## Nonlinear coloration and damage of wide gap glasses by femtosecond laser at 0.85 µm

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# **ABSTRACT**

The processes arising in glasses as an example of solid dielectrics under high-power femtosecond laser radiation have been investigated. In the investigation samples of fused silica were used as well as a wide selection commercial borosilicate glasses K8 (Russia) and BK7 (USA) which have the band gap near 6 eV. The glass samples were irradiated with the output of a laser system allowing the production of laser pulses at 0.85  $\mu$ m wavelength and duration ~ 10<sup>-13</sup> s. It is found that color centers formation and intrinsic luminescence of borosilicate glasses are observed under high-power femtosecond radiation at 0.85  $\mu$ m. These processes result from supercontinuum generation in the bulk glass and are followed by two-photon absorption of the short-wavelength part of this supercontinuum. These same processes lead to the damage of glasses under conditions of intensive ionization of material matrix, and show that the damage process is not connected with avalanche ionization.

Keywords: borosilicate glasses, femtosecond laser radiation, color centers formation, intrinsic luminescence

## **<u>1. INTRODUCTION</u>**

It is well known that the refractive index variations of condensed media arising under highpower laser radiation with pico- and femtosecond duration result in changes of the amplitude, phase and frequency of these laser pulses. These effects induce substantial spectral broadening of the transmitted radiation beams, both in the long-wavelength and in the short-wavelength regions ("white" light generation) due to self-phase modulation, induced-phase modulation, or cross-phase modulation (e.g., [1], [2] and references therein). The first works devoted to the observation of "white" light generation in crystals and glasses were performed in 1970 [3,4]. It was found that from five to ten small-scale filaments occurred under picosecond pulse excitation at 0.53  $\mu$ m. The irradiance in these filaments could achieve  $10^{13}$  W/cm<sup>2</sup>. In this case intensive radiation in wide spectral diapason from 0.4 to 0.7  $\mu$ m was observed. In subsequent works the analogous effects were obtained at other wavelength [5,6], induced spectral broadening of weak picosecond pulse in glass was demonstrated [7], the reasons of temporal form change was explained [8], and the main mechanisms of this process was understood [1,2].

The phenomenon of generation of "white" laser pulses has found wide application in different areas: for the production of the ultrashort pulses [9], for time-resolved absorption

spectroscopy [10], in fiber diagnostics [11], and in other applications (e.g., [12]). However, use of this phenomenon has a limitation because the converted radiation has a high intensity in wide spectral region. It can lead to nonlinear absorption, and color centers formation. The last process will result in the change of absorption coefficient [13-15] and the refractive index [16] of medium, that is the formation of optical inhomogeneities. In this connection the present work goal is to study the processes arising in glasses as an example of solid dielectrics under high-power femtosecond laser radiation.

#### 2. EXPERIMENTAL

In the investigation samples of fused silica were used as well as a wide selection commercial borosilicate glasses K8 (Russia) and BK7 (USA) which have the band gap near 6 eV. The mechanisms of nonlinear absorption in such glasses were studied earlier [13,14]. It was found that two-photon glass matrix ionization occurs under laser radiation exposure of glasses with  $E_g/2 < hv < E_g$  leading to color centers formation and to fundamental luminescence. It should be emphasized that in this case the effect of color centers formation was clearly registered at an intensity  $I = 10^{-3} \cdot I_o$  (where  $I_o$  is optical breakdown threshold of glass), while fundamental luminescence was registered at an intensity  $I = 10^{-4} \cdot I_o$  [17]. At radiation exposures of glasses with  $hv < E_g/2$  these effects were not found even at  $I = 0.98 \cdot I_o$ , that is it is impossible to register three- and more quantum processes.

The glass samples were irradiated with the output of a laser system allowing the production of laser pulses at 0.85  $\mu$ m wavelength and duration ~ 10<sup>-13</sup> s. A Kerr lens modelocked Ti:sapphire laser was used as the femtosecond pulse source. The output pulses of this laser were expanded in an anti-parallel grating-pair pulse stretcher and injected into a regenerative amplifier. Pulse slicers used before and after the regenerative amplifier selected a single pulse out of the pulse train. This single pulse was recompressed in a grating-pair pulse compressor. The output beam had the diameter ~ 25 mm and the energy to 1 mJ.

To measure the laser-induced damage thresholds the radiation was focused to spot sizes 2-3 microns by an objective lens with a focal length of 50 mm. The calculated distribution of irradiance in the focal plane at the depth of 1.1 mm inside of sample is shown in Fig. 1. To study the interaction processes the radiation was focused to a spot size of 230 microns on the front surface of the sample by a lens with a focal length 500 mm.

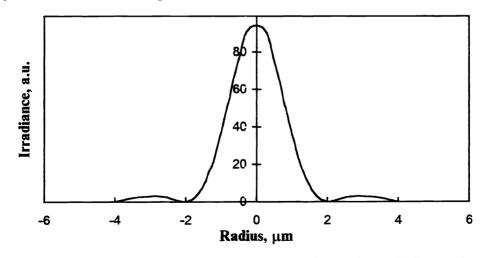


Fig.1. Calculated distribution of irradiance in focal plane of lens (F:D = 1:1) at 0.85  $\mu$ m.

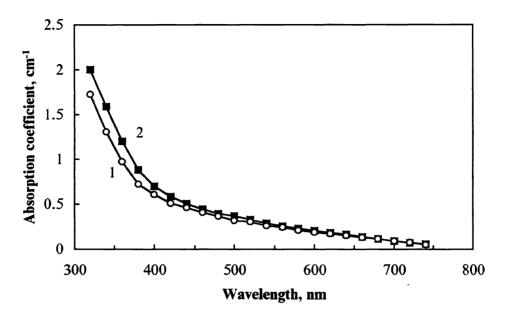


Fig.2. Spectra of additional absorption of K8 glass irradiated by laser radiation (1) and  $\gamma$ -radiation (2).

### **<u>3. RESULTS AND DISCUSSION</u>**

It was established that white light generation is brought about by laser radiation with irradiances of  $> 10^{13}$  W/cm<sup>2</sup>. This light was propagated collinear to the main radiation. Besides, a bright luminosity which is characteristic for the intrinsic luminescence of alkali silicate glasses, and the darkening along track of passing beam arose. These effects are observed only in K8 and BK7 glasses and were absent in fused silica. The measurement of additional absorption spectra of glass darkening (Fig. 2, curve 1) showed that they are in agreement with spectra of color centers absorption in these glasses (Fig. 2, curve 2). The process of color centers formation can be connected either with four-photon absorption of radiation at 0.85 µm or with linear or two-photon absorption of the supercontinuum. However, as has already been noted, three- and more quantum processes are not observed in the glasses investigated. Closer examination of this effect showed that coloration and intrinsic luminescence of the samples occurs only from a depth of 0.5 - 0.7 mm beneath the entrance surface (Fig. 3). It means that matrix ionization and coloration of the glasses are not connected with multi-photon absorption of laser radiation at 0.85 µm. It is likely that this process results from intensive light generation in a wide spectral region, followed by linear or twophoton excitation of the glass matrix under this radiation. The reason for coloration of these glasses at some depth beneath the front surface is due to the linear dependence of the spectral broadening on the medium length. Taking into account that two-photon absorption of the glasses investigated begins at intensities I ~  $10^6$  W/cm<sup>2</sup> [13-15], and that there is no well-defined intense spike at the laser frequency in the supercontinuum [1], it may be concluded that the excitation of the glass matrix resulted from two-photon absorption of the short-wavelength part of the white laser pulses.

The measurements of optical breakdown thresholds show that visible damage of K8 and BK7 glasses bulk arises at an irradiance of  $7 \cdot 10^{13}$  W/cm<sup>2</sup>, and in fused silica at an irradiance of  $2.5 \cdot 10^{14}$  W/cm<sup>2</sup>. It should be noted that luminosity is observed in the interaction volume before the

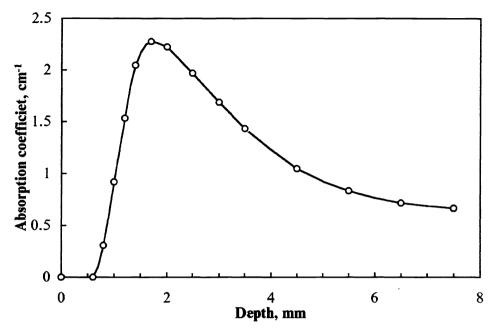


Fig.3. The dependence of additional absorption coefficient vs depth under entrance surface of K8 glass.

appearance of visible damage. The threshold of the appearance of this luminosity is less then the damage threshold by nearly 5 times. It indicates that damage occurs under conditions of intensive ionization of the glass matrix. However, in spite of the high beforethreshold concentration of electrons in interaction bulk, the damage occurs only at irradiance  $7 \cdot 10^{13}$  W/cm<sup>2</sup>. This fact indicates that laser-induced damage should not be connected with avalanche ionization.

## 4. CONCLUSION

Thus color centers formation and intrinsic luminescence of borosilicate glasses are observed under high-power femtosecond radiation at 0.85  $\mu$ m. These processes result from supercontinuum generation in the bulk glass and are followed by two-photon absorption of the short-wavelength part of this supercontinuum. These same processes lead to the damage of glasses under conditions of intensive ionization of material matrix, and show that the damage process is not connected with avalanche ionization.

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