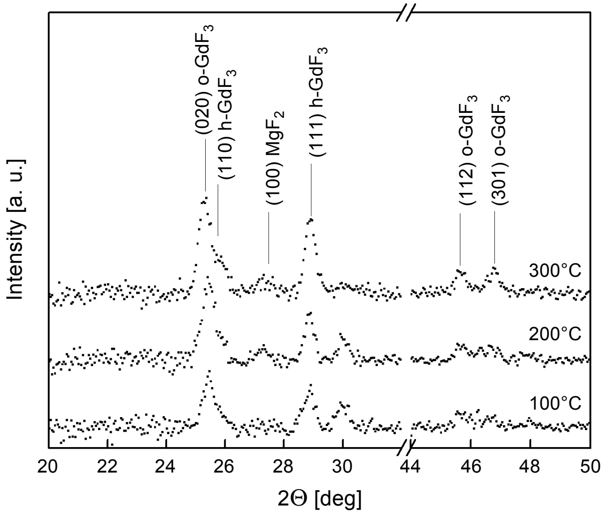

Fig. 5. The XRD patterns of GdF_3/MgF_2 antireflective coatings deposited on substrates heated at different temperatures.

XRD patterns prove the presence of the crystal phase of tetragonal MgF_2, orthorhombic and hexagonal GdF_3. Moreover, based on the detailed analysis, it could be noticed that the intensity of signals assigned to GdF_3 increased almost twice, when the substrate temperature was changed from 100 to 300°C, and the intensities of signals associated with MgF_2 were comparable. These results are in accordance with the results of measurement obtained by means of spectroscopic ellipsometry (Fig. 1). An increase of GdF_3 refractive index was caused by the crystallinity improvement, while in the case of MgF_2 the rise in substrate temperature had no impact on the crystalline structure, so the values of refractive indices are the same (Fig. 1).

3.3. Optical properties of GdF_3/MgF_2 bilayer systems

The quality and performance of obtained GdF_3/MgF_2 layers, aimed to work as a narrow -band filter for wavelength of 238 nm, were evaluated by the analysis of their optical characteristics. Measurements of spectral dependence of transmittance and reflectance

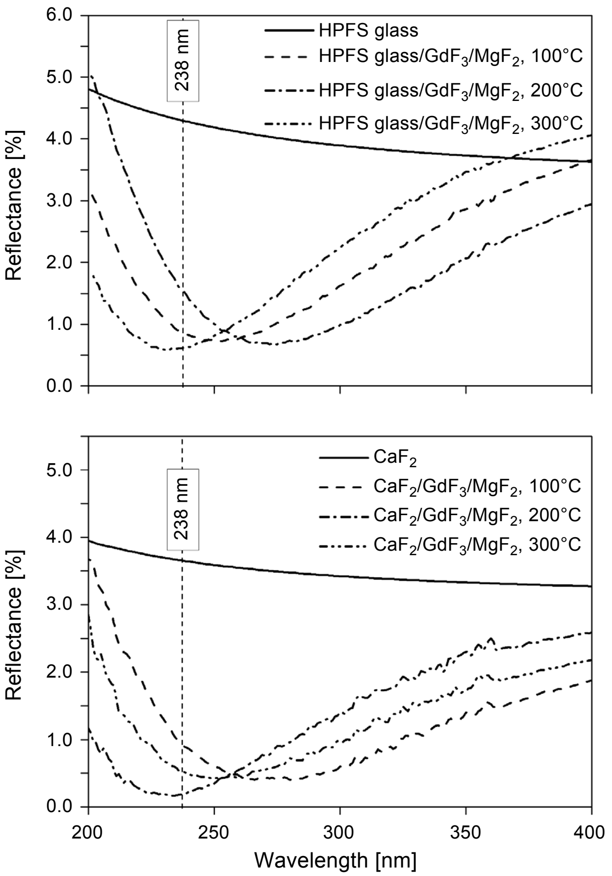

Fig. 6. The reflectance spectra of uncovered substrates and substrates covered with GdF_3/MgF_2 antireflective coatings deposited on a substrate heated to different temperatures.

in the wavelength range between 200–400 nm with a mean accuracy of 1 nm were performed for uncovered HPFS quartz glass, CaF_2 substrate and for samples with GdF_3/MgF_2 films obtained at different substrate temperatures.

The analysis of reflectance spectra (Fig. 6) revealed that in the case of samples deposited on HPFS glass, the lowest reflectance equal to 0.62% at wavelength of 238 nm was achieved by the bilayer system obtained in a substrate heated to 300°C, but it was not the minimal value of reflectance for this system. The minimal value of reflectance was equal to 0.58% at 231 nm. The GdF_3/MgF_2 layers obtained at other substrate temperatures are characterized by higher reflectance and their minima are placed at higher wavelength. The bilayer system obtained on quartz glass heated to 100°C achieved reflectance equal to 0.73% at wavelength of 253 nm, while for layers deposited at 200°C minimal reflectance had a value of 0.68% at 275 nm.

In case of bilayers obtained on a CaF_2 substrate for the wavelength of 238 nm, the best results were obtained by samples prepared on a substrate heated to 200°C. Re-

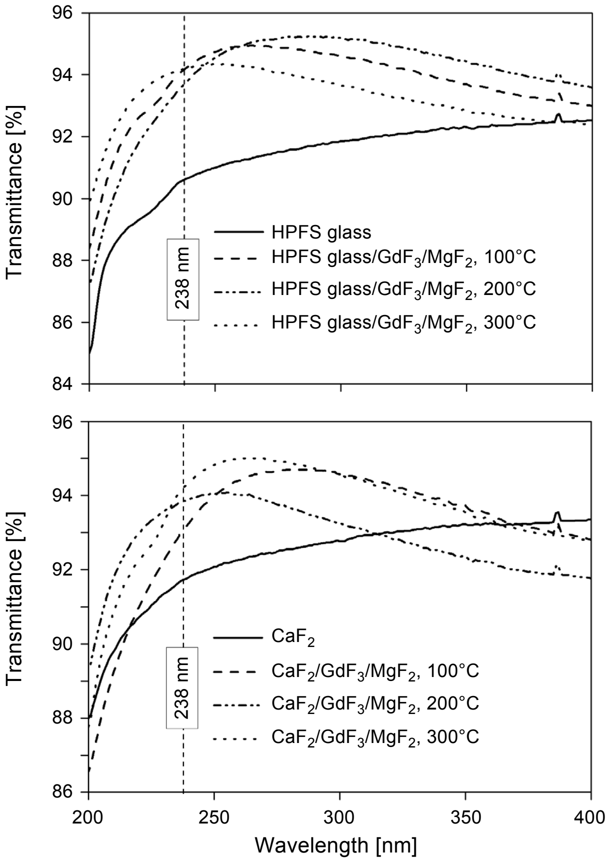

Fig. 7. The transmittance spectra of uncovered substrates and substrates covered with GdF_3/MgF_2 antireflective coatings deposited on a substrate heated to different temperatures.

flectance for this film at 238 nm was equal to 0.18%, whereas for layers obtained at 300°C and 100°C it was 0.52% and 0.90%, respectively.

For all obtained bilayer systems, spectral transmittances were also measured within the range of 200–400 nm. Based on the transmittance spectra for layers deposited on HPFS quartz glass, presented in Fig. 7, it was revealed that all layers improve the transmittance of quartz glass but the best performance characterizes the GdF_3/MgF_2 system obtained on a substrate heated to 200°C. However, the maximum of transmittance is shifted to higher wavelengths than 238 nm.

For the wavelength of 238 nm, the highest transmittance equal to 94.2 was achieved by the bilayer system obtained at a substrate heated to 300°C. In case of bilayers obtained on a CaF_2 substrate, the situation was similar. For the wavelength of 238 nm, the best results were obtained by the sample prepared on a substrate heated to 300°C, however it is not the maximal transmittance, the maximal value of transmittance was equal to 95.02% at 269 nm. For other bilayers, the maximal values of transmittance were lower and also were shifted to higher wavelengths.

For all obtained bilayer systems, the maximal values of transmittance were assigned to higher wavelength than 238 nm, that was caused by the fact that the actual values of thicknesses were different from optimal thicknesses determined by the optical characteristics simulation. It could be caused by unintentional variations of a deposition rate. Some divergences could be also caused by some heterogeneity of the layer thickness.

4. Conclusions

The aim of the studies was to design the narrow-band filter dedicated to gas-ion/BBO crystal laser generating irradiation of wavelength equal to 238 nm. The antireflective coating was created by application of MgF_2 – as a material with a low refractive index and GdF_3 as a material with a medium refractive index. The bilayers were deposited by thermal source evaporation on CaF_2 and a quartz glass substrate. During the studies the influence of substrate temperature during the deposition on optical and structural properties was considered. Based on the X-ray diffraction it was revealed that the rise in substrate temperature causes the increase in the volume of a crystalline phase of GdF_3 but it does not affect MgF_2 crystalline structure. These results were confirmed by measurements of refractive indices of single layers performed by means of spectroscopic ellipsometry. Refractive indices of MgF_2 layers deposited on substrates heated to different temperatures were almost the same, whereas the refractive index of GdF_3 layer improved when the substrate temperature was increased. Measured values of refractive indices of single layers were used to optical characteristics simulation design to determine optimal thicknesses of single layers forming the bilayer system. Moreover, based on the simulated spectral dependences of reflectance, it was found that by an increase in substrate temperature from 100 to 300°C the reflectance at 238 nm can be reduced from 0.65% to 0.32%. Such dependence was not observed in experimentally measured reflectance and transmittance spectra but it was caused by the differences between actual and optimal values of thicknesses of single layers. The evidence of the mismatch in layers thicknesses was also the shifting of reflectance minimum and transmittance maximum with respect to wavelength of 238 nm. Thought the minimal reflectance of applied GdF_3/MgF_2 coating was not placed at 238 nm, the obtained results confirmed that such bilayer system could be successfully used as an antireflective coating for UV region of electromagnetic spectrum, however, the precise control of a deposition condition is required.

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