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Formation of color centers and light scattering structures by femtosecond laser pulses in sodium fluoride

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ABSTRACT

Modification of sodium fluoride crystal lattice by means of femtosecond laser pulses with $\lambda_{max}=800$ nm, energy 0.5 mJ, duration 30 fs and repetition rate 1 kHz has been considered in the paper. Effective formation of simple and complex aggregate color centers and light scattering nanodefects in the channel of a laser beam in NaF crystal have been shown for the first time. Dependences of color centers concentration on the distance between the channel center and its periphery and along the channel have been presented. Influence of external focusing on color centers creation has been revealed. Explanations of the observed phenomena have been presented on the basis of nonlinear processes taking place under the effect of high-intensity femtosecond pulses.

1. Introduction

It is known that femtosecond processing of transparent crystal materials attracts interest of researchers because it is possible to receive micro- and nano-devices in the form of optical data carriers, diffraction gratings, waveguides and others with the help of femtosecond lasers on their basis [1–3].

In our opinion, alkali fluoride crystals occupy a special place in defect formation by femtosecond radiation in transparent media, because color centers (CCs) are easily created in them, which allows to record information and microstructures in material.

In the course of transparent material processing by means of femtosecond pulses different volume and point defects are produced. If the femtosecond laser radiation intensity is greater than threshold for plasma formation (about 10^{13} W/cm^2 for transparent crystals) melting and material recrystallization, local micro-explosions, emptiness and cracks in the femtosecond beam channel can occur [4,5]. The structures in the form of waveguides with the changed refractive index in the femtosecond beam channel can be created at a lower radiation power and they can either show or not show anisotropic properties [6,7]. In addition, femtosecond radiation forms color centers, for example, in fluorides of alkaline and alkaline-earth metals [8]. The authors of this paper investigated features of creation and transformation of color centers under action γ and fs laser radiations in magnesium fluoride. They found a luminescence of the new centers at $\lambda_{max}=656$ and $780\div800$ nm in MgF_2 and revealed common features of color centers formations in fluorides of alkaline and alkaline-earth metals. Efficiency of CCs formation depends on energy and duration, repetition frequency of the pulses and conditions of radiation focusing.

Femtosecond processing of glasses, polymers and other transparent materials has been studied well enough. There are also several papers devoted to femtosecond irradiation of lithium fluoride crystals (LiF) [9–13]. However, we could not find any recent scientific publications devoted to femtosecond irradiation of sodium fluoride crystals (NaF) though this crystal has similar properties to LiF and thus has prospects for application.

We studied formation of the color centers and other defects accompanying them under the influence of high-intensity femtosecond pulses in NaF crystals with the purpose to examine the possibility for creation of micro- and nano-devices on their basis.

2. Experimental results and their discussion

2.1. Properties of defects in NaF induced by femtosecond laser radiation

We have formed 6 channels in NaF by titanium sapphire laser pulses with maximum wavelength $\lambda_{max}=800$ nm (Fig. 1). The conditions of irradiation for each channel were as follows: pulse energy 0.5 mJ, duration 30 fs, repetition rate 1 kHz, and irradiation

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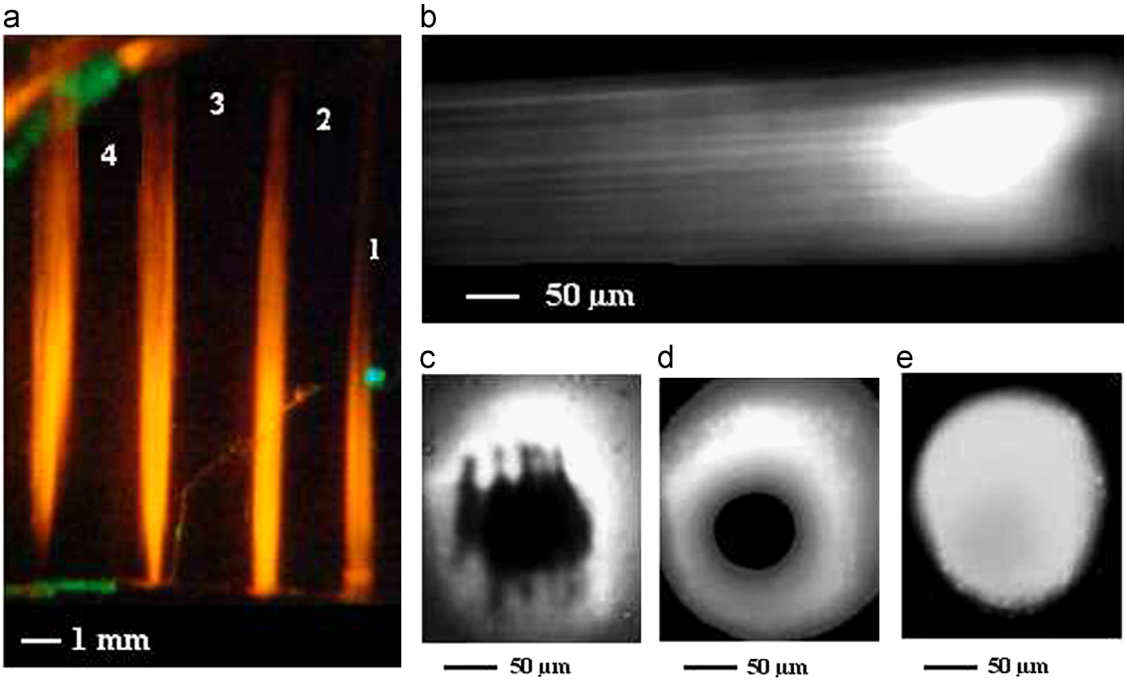

Fig. 1. The luminescence of color centers in the channels induced by femtosecond laser pulses in NaF, at excitation by the semiconductor laser with λ_{max}=445 nm: a—longitudinal images of the CCD camera of the channels along the direction of a laser beam in a crystal (the input surface at the bottom of the image, crystal height from the input surface to the image top is 1 cm. The input surface of a crystal is visible in the form of the light horizontal line); b–e—the images of the 2nd channel obtained by means of an optical microscope Olympus IX71 (b—the longitudinal image (an input surface on the right), c, d and e—cross sections of the channel at different distances from the input surface: 0, 300 and 700 μm, respectively.

time 5 s. Using an external lens with the focal length of 30 cm, we consistently increased the distance between the input surface and the geometrical focus by 0.5 cm in each subsequent channel (the first channel has a geometrical focus on the input surface). In all channels effective creation of the color centers occurred. It is indicated by the spot intensively painted with CCs in the cross section of the irradiated channels (Fig. 1c–e).

We gave our special consideration to the 2nd channel because it has all characteristic features of irradiation. The external focus of femtosecond radiation was approximately in the middle of the 2nd channel (Fig. 1a).

In Fig. 1b the longitudinal image of the 2nd channel is demonstrated. The diameter of the spot area painted by the color centers (Fig. 1c–e) is about 200 μm. The second channel has intensive formation of CCs directly from the crystal surface because intensity of the laser light falling on a crystal under existing conditions of experiment is sufficient for plasma formation, photoionization of material and creation of the color centers. The concentration of CCs in the channels (Fig. 1a) decreases with the increase of the distance between the input surface of the crystal and the geometrical focus of laser beam. Multiple filamentation in the crystal, i.e. disintegration of femtosecond beam into a set of luminescent threads (or filaments), begins with input crystal surfaces, but clearly reveals itself in the second and last third of the channel length (Fig. 1b). The luminescence is caused by color centers formed in the filaments. The mechanism of the color centers formation by femtosecond laser pulses is considered to be established. It is multiphoton-tunnel absorption of laser pulses that transfers the electron of valence band to conduction band of the crystal, creating electron–hole pair [14,15]. The energy of one photon of the titanium sapphire laser is equal to 1.55 eV. One electron of valence band must receive 8 photons simultaneously to be able to overcome the width of the forbidden band in sodium fluoride (12.2 eV according to [16]). The free electrons or excitons (formed as the free electrons decay) are formed as a result of this process. The strong electron-lattice coupling in alkali halides causes the self-trapping of excitons, producing self-trapped excitons (STEs). Decomposition of self-trapped excitons into primary radiation defects (F–H pairs of centers) occurs subsequently. A Frenkel pair (F–H pair) consists of an F center (an electron in the field of an anion vacancy) and an H center, which is a dihalide F_2^- molecule located at one anion site. Then the aggregate color centers are formed from primary radiation defects. The color of the crystal in the field of irradiated channels is the evidence of CCs creation. Besides, we observed absorption and luminescence of CCs by means of a confocal microscope.

It should be noted that the consecutive movement of external focusing of a femtosecond beam from an input surface of a crystal to the output surface of a crystal and further behind the output surface of a crystal provides the increase of a self-focusing distance in a crystal according to the size of the geometrical focus shift (Fig. 1a). Comparison of channels 2, 3 and 4 allows to note that in the 4th channel the self-focusing of laser radiation occurs at the distance of 1–2 mm from the input surface of a crystal. External focusing of femtosecond radiation was in this case at the distance of 5 mm behind an output surface of a crystal. The formation of CCs in the 4th channel also began at the distance of 1–2 mm from an input surface of a crystal.

2.2. Luminescence of the color centers in NaF channels, induced by femtosecond laser pulses

Fig. 1c shows luminescence of color centers on an input surface of NaF crystal in the cross section of the 2nd filament. The diameter of the painted area is about 200 μm. One can see that color centers have been very intensively formed directly on a crystal surface. The black area in the center of the picture means that large aggregate color centers, such as R, N and more complex ones, consisting of more than four F centers, were formed in this area. The R-center is neutral aggregate consisting of three adjacent