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Effect of KrF laser radiation on electron-beam-induced absorption in fluorite and quartz glasses

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***Abstract*. The behaviour of high-purity fluorite and KU-1, KS-4V and Corning 7980 quartz glasses exposed to an electron beam and KrF laser radiation is studied. Exposing these materials to $\sim$ 5-MW cm^{-2} KrF-laser radiation both during the action of the electron beam and after it reduces their residual absorption at least by a factor of 1.5.**

Keywords: *optical materials, KrF-laser radiation, ionising radiation, radiation resistance.*

1. Introduction

Quartz glasses and fluorite are the basic materials for fabricating the windows of high-power KrF lasers and other electron-beam-pumped excimer lasers. The efficiency and hence the possibility of using these lasers depends to a considerable extent on the radiation resistance of the materials of these windows subjected to the action of the electrons scattered by the pump beam, X-rays and high-intensity laser radiation [1 – 4]. Problems of radiation resistance also appear during operation with synchrotron radiation, in resonator optics of free-electron lasers [5, 6] and in reactor windows in laser fusion [7, 8]. All this points towards the necessity and importance of studying the potentialities of optical materials (OMs) subjected to the action of various types of ionising and laser radiation.

By studying the outlook for using an electron-beam-pumped KrF laser as a driver for laser fusion, we analysed the behaviour of a number of modern quartz glasses and high-purity CaF_2 crystals under prolonged action of electron beam pulses [2, 3, 9]. The present study is a continuation of this research. Our aim was to study the effect of KrF-laser radiation on the electron-beam-induced residual absorption in previously studied KU-1, KS-4V and Corning 7980 glass samples as well as in high-purity CaF_2. The experiments were planned in such a way that the obtained results could throw light on the processes occurring in OMs under the action of an electron beam and laser radiation, and could serve as the basis for refining their models.

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2. Radiation resistance of quartz glasses exposed simultaneously to an electron beam and KrF-laser radiation

It was shown in [2, 3, 9] that after prolonged exposure of quartz glass samples and high-purity fluorite to an electron beam their transparency is reduced to a new quasi-stationary level. But the windows of electron-beam-pumped lasers are exposed not only to ionising radiation consisting of electrons scattered from the pump beam and X-rays, but also to laser radiation. The behaviour of OMs subjected to such a complex action is interesting not only from a practical point of view but for scientific research also. In this section, we present the experimental results that provide an answer to this question for quartz glasses exposed simultaneously to an electron beam and KrF laser radiation.

The experiments were performed on the ELA setup (in our earlier investigations [10, 11], the EL-1 setup was used). Under normal operating conditions, the ionising radiation energy density per pulse at the laser chamber window was $F_1 \approx 0.1$ J cm^{-2} [1]. For such a density and under normal pulse repetition rate of the setup (5×10^{-3} Hz), the highest induced absorption does not exceed 9 % even in KU-1 samples, which has the lowest radiation resistance among all investigated quartz glasses. For such a value of F_1, the induced absorption does not exceed 2 % – 3 % in the KS-4V quartz glass, which is close to the sensitivity limit of spectrophotometers. To increase the ionising-radiation-induced absorption, it was necessary to increase F_1.

For this purpose, permanent magnets were installed in the ELA laser chamber. Their magnetic field, perpendicular to the propagation direction of the electron pump beam rotated a part of this beam towards one of the output windows of the laser. Calorimetric measurements of the electron beam energy density showed that in this case the value of F_1 increases to 0.4 J cm^{-2} on the window surface. The distribution of the electron beam energy density over the free surface of the window was quite uniform, as shown by the impressions on a vinyl plastic plate used instead of the window in adjustment experiments. The plate was then replaced by the output window made of the OM under study. A plane resonator mirror with $R_b = 100$ % at 248 nm was placed in the second laser window. The output plane resonator mirror with $R_o = 44$ % was mounted outside the laser chamber near the output window.

A specific feature of this experiment is that an additional diaphragm was installed near the mirror with $R = 100$ % inside the laser chamber to cover half the laser beam. As a result, lasing occurred over one half of the circular cross-section of the optical aperture of the laser. Only half of the OM used for the laser window was exposed to laser radiation while the electron beam fell on the entire material. A comparison of induced absorption in each half of the window after a series of shots on the ELA setup showed the difference between absorption in the part exposed only to the electron beam and the part exposed to the electron beam and laser radiation. All the other differences in experimental factors (window material and conditions of irradiation) which could affect the induced absorption were eliminated in this case.

The KrF laser used in experiments operated on the $Ar - Kr - F_2$ gas mixture with the ratio $Ar : Kr = 10 : 2$ and the F_2 pressure not exceeding 0.005 atm in an overall pressure of 1.1 atm. Under such a pressure, the average density of the electron beam energy at the window was 0.4 J cm^{-2}. The difference in the densities of the laser gas mixture and the atmosphere in which the energy of the electron beam was measured near the window in calibration experiments was compensated by placing an additional aluminium foil filter in front of the calorimeter.

Because the window being studied was located inside the laser cavity, the total laser radiation intensity E_w and, hence, the laser energy density on the window was the sum of the intensities of forward and backward beams. For $R_b = 100\,\%$, the quantity E_w is related in this case with the output laser energy density E_o by the expression

$$E_w = E_o(1 + R_o)/(1 - R_o). \tag{1}$$

For $R_o = 44\,\%$, we have

$$E_w = 2.57E_o.$$

The output energy density was measured with a BKDM calorimeter in each shot. The quantity E_o was determined by dividing the average output laser energy over the entire series of pulses with the window being studied by the cross-section area of the output laser beam. The laser radiation intensity at the window was calculated from the expression

$$I_w = E_w/\tau, \tag{2}$$

where $\tau = 80$ ns is the laser pulse duration.

This technique was used to study five samples of quartz glasses, including three Corning 7980 samples (Standard Grade – C-0, KrF grade – C-KrF, ArF grade – C-ArF), one KU-1 sample and one KS-4V sample. Because the diameter and thickness of the Corning samples were 52 mm and 10.5 mm, a small intermediate flange was used for mounting them on the window of the laser chamber. The laser-beam diameter was reduced in this case from the conventional 52 mm to 42 mm. For the KU-1 and KS-4V samples, whose diameter and thickness were 60 mm and 17 mm, respectively, the laser-beam diameter was 52 mm.

The window samples being investigated operated in the laser chamber for 6 – 7 days during which about 400 shots were fired. The total energy density F of the electron beam at the surface of these windows achieved ~ 160 J cm^{-2} during this period. The average energy density E_w of the KrF laser radiation per pulse on the exposed half of the window in these experiments was ~ 0.3 J cm^{-2} for an intensity $I_w \approx 4$ MW cm^{-2}. The absorption spectra of each half of the window were recorded with a Spectronics spectrophotometer in the interval 200 – 1000 nm within one hour and one day after the last shot.

Figure 1 shows the typical transmission spectra of one of the investigated samples (C-ArF) before and after exposing to the electron beam only as well as before and after exposing to the electron beam and laser radiation simultaneously. One can see that the region exposed to the electron beam and laser radiation has a higher transmittance than the region exposed to the electron beam only. Such a difference was observed in the entire visible spectral range for $\lambda > 350$ nm where no absorption is induced by the electron beam [2, 9]. Apparently, this effect is associated with laser cleansing of the surface. It was observed in all the quartz samples investigated by us.

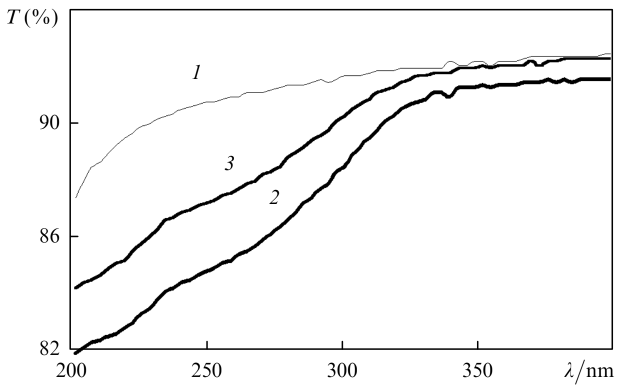

Figure 1. Transmission spectra of the corresponding halves of the Corning 7980 glass sample (ArF grade – C-ArF) before [curve (*1*)] and after exposure to the electron beam [curve (*2*)], as well as for simultaneous exposure to the electron beam and KrF laser radiation [curve (*3*)].

The transmission spectra of the samples exposed to the electron beam only (E) and to the electron beam and laser radiation simultaneously (L) [curves (*2*) and (*3*) in Fig. 1] were recalculated by points by the expression

$$\mathrm{OD} = \ln(T_0/T) \tag{3}$$

to the optical density spectra (Fig. 2). One can see that in the case of the simultaneous action of the electron beam and laser radiation, the induced optical density is much lower. We describe this difference with the help of the coefficient

$$K_{\mathrm{EL}} = \frac{(\mathrm{OD}_{250} - \mathrm{OD}_{400})_{\mathrm{L}}}{(\mathrm{OD}_{250} - \mathrm{OD}_{400})_{\mathrm{E}}}, \tag{4}$$

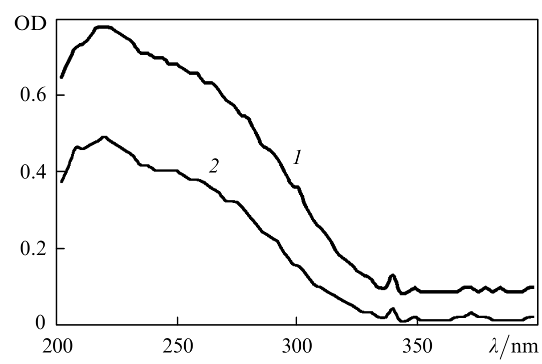

Figure 2. Optical density spectra for halves of the C-ArF sample exposed to the electron beam [curve (*1*)], as well as for simultaneous exposure to the electron beam and KrF laser radiation [curve (*2*)].

where OD_{250} and OD_{400} are the optical density at $\lambda = 250$ and 400 nm respectively. Expression (4) allows us to eliminate the uncertainty related to the choice of the zero level of optical density for sample surface regions with different levels of purity.

Table 1 presents the coefficients K_{EL} obtained in these experiments for all quartz glass samples. The average value of K_{EL} for all samples is 1.5. The error level does not exceed 20 %. Hence, within the error of measurement, the value of K_{EL} for all glass samples investigated by us is the same and amounts to 1.5. In other words, the absorption induced by ionising radiation in the windows of electron-beam KrF lasers using the quartz glass samples investigated by us will be 1.5 times lower than in the case of exposure to ionising radiation only.

Table 1.

Type of glass	K_{EL}
C-0	2
C-KrF	1.32
C-ArF	1.5
KS-4V	1.55
KU-1	1.2

Analogous experiments with samples of high-purity fluorite crystals were not performed because for exposure of the samples to electron beams in the experiments, the absorption induced in CaF_2 does not exceed 1 %, which is close to the sensitivity threshold of the spectrophotometer. Hence, it is virtually impossible to reveal the difference in absorptions for various irradiation regime.

3. Annealing of electron-beam-induced absorption in OMs by KrF laser radiation

Absorption induced in quartz glasses by ionising radiation under the simultaneous action of ionising and KrF laser radiation may decrease for two main reasons. The first one is the decrease in the production of long-lived defects in the glass during each pulse of such a combined action of radiation. The second reason is the laser-induced annealing of defects produced during the previous ionising radiation pulses. Separate experiments involving the action of ionising radiation followed by exposure to laser radiation may reveal the main reason behind the decrease in absorption in OMs.

In the course of prolonged experiments aimed at studying the behaviour of OMs under the action of an electron beam [2, 3, 9], we have collected a set of samples irradiated by various fluences and having a considerable absorption in the UV spectral region. Some of these samples were used for studying the effect of KrF laser radiation on the residual absorption induced by an electron beam.

Two KU-1 glass samples (No. 2/2 with $F = 18.7$ kJ cm^{-2} and No. 2/3 with $F = 3.2$ kJ cm^{-2}) and two KS-4V glass samples (No. 1, $F = 20.6$ kJ cm^{-2} and No. 2, $F = 4.1$ kJ cm^{-2}) were used in our experiments. Their exposure on the ELA setup was completed on March 1, 2004 [9]. Relaxation of the electron-beam-induced absorption was studied for these samples for about one and a half years. In addition, we also studied three samples of Corning 7980 glass (C-0, C-KrF and C-ArF) exposed to an electron beam with $F = 6.4$ kJ cm^{-2}. The electron beam exposure of these samples was completed by July 5, 2005. The relaxation of the induced absorption was studied in these samples for about two months and then experiments on laser-induced annealing were started.

Samples were irradiated by an EMG 150 MSC electric discharge excimer laser (Lambda Physik). The energy density produced by a 20-ns pulse from the KrF laser at the samples was about 0.1 J cm^{-2}. The pulse repetition rate in a train of pulses was 5 or 10 Hz. After exposure with the required overall fluence F_L (~ 100 J cm^{-2} as a rule), the sample transmittance was measured on a spectrophotometer. About two weeks elapsed between successive series of sample irradiation.

Figure 3 shows a typical family of transmission spectra after various series of KrF laser exposures of KU-1 sample No. 2/2 (chosen as an example) with the corresponding values of fluence shown for each curve. One can see that, as the fluence increases, the transmission spectra converge to a certain limit, i.e., the transmission attains a new quasi-stationary level.

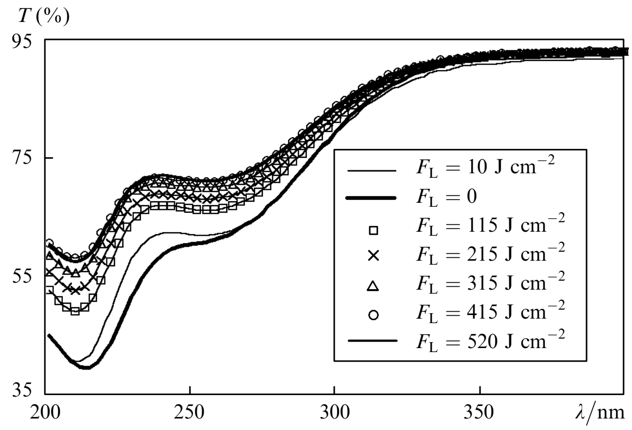

Figure 3. Transmission spectra of KU-1 sample No. 2/2 before and after exposure to KrF laser radiation.

To simplify the analysis of processes occurring in the samples, we recalculated the transmission spectra to the optical density (OD) spectra by using expression (3). (Note that Figs 3–6 also show the corresponding spectra of the samples before the beginning of laser-induced annealing.) Figures 4 and 5 show some selected OD (F_L) spectra after laser irradiation of KU-1 sample No. 2/2 and KS-4V sample No. 1, respectively. (The value of F_L in J cm^{-2} is shown in parentheses after OD in Figs 4 and 5 and subsequently in the text.)

One can see from Figs 3 and 4 that for small F_L ($\lesssim 10$ J cm^{-2}), the absorption band of the KU-1 samples is burnt. The corresponding spectrum [OD(0)–OD(10)] is shown by the dashed curve in Fig. 4. This band has a maximum at 226 nm and its FWHM is 23 nm. It was also observed in KU-1 sample No. 2/3 and in the Corning samples. This band, which is typical of type III glasses, probably belongs to the surface centres, which have not been observed experimentally so far [12]. After burning out of this band, absorption over the entire UV region decreases proportionally with increasing F_L, as follows from the constant ratio OD(520)/OD(115) in the interval 200–330 nm in Fig. 4. A similar behaviour of absorption

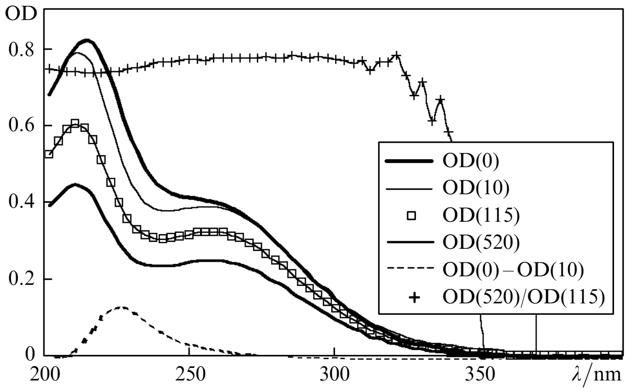

Figure 4. OD (F_L) spectra of KU-1 sample No. 2/2 for various values of F_L.

with increasing F_L was also observed in Corning 7980 glass samples.

The KS-4V samples also have a band that is rapidly burnt out by the KrF laser radiation, but it has a slightly different shape (dashed curve in Fig. 5). This band was obtained by subtracting from the curve OD(0) the curve OD(10) multiplied by the coefficient $k = OD_{200}(0)/OD_{200}(10)$, which ensured the coincidence of the tails of the corresponding spectra. The resultant band with a plateau in the interval 226-250 nm apparently consists of several 'elementary' bands. As F_L was further increased, absorption in KS-4V samples as in other glasses, decreased uniformly over the entire UV region and achieved a quasi-stationary level.

Figure 6 shows the experimental dependences of OD_{250} on F_L of the 248-nm KrF laser radiation for all the quartz glass samples studied by us. These dependences clearly show that the absorption induced in quartz samples by KrF laser radiation decreases to a new quasi-stationary level.

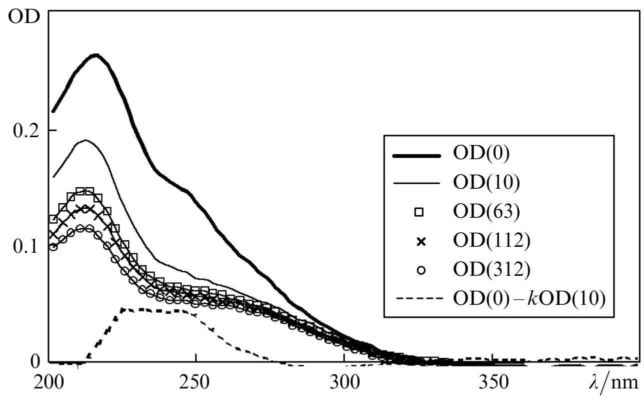

Figure 5. OD(F_L) spectra of KS-4V sample No. 1 for various values of F_L.

Let us describe this decrease by the coefficient

$$K_L = [OD_{250}^{max} - \Delta OD_{250}^{S}]/OD_{250}^{min}. \quad (5)$$

Here, OD_{250}^{max} is the maximal optical density of the sample at 250 nm before the beginning of its exposure to laser radiation; ΔOD_{250}^{S} is the change in OD_{250} due to burning out of the surface absorption; and OD_{250}^{min} is the minimum of OD_{250} after irradiation of the sample with the maximal fluence. Table 2 presents the coefficients K_L for all the investigated quartz samples.

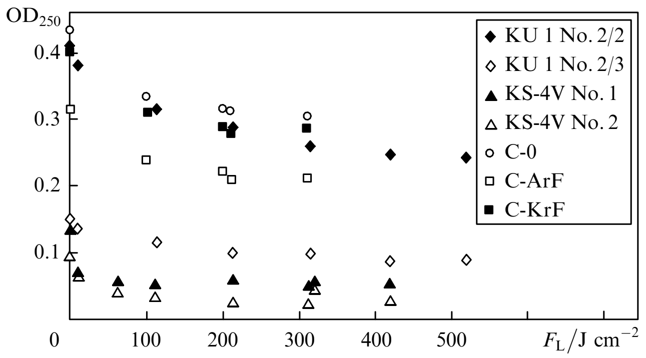

Figure 6. Dependence of OD at $\lambda = 250$ nm on F_L for all the investigated quartz glass samples.

Table 2.

Sample	K_L
KU 1 No. 2/2	1.7
KU 1 No. 2/3	1.7
C-0	1.5
C-KrF	1.4
C-ArF	1.5
KS-4V No. 1	1.8
KS-4V No. 2	2

Note that the values of K_L for the Corning glass samples were obtained for $F_L = 300$ J cm^{-2}, and hence they are slightly lower than the corresponding values for KU-1 samples. Because of the smallness of OD_{250}^{min}, the error in determining K_L for KS-4V samples is about double the error for other samples in which it does not exceed 10 %. One can see that the average value of K_L for all the samples studied by us is 1.7. All the values of K_L obtained by us coincide with this value to within 10 %. Hence, the KrF laser radiation can be used to burn out UV bands of the absorption induced in quartz glasses by ionising radiation to a level that is at least 1.7 times lower than the initial level.

The slight difference in the values of coefficients K_{EL} and K_L indicates that the decrease in the induced absorption in quartz glasses under the simultaneous action of an electron beam and laser radiation is due to laser burn out of long-lived centres formed during previous pulses, and not due to a change in the efficiency of their formation.

Similar experiments were performed with three high-purity CaF_2 samples out of the six samples irradiated by an electron beam on the ELA setup with an overall fluence o 26.2 kJ cm^{-2} [3, 9]. The interval between the end of exposure of these samples to the electron beam and the beginning of experiments with laser radiation was 38 days. This made it possible to neutralise the effect of natural relaxation of induced absorption on the results of experiments with laser radiation.

Figure 7 shows the transmission spectra of one of the CaF_2 samples (sample No. 4) before and after exposure to KrF laser radiation with an overall fluence of 50, 150 and 250 J cm^{-2}. It is hard to draw any conclusion from these spectra about the effect of laser radiation on the induced absorption because of the systematic error introduced by a parallel displacement of the transmission spectra during measurements on a spectrophotometer [9].

As in [9], we used the dip ΔD in transmission spectra in the F-centre absorption region at $\lambda = 379$ nm for a quantitative description of variations occurring in CaF_2 samples

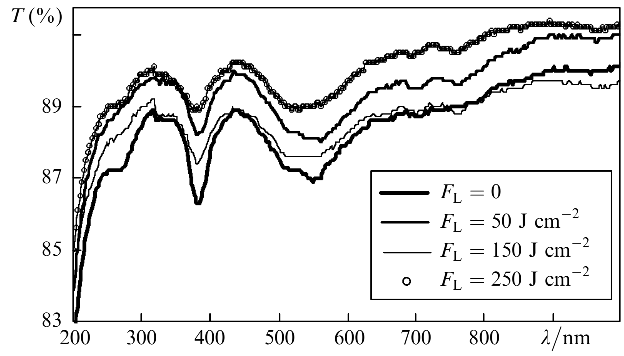

Figure 7. Transmission spectra of CaF_2 sample No. 4 before and after exposure to laser radiation.

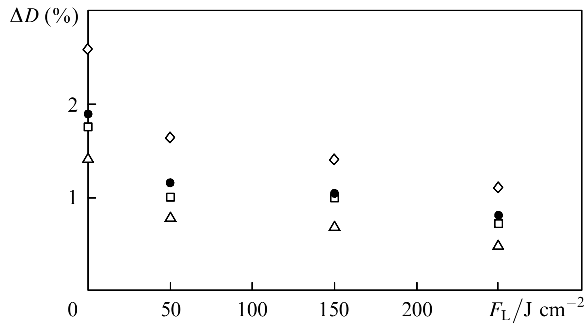

Figure 8. Dependence of ΔD at $\lambda = 379$ nm on F_L for CaF_2 samples No. 4 (◇), 12 (□) and 14 (▽), as well as ΔD averaged over three values (●).

exposed to laser radiation. In the present case, this dip is defined as

$$\Delta D = [(T_1 - T_m) + (T_2 - T_m)]/2. \quad (6)$$

Here, T_m is the transmission for $\lambda = 379$ nm, and T_1 and T_2 are the maximum values of transmission from the left and right of the dip. Using this quantity as a quantitative parameter of the induced absorption in CaF_2, we can eliminate the systematic error introduced by a parallel displacement of the spectra during measurements [9].

Figure 8 shows the experimental dependences of ΔD on F_L for three CaF_2 samples investigated by us (samples No. 4, 12 and 14), as well as the values of ΔD obtained by averaging over three values. These dependences show that the electron-beam-induced residual absorption at the intrinsic colour centres in CaF_2 decreases under the action of KrF laser radiation with an intensity of about 5 MW cm^{-2}. This decrease can be defined quantitatively through the coefficient

$$K_C = \Delta D_{max}/\Delta D_{min}. \quad (7)$$

Here, ΔD_{max} and ΔD_{min} are the values of ΔD before exposure of the sample to laser radiation and after exposure to laser radiation with the maximum F_L. For average values of ΔD in Fig. 8, the value of $K_C = 2.4$. Note that this is the case for samples with noticeable traces of the oxygen impurity [3]. In extremely pure samples of CaF_2, the value of ΔD decreases to about half even without laser radiation exposure during three days after the termination of exposure to the electron beam [9]. Exposure of such samples to KrF laser radiation with an intensity of about 5 MW cm^{-2} accelerates the relaxation of the absorption induced by the electron beam.

4. Conclusions

The experimental results obtained in this study show that prolonged exposure of KU-1, KS-4V and Corning 7980 quartz glasses to KrF laser radiation with an intensity of about 5 MW cm^{-2} lowers the ionising-radiation-induced quasi-stationary absorption by a factor of at least 1.5 both at the instant of exposure to ionising radiation and afterwards on all their bands in the UV range. This effect is manifested even more strongly in high-purity samples of CaF_2.

The effects of annealing of the ionising-radiation-induced quasistationary absorption under the action of KrF laser radiation in the OMs revealed in our experiments raise hopes of increasing the service life of windows of high-power electron-beam-pumped KrF lasers intended, among other things, for use as drivers in laser fusion. Among all the investigated materials, Russian quartz glass of type KS-4V was found to be the most suitable material for large windows of such lasers. The effects studied here improve the resistance of high-purity CaF_2 crystals to electron beams and X-rays, which even otherwise have a high stability to such radiation.

The entire body of the experimental data on the effect of ionising and laser radiation on modern high-purity OMs presented in this work will help in improving the understanding of the physics of radiative processes occurring in such materials. These results will also be useful for manufacturers of OMs and designers of high-power electron-beam-pumped excimer lasers.

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