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THERMOSTIMULATED LUMINESCENCE OF LiF: Mg, Ti IRRADIATED BY FEMTOSECOND LASER PULSES IN THE MODE OF FILAMENTATION[1]

The results of the research into the mechanism of light sum storing in LiF: Mg, Ti crystals irradiated by femtosecond laser pulses in the filamentation mode are presented. The data obtained are compared with the results for thermally stimulated luminescence (TSL) of the same crystals under beta radiation. It is found that under the action of intense femtosecond pulses of Ti:sapphire laser in the near infrared region of ~ 800 nm the peaks are formed more effectively. This is caused by higher excitation density.

Keywords: *thermostimulated, fluorescence, femtosecond, laser, filamentation.*

Methods of thermally stimulated fluorescence (TSF) are widely used to study the formation of defects, study of the nature of fluorescent centers and trapping centers, identify critical performance fluorescent and laser materials, saturable absorbers, photosensitive storage media, and other functional materials. These materials include lithium fluoride crystals, widely used in personal dosimetry, as well as the development of tunable laser media, optical shutters and optical memory materials.

The objects of our study are single crystals of dosimetric LiF: Mg (~ 100 ppm), Ti (~ 10 ppm), widely used in the γ-dosimetry. The results of studies of this type of TSF crystals irradiated with X-rays, α-particles and the vacuum ultraviolet, published in [4]. In TSF of this type crystals recorded during heating at a constant rate $\beta = 1–3\ K \cdot s^{-1}$, dominant peak with a maximum at ~ 485 K and light sum up to ~ 10 Gy is linearly proportional to the absorbed dose. With increasing of radiation dose the area of superlinear dependence of the light sum of the absorbed dose is observed, followed by saturation and even weakening of dosimetric peak [4]. TSF of lithium fluoride crystals irradiated with intense femtosecond laser pulses was not investigated before this work.

The aim of this study was to investigate the mechanism of detection of accumulation of light sum by crystals LiF: Mg, Ti with intense femtosecond pulses of Ti: sapphire laser in the near infrared region ~ 800 nm, and a study of the photofluorescence of the irradiated samples and comparing the results with the results of the TSF of the same crystals irradiated with beta radiation and other types of radiation. Mechanisms of creating color centers in LiF crystals and photofluorescence excitation by intense femtosecond laser pulses are considered in [5]. In this work it is for the first time shown that a necessary condition for the formation of color centers under the influence of the Ti: sapphire laser (~ 800 nm, ~ 1,5 eV) with low-aperture external focusing lens is filamentation accompanied by a high increase of laser light intensity that drives the highly nonlinear absorption of laser radiation.

The experimental setup used for the irradiation of these crystals by femtosecond laser pulses includes titanium-sapphire laser, generating 30 fs pulses with energy about 0,55 mJ and repetition rate of 1–1000 Hz. The exciting radiation was focused by a lens with a focal length of 30 cm. The test samples were located at a distance of 1,5 cm in front of the focus. The total energy of the laser radiation incident on the sample is determined by the number of pulses (1–120000).

Photofluorescence study of irradiated laser samples was performed using a scanning confocal fluorescent microscope PicoQuant MicroTime 200 with a time resolution. This includes studying the spatial distribution of color centers in crystals formed upon filamentation of laser radiation. Photofluorescence spectra excited by a laser beam with a wavelength of 470 nm were recorded with spectrometer Ocean Optics 6500. TSF studies were carried out with a specialized instrumentin within temperature range from 295 to 673 K with a constant heating rate $\beta = 1–3\ K \cdot s^{-1}$. After measuring the TSF curves of the crystal irradiated by femtosecond radiation TSF of the same sample irradiated by X-ray and low-energy beta radiation isotope source $^{90}Sr–^{90}Y$ a dose rate 0,6 Gy/min was additionally investigated. The exposure time of the sample beta particles was 30 s. The curves TSF were measured in 24 h after irradiation, with the aim to eliminate the influence of relaxation processes.

In Fig. 1 the photofluorescence spectra are shown with excitation wavelength 470 nm for samples irradiated by femtosecond laser radiation with a total energy exposure 2,75 mJ (*1*), 4,4 J (*2*) и 990 J (*3*).

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From the presented results it is seen that in the fluorescence spectra of irradiated crystals of lithium fluoride characteristic bands of emission F^2 centers with a maximum emission wavelength λ = 680 nm and F^{3+} centers with a maximum λ = 540 nm are observed. Studies have found that with the increase of laser irradiation to 9 J total photofluorescence intensity first increases and then begins to decline due to concentration quenching. Therefore, the curve *3* in Fig. 1 is located below curve *2*.

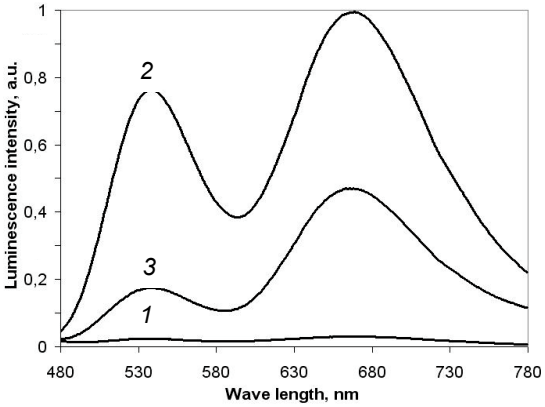
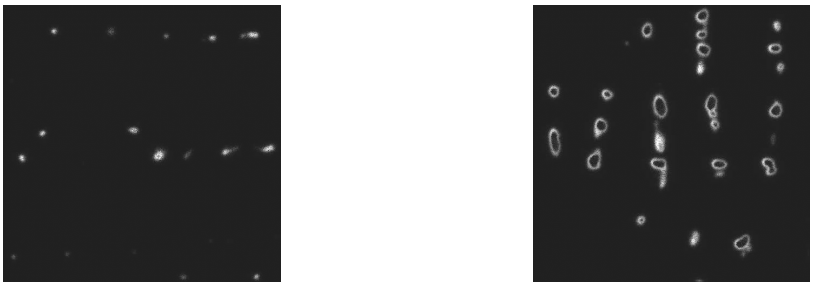


Fig. 1. Photofluorescence spectra of irradiated crystals LiF: Mg, Ti at different laser energy directed at a sample: 2,75 mJ (*1*), 4,4 J (*2*) and 990 J (*3*).

Microscopic studies have shown that the color centers in crystals irradiated by femtosecond radiation are distributed in threadlike channels (blast holes) formed in the region of the laser filaments resulting from multiple self-focusing. This can be seen in the photographs (Fig. 2), which shows the cross-fluorescence intensity distribution of color centers in the hole with two different doses of laser irradiation.

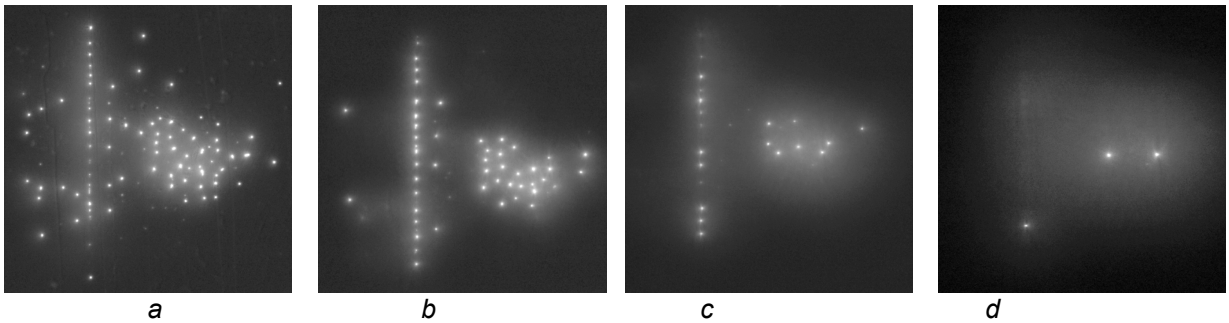


a *b*

Fig. 2. The transverse distribution of the fluorescence intensity of the color centers formed by laser irradiation (scan area 80×80 μm): *a* – energy radiation 13,75 mJ (25 counts); *b* – the energy of radiation 550 mJ (1000 pulses).

Studies have found that the number of filaments is not increased significantly with the number of pulses. Filaments during the irradiation were located in the same places. However, the cross-sectional diameter of individual holes (traces of filaments) is significantly increased (as seen at Fig. 2*b*). The concentration of color centers is decreased from the center to the periphery of the hole. Cross-sectional images of holes in rings at higher doses due to concentration quenching, increasing from the periphery to the center of the hole.

Arrangement of filaments in multiple self-focus is determined by the presence or irregularities in the original laser beam, or in the media. Surface of the samples were imperfect natural chipped crystals along cleavage planes, resulting in variations of the location of filaments in samples. This can be seen at Fig. 3, that some of the filaments located along the step formed by the split of a single crystal along a cleavage plane.



a *b* *c* *d*

Fig. 3. The transverse distribution of the intensity of the color centers at different depths from the front surface of the sample (the depth increases from *a* to *d*).

The experimental results show that the femtosecond pump main highlight is the usual dosimetric TSF peak [4] with a maximum of 485 K. In addition it is clear that a more efficient compared to the b-irradiation stocking high peaks (curve *1* at Fig. 4). This fact is not trivial and non-negotiable.

It is generally accepted that the action of femtosecond pulses on dielectric crystal is reduced to highly nonlinear ionization. In the experiment, when the energy of laser radiation several times (8–9 times) less energy interband transitions, ionization processes occur only after the self-focusing and filamentation of laser light, when its intensity increases dramatically. This can be seen at Fig. 2. In any case, the result of such exposure is the formation of electrons and holes. It was natural to expect that the curves in the femtosecond storing TSF will have the same form as in other forms of creation of electron-hole pairs, for example, the action of X-rays, electrons, or photons for band-band transitions.

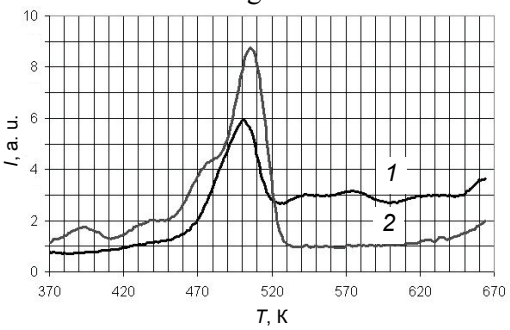


Fig. 4. TSF curves crystals LiF: Mg, Ti exposed: *1* – laser beam with an energy of 1,1 mJ; *2* – beta radiation isotope source.

If we compare the curves *1* and *2* in Fig. 4, we can see that at low doses of beta-irradiation (0,3 Gy) high peaks are induced at all. In addition, for femtosecond irradiation of the same sample used only two laser pulses with a total energy of 1,1 mJ. In these conditions, it follows from a comparison of curves *1* and *2*, and highlighted by the accumulared light sum (area under the curve) for femtosecond electron irradiation and were almost identical.

This raises the question why does the same sample at the femtosecond laser irradiation effectively produce high peaks, in contrast to the electron or X-rays, although both irradiation creates in the material in the first stage of electron-hole pairs? To answer this question, we estimate the excitation density of matter. Specifically, the experiments on the microscopic study of the topography of holes induced by laser radiation showed that after irradiation individual pulses produced about 50 holes with a size ∅1,3μ × 30μ. Excitation density is comparable with the crystal under the action of light sum storage of X-rays, on the one hand, and the laser beam filamentation mode, on the other hand. To do this, we draw attention to the fact that the light sum, flashed in the TSF for the two curves shown in the following figure, is approximately the same. However, the action of X-ray light sum is distributed throughout the sample ($2 \cdot 10^{-8}$ m^3), while under the influence of laser radiation is distributed over the volume of holes ($2{,}5 \cdot 10^{-15}$ m^3). Thus, the density of the excitation of the crystal under the action of laser radiation is about 10^7 times higher than the X-ray radiation.

In the literature, high peaks in the TSF detectors based on LiF–Mg, Ti are induced in the tracks of heavy particles, where the excitation density is much higher than the X-ray or beta radiation. We relate creation of high peaks caused by the laser radiation to a higher excitation density of material.

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