LASER APPLICATIONS AND OTHER TOPICS IN QUANTUM ELECTRONICS

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Development and study of dielectric coatings with a high radiation resistance

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Abstract. **Technological methods for applying dielectric coatings on glass substrates are proposed and studied, which substantially enhanced the radiation resistance of the coating to irradiation by nanosecond pulses. A rapid method for measuring the radiation resistance of optical elements by using an array of Gaussian laser beams is described.**

Keywords: *neodymium glass laser, coating deposition technology, dielectric mirror, radiation resistance.*

1. Introduction

In the 1990s, the development of new-generation, high-power laser systems – the 2-MJ NIF [1] and LMJ [2] facilities and the 300-kJ Iskra-6 facility [3] (the output energies are given at 337 nm) was initiated in a number of leading laser laboratories. At present a module of the laser system Iskra-6 – the Luch facility is built [4, 5]. The output energy density in the laser channel of these facilities is ~ 10 J cm^{-2} for 3 – 5-ns pulses. The optical elements of such facilities should have the warranted radiation-resistance margin exceeding the output energy density (10 J cm^{-2}) by a factor of 1.5 – 2 [6].

The results of testing the radiation resistance of optical elements developed for the Luch and Iskra-6 facilities in 2001 – 2003 are presented in paper [7]. Below, we discuss the investigations devoted to the manufacturing of dielectric coatings with a high radiation resistance, which were performed at the Luch Research and Production Association and the Institute of Laser Physical Research, Russian Federal Nuclear Center, All-Russian Scientific Research Institute for Experimental Physics (ILFI RFNC-VNIIEF). We also describe a rapid method for measuring the radiation resistance and present the results of testing dielectric coatings manufactured at the Luch RPA.

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2. Fabrication of dielectric coatings with a high radiation resistance

One of the factors preventing the development of high-power pulsed lasers is a low radiation resistance of dielectric coatings. Most of the studies of the effect of the structure of laser coatings, their material, and different technological factors of their deposition have been performed in the in the spectral range between 1.05 and 1.06 μm. Based on these investigations, a number of problems can be indicated that should be solved to obtain the maximum radiation resistance:

(i) the choice of appropriate materials;

(ii) the provision of a high quality of a substrate surface;

(iii) the use of the optimal deposition technology providing the preparation of a non-absorbing coating without defects.

In this spectral range, the oxides of different metals and semiconductors have long been successfully used (TiO_2, ZrO_2, HfO_2, Ta_2O_5, SiO_2, etc.). The maximum radiation resistance at 1.06 μm was obtained for the Ta_2O_5/SiO_2, HfO_2/SiO_2, and ZrO_2/SiO_2 coatings, the HfO_2/SiO_2 coatings having the highest threshold resistance.

The manufacturing technology of high-quality polished substrates is well developed and at present it does not prevent fabricating highly reflecting interference laser mirrors with a high radiation resistance. However, when anti-reflection (AR) coatings are used, a charged near-surface layer substantially reduces the radiation resistance. This is obviously explained by the fact that in this case all the energy of laser radiation is incident on the substrate surface, and the smallest defects of the surface such as particles of a polished compound embedded into the substrate surface and microscopic cracks begin to play a noticeable role. Numerous studies reliably confirmed the dependence of the radiation resistance of the substrate surface and AR coatings on the polishing methods, abrasives, and surface cleaning after polishing [8].

Japanese researchers reported [9] that the depth of penetration of a polished compound into the substrate surface does not exceed 100 nm and there exists an efficient method for increasing the radiation resistance of the substrate surface and AR coating by several times – the method of deep ion etching. Studies [8] and [9] were performed by using UV lasers. To verify the applicability of the results of these papers to the 1.05 – 1.06-μm spectral range, we investigated the dependences of the radiation resistance of AR coatings on the thickness of a layer removed from the substrate surface upon ion etching before

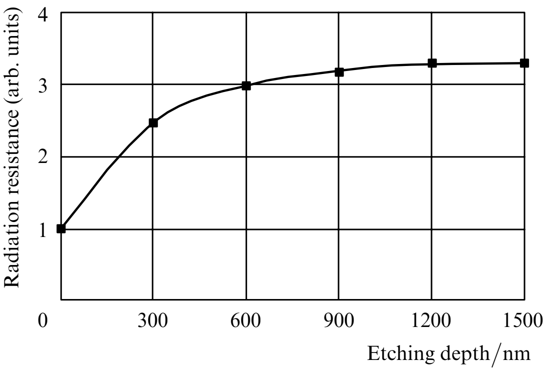

Figure 1. Dependence of the radiation resistance of AR coatings on the depth of substrate ion etching before coating deposition.

their deposition. The results of our studies are presented in Fig. 1.

The thickness of the surface layer of the substrate removed by ion etching was measured by variations in the frequency of a quartz resonator with a SiO_2 layer deposited on it, which was located near the substrate, and also directly by using a broadband photometric control system. The radiation resistance of coatings is presented in arbitrary units because we could not measure absolute values with our equipment. For this reason, only comparative tests were performed. We found that the ion etching of a substrate by the depth 100 – 150 nm before the deposition of the AR coating enhanced the radiative resistance of the latter by 1.5 – 5 times, depending on the polishing method and subsequent chemical preparation of the substrate. The positive effect of ion etching was also observed upon the deposition of output mirrors with low reflection coefficients; the lower was the reflection coefficient of the deposited mirror, i.e., the smaller was the number of layers, the greater effect was achieved.

The optimal coating deposition technology should provide in the general case the deposition of a non-absorbing coating without defects. It is known that most of the metal oxides have the phase transition at the temperature slightly below the melting temperature, at which the density abruptly changes. For example, the phase transition of hafnium monoxide from the monoclinic to tetragonal phase, which occurs at the temperature about 1700 °C, is accompanied by the change in its volume by 3.8 %. The characteristic evaporation temperature is 2200 °C. Therefore, under the evaporation surface there exists a surface with a constant temperature 1700 °C, from which microparticles are ejected when the specific volume of its material changes. Hafnium or zirconium oxide stabilised by yttrium does not have a phase transition and is successfully used for applying coatings with a high radiation resistance [10].

If we assume that the number of defects is proportional to the layer thickness, then multilayer coatings should contain a greater number of defects and, therefore, will have a lower radiation resistance. To obtain the reflectivity 99.5 % for the ZrO_2/SiO_2 coatings by using ZrO_2 with the refractive index $n = 1.95$, 21 layer should be used. If, however, the ZrO_2 layers with $n = 2.1$ are deposited, only 17 layers are required. That is why the technology should provide the deposition of materials with the maximum possible refractive index. This condition was fulfilled in the ion-assisted deposition of coatings by using a modernised End-Hall ion source. The bombardment of a growing oxide film with a 200 – 250-eV beam of oxygen ions (by using a mixture of oxygen with heavy noble gases) allowed us not only to increase the refractive index and, therefore, the film density up to the values typical for the corresponding bulk materials, but also to decrease absorption in the coating by more than an order of magnitude. Figure 2 shows the dependence of the refractive index of a ZrO_2 film on the content of xenon in an oxygen ion beam [11].

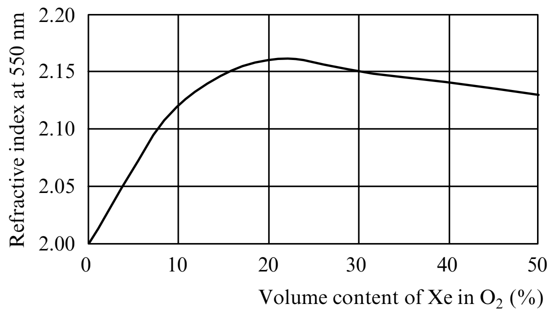

Figure 2. Dependence of the refractive index of a ZrO_2 film on the xenon content in oxygen.

The average ion energy and the average ion current density in the ion beam incident on the substrate were 190 eV and 200 μA cm^{-2}, respectively. One can see that the refractive index is maximal for the xenon content of about 20 % and then decreases with increasing the xenon content, which is accompanied by the increase of absorption in the film.

The increase in the refractive index of a highly reflection material results in turn in a decrease in the number of layers required to obtain the specified refractive index, and in a decrease in the number of defects, thereby improving the radiation resistance of the coating. Moreover, the action of the ion beam in combination with a low temperature of the substrate during the ion-assisted deposition of coatings forms layers with the structure close to amorphous, which also improves the radiation resistance of the coating.

Based on the above considerations, the HfO_2/SiO_2 dielectric mirrors and AR coatings were manufactured at the Luch RPA for the radiation-resistance testing at the ILFI, RFNC-VNIIEF.

3. Method for rapid radiation-resistance measuring

The radiation resistance of optical elements is studied, as a rule, by a classical method [12], when a sample is irradiated by one Gaussian beam with the effective diameter 1 – 2 mm. To determine the damage threshold of the sample, it is necessary to perform several shots, and when wide-aperture samples are studied, the radiation resistance should be measured at several points, which results in the increase in the number of experiments and in the measurement time.

We developed the method for rapid measuring the radiation resistance of optical elements in which a sample was irradiated not by one laser beam but by an array of Gaussian laser beams (the intensity envelope of the laser beams and individual laser beams in the array have the intensity distributions close to Gaussian). The damage threshold was determined by this method in most experiments for one shot with an accuracy of $\sim 10\,\%$.

An array of Gaussian beams was formed in the following way (Fig. 3). A diffraction-limited laser beam with the aperture diameter 12 mm is incident on a grid aperture in the form of a 7×7-mm square containing a copper wire grid (the wire diameter was 0.7 mm and the grid period was 1.4 mm). Laser radiation transmitted through the grid aperture is incident on a spatial-angular selecting square 12×12-mm aperture located at a distance of ~ 4.3 m from the grid aperture. The wire diameter and the grid step were chosen to provide the efficient spatial-angular selection of radiation at an acceptable distance without considerable energy losses. The image of the grid aperture formed in this way is projected on the sample.

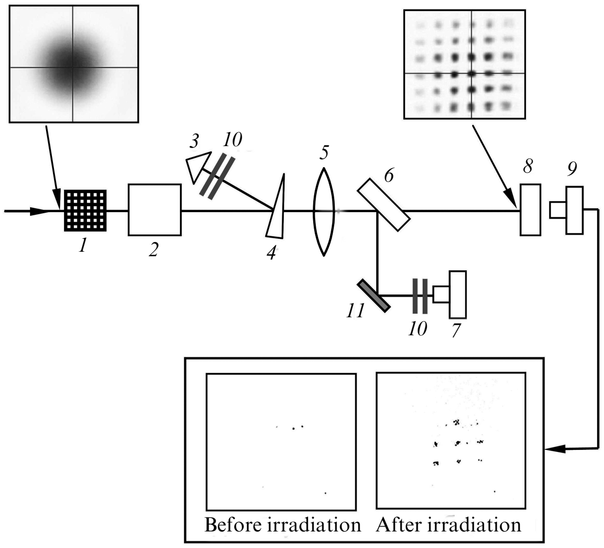

Figure 3. Optical scheme of an array of Gaussian beams: (*1*) grid aperture; (*2*) spatial-angular selecting diaphragm; (*3*) calorimeter; (*4*) wedge; (*5*) lens; (*6*) plane – parallel plate; (*7*) CCD camera; (*8*) sample; (*9*) microscope with a CCD camera; (10) optical filters; (11) dielectric mirror.

To compare our method with the classical method, we performed test measurements of the radiation resistance of a K8 glass. The laser radiation energy incident on a sample was measured in these experiments with an EPM1000 power meter with a J50LP-2 pyroelectric detector (Molectron). The incident radiation intensity distribution was measured with a WinCamD CCD camera (Gentec). The camera was located at the same distance from the lens as the sample. The sample damage was recorded with a microscope equipped with a CCD camera. The images of the sample surface were recorded before and after laser irradiation. The damage criterion was the appearance of irreversible structural changes on the sample surface after laser irradiation, which were observed with the CCD camera.

The energy damage threshold of samples was characterised by the maximum energy density of one beam, which was defined as

$$\varepsilon = E/S_{\text{eff}},$$

where E is the energy of one beam; $S_{\text{eff}} = \pi d_{\text{eff}}^2/4$ is the area of the equivalent laser spot with the uniform distribution of the energy density equal to the maximum energy density in a real spot containing the same energy; and d_{eff} is the diameter of the equivalent laser spot. The maximum energy density of one of the beams was calculated by using a special program written in Matlab, which was used to process CCD-camera images.

The laser radiation parameters in test experiments were as follows: the radiation wavelength $\lambda = 1.054$ μm, the pulse duration $\tau_{0.5} = 3.2$ ns, the effective beam diameter in the array $d_{\text{eff}} = 0.2$ mm (the 6×6 array of 3×3-mm size); and the effective beam diameter in the classical method $d_{\text{eff}} = 1.4$ mm. The typical distributions of the laser radiation intensity on a sample obtained by different methods are presented in Fig. 4.

One can see from Fig. 4a that the developed optical scheme allows us to transform the initial diffraction-limited beam so that the intensity envelope of laser beams in the array and individual laser beams will have the Gaussian intensity distribution (coincidence with the Gaussian distribution was obtained after processing of the 8-bit image presented in Fig. 4a).

From the laser radiation intensity distributions on a sample and its damage pattern, two beams were determined – one with the maximum energy density at which the sample damage was not observed yet, and another with the minimum energy density at which the damage was detected. The threshold energy density of the sample damage is the average value of these two energy densities. The difference of the intensities of the beams near the sample damage threshold was, as a rule, $\sim 10\,\%$.

The radiation resistances of glass samples obtained by these two methods coincide within ± 2 J cm^{-2} (about 8 %) and are equal to 26 J cm^{-2}. Therefore, test experiments showed that the rapid method can be used for determining the radiation resistance of optical elements.

This method was applied for measuring the radiation resistance of optical elements fabricated at the Luch RPA (Table 1). One can see from Table 1 that dielectric mirrors (with reflectivity higher than 97 %) have a high radiation resistance – up to 32 J cm^{-2}. AR coatings have the radiation resistance 20 and 15 J cm^{-2} upon irradiation from the coating and glass sides, respectively.

4. Conclusions

We have studied experimentally technological methods of deposition of dielectric coatings with the minimal absorption and maximum possible refractive index and density. This provided the increase in the radiation resistance of the HfO_2/SiO_2 coating up to 30 J cm^{-2} and higher for 1.054-μm, 3-ns laser pulses.

The rapid method is developed for measuring the radiation resistance of optical elements by using arrays of Gaussian beams. The accuracy of measuring the damage threshold energy density of coatings for one shot was $\pm 10\,\%$.

The radiation resistance of mirrors and AR coatings fabricated at the Luch RPA was measured to be up to 32 and 20 J cm^{-2}, respectively.

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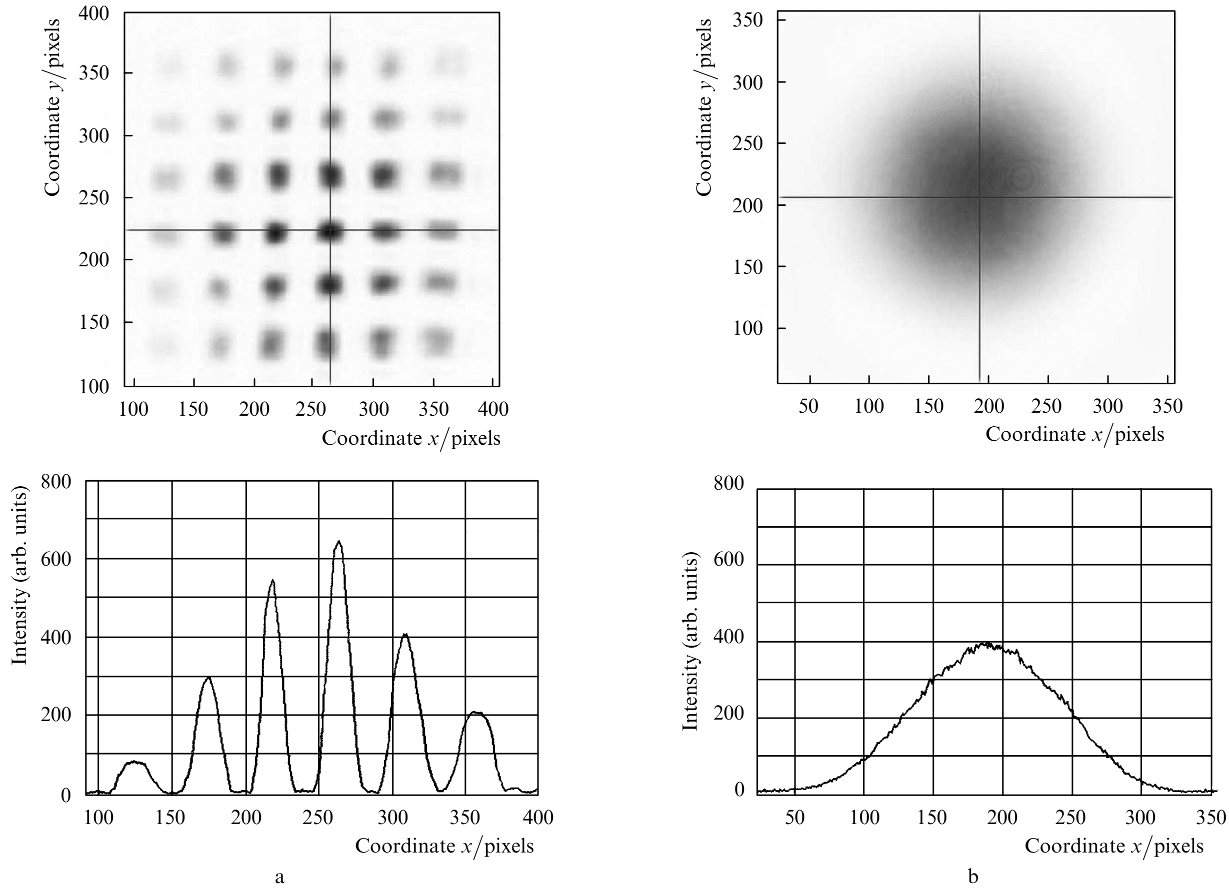

Figure 4. Typical laser radiation intensity distributions on a sample obtained by the rapid (a) and classical (b) methods.

Table 1. Radiation resistance of dielectric coatings measured in the paper.

Sample	Radiation resistance/J cm^{-2}	Irradiation conditions
Dielectric mirror 1	32.0 ± 1.0	Irradiation from the coating side
Dielectric mirror 2	29.5 ± 2.5	Irradiation from the coating side
AR coating	20.0 ± 1.0	Irradiation from the coating side
	15.0 ± 2.0	Irradiation from the glass side

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