Laser performance investigation of a new UV active media LiY$_{0.3}$Lu$_{0.7}$F$_4$:Ce$^{3+}$ and LiY$_{0.3}$Lu$_{0.7}$F$_4$:Ce$^{3+}$+Yb$^{3+}$

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Abstract. We show that the LiY$_{0.3}$Lu$_{0.7}$F$_4$:Ce$^{3+}$ and LiY$_{0.3}$Lu$_{0.7}$F$_4$:Ce$^{3+}$+Yb$^{3+}$ crystals are promising active media of UV spectral range with low-threshold lasing (30-90 mJ/cm$^2$). Due to crystal-chemical approach (additional doping by ions Yb$^{3+}$) we obtained the effect of suppression of spurious photodynamic processes. The active medium LiY$_{0.3}$Lu$_{0.7}$F$_4$: Ce$^{3+}$+ Yb$^{3+}$ combines properties of saturable amplifier and oscillator with small saturation energy, and promising to generate pulses of ultrashort duration in the UV spectrum.

1. Introduction

In comparison with a set of the solid-state lasers of infrared and visible ranges, the number of the solid-state lasers radiating in ultraviolet (UV) and vacuum ultra-violet (VUV) ranges is limited. There are problems [1] on a way of creation of these lasers, such as material solarization, excited state absorption (ESA) [2]. However, compact solid-state lasers of UV and VUV spectrum ranges development will allow solving variety of problems in various areas of science and technology as medicine, biology, microelectronics, nanotechnology, laser spectroscopy, materials precision processing, etc.

One of the approaches to solve the material solarization problem is in increasing crystal photochemical stability of solid-state materials with respect to UV radiation.

Objects of the investigation LiY$_{0.3}$Lu$_{0.7}$F$_4$:Ce$^{3+}$ and LiY$_{0.3}$Lu$_{0.7}$F$_4$:Ce$^{3+}$+Yb$^{3+}$ crystals can be attributed to the materials suitable for the construction of lasers operating in the UV region of the spectrum.

The purpose of this study is to investigate the lasing characteristics of active media LiY$_{0.3}$Lu$_{0.7}$F$_4$:Ce$^{3+}$ and LiY$_{0.3}$Lu$_{0.7}$F$_4$:Ce$^{3+}$+Yb$^{3+}$ and intracavity losses connected to these active media.

2. Objects of study and experimental setup

Double fluoride mixed crystals of scheelite structure, doped by ions Ce$^{3+}$, which are the objects of our investigation are homologous to known Ce:LiYF$_4$ and CeLiLuF$_4$ active media. Previous studies [3] have shown that the ratio of yttrium ions (30%), and lutetium (70%) provides the maximum ratio of cerium ions entering the matrix. Ce$^{3+}$ content was 1 at. % in both samples and Yb$^{3+}$ ions content was 1 at. %.
These crystals were synthesized in the crystal growth laboratory, the Department of Quantum Electronics and radiospectroscopy of Institute of Physics at the Kazan Federal University. These crystals are the new active media, generating in the ultraviolet range of the spectrum.

The samples were prepared for side pumping scheme. They were a half-cylinders of length 7.1 mm with polished parallel ends, which were side-polished plane angle, the value of which is chosen from Brewster fall of reflected from the mirrors of the lasing beam with a wavelength of 309 nm. Such a choice of the angles between the faces and the side planes is due to the need to obtain π-polarized light laser generation with minimal losses.

The experimental setup for measuring the lasing characteristics is shown in Figure 1. It comprises side pumping by 1st Stokes component of 266 nm laser radiation obtained from Raman shifter cell with H$_2$ and nonselective cavity for investigated crystals.

3. Results. Lasing characteristics

Experimental plots of the output energy depending on the pump energy for these crystals for output mirrors with different reflection coefficients R are shown in Figures 2a and 2b.

As it is evident from the nature of the energy dependence of lasing on the pump energy (Fig.2a), there is a hysteresis in the generation characteristics: the course of the value of energy of laser oscillation with increasing pump energy is not the same as the corresponding dependence with decreasing pump energy. When reducing the pumping energy, the energy of the lasing is higher than the value when we increase pumping energy. Hysteresis confirms that dynamic processes induced in the active media which cause additional intracavity losses. Additionally, these losses are changing their value during the experiment.

For the active medium without ytterbium doping the maximum slope efficiency was η = 14% (R=70%). For the corresponding output mirror in the cavity there is a large part of the laser radiation, as compared to other mirrors. The fact that color centers are destroyed due to the absorption of laser radiation provides relatively high slope efficiency. This also explains the hysteresis of the lasing characteristics of Fig. 2a. An increasing number of color centers are being destroyed by the end of the experiment.

Also the graph (Figure 2a) shows that the area of the hysteresis loop is minimal when the output mirror with a reflection coefficient R=70% is used, which confirms the fact that the formed color centers are destroyed more efficiently.

For a sample additionally doped by ytterbium ions LiY$_{0,3}$Lu$_{0,7}$F$_4$:Ce$^{3+}$+Yb$^{3+}$ the hysteresis dependence was not observed. This suggests that photodynamic processes were suppressed by the addition of an
effective electron traps (Yb$^{3+}$). However, the lasing saturation was observed. The maximum slope efficiency was found to be $\Delta \eta = 11\%$ with the output mirror reflecting $R = 55\%$ of the laser generation.

It is known that the dynamic saturation of the amplifying medium can be used for mode locking in schemes with slow saturable absorber. Thus, the active medium with ytterbium ions is promising for ultrashort pulses lasing.

![Fig 2a. Output lasing energy depending on the pump energy for LiY$_{0,3}$Lu$_{0,7}$F$_4$:Ce$^{3+}$ crystal for output mirrors with different reflection coefficients R](image1)

![Fig 2b. Output lasing energy depending on the pump energy for LiY$_{0,3}$Lu$_{0,7}$F$_4$:Ce$^{3+}$+Yb$^{3+}$ crystal for output mirrors with different reflection coefficients R](image2)

Table 1. Slope efficiency for mirrors with different reflection coefficients

| R, reflection of mirror, % | LiY$_{0,3}$Lu$_{0,7}$F$_4$:Ce$^{3+}$ | LiY$_{0,3}$Lu$_{0,7}$F$_4$:Ce$^{3+}$+Yb$^{3+}$ |
|---------------------------|---------------------------------|---------------------------------|
| 30                        | 5,7 ± 0,1                       | 10,7 ± 0,8                      |
| 48                        | 9,5 ± 0,3                       | 8,2 ± 0,4                       |
| 55                        | 5,8 ± 0,3                       | 11,5 ± 0,4                      |
| 64                        | -                               | 8,0 ± 0,2                       |
| 70                        | 14,2 ± 0,4                      | -                               |

4. Intracavity losses calculation

Producing a further analysis of the data, we have calculated the intracavity losses. From [4] the energy balance equation is known:

$$E_g = \eta \cdot \Delta \cdot \frac{K_c}{K_r + \rho} \cdot (E_p - E_{th}), \quad (1)$$

Where $E_g$ – lasing energy, $\eta$ – quantum efficiency of luminescence, $\Delta = h\nu / h\nu_p$ – quantum efficiency of laser generation, $K_c$ – “useful losses” coefficient corresponding to couplers reflectance, $E_p$ – pumping energy, $E_{th}$ – threshold pump energy and constant $\rho$ – coefficient of the intracavity losses. However, in our case, the coefficient $\rho$ should be a function $\rho = \rho_0 + \rho_\phi \left(E_p, E_g, R, t\right)$ [5], where
$\rho_b$ – passive intracavity losses, \( R \) – output mirror reflection coefficient, \( t \) – time. Functional dependence $\rho_b=f(E_p, E_{g\text{cav}}, R, t)$ takes into account both the processes of formation of color centers under the pump, and their destruction, spontaneous (due to thermal vibrations of the lattice) and due to ionization of color center by laser radiation circulating inside the cavity (this process is called photobleaching).

After some simple mathematical transformations we can receive the final formula for the calculation of intracavity losses [5]:

$$\rho = K_r \cdot \left( \frac{\eta \cdot \Delta \cdot E_p - \eta \cdot \Delta \cdot K_r \cdot a - E_g}{\eta \cdot \Delta \cdot K_r \cdot a + E_g} \right), \quad (2)$$

$$a = \frac{h\nu_p \cdot V}{\sigma_g \cdot \eta}, \quad (3)$$

Where \( h\nu_p \) – pump photon energy, \( V \) – pumping volume, \( \sigma_g \) – laser transition cross section.

Figure 3 shows intracavity losses depending on the pump energy $\rho(E_p)$ for output mirrors with different reflection coefficients \( R \) for the sample without Yb (Fig 3a) and with ytterbium (Fig 3b).

From the curves shown in Figures 3a and 3b we can see that by increasing of pumping energy total intracavity losses are reduced. This dependence is explained by the formation of color centers by pumping radiation and their subsequent destruction by laser radiation. The more lasing remains in the cavity, the lower amount of the color centers.

Curves for the sample without ytterbium shows that increasing the pump energy curve (Figure 3a) is not the same as pump reduction curve, which also illustrates the existence of color centers creation determined dynamic processes in the sample. If sample was additionally activated by ions of Yb$^{3+}$ dependence is not so pronounced, and the intracavity losses are independent of the conditions of the experiment, therefore, we were able to suppress spurious photodynamic processes through preparing crystal compound, a crystal-chemical approach.

As mentioned earlier, the crystal with ytterbium can be used to generate ultrashort pulses. The obtained relation shows that for small values of the pump energy, and, consequently, the value of lasing losing more, and as a result, the leading edge of the pulse laser generation will suffer heavy losses. The trailing edge will be shortened due to saturation of the gain medium LiY$_{0.3}$Lu$_{0.7}$F$_4$:Ce$^{3+}$+Yb$^{3+}$. 

![Figure 3a. Intracavity losses $\rho(E_p)$ dependence on pumping energy for LiY$_{0.3}$Lu$_{0.7}$F$_4$:Ce$^{3+}$ active medium with different reflection coefficients R of output mirror](image-url)

![Figure 3b. Intracavity losses $\rho(E_p)$ dependence on pumping energy for LiY$_{0.3}$Lu$_{0.7}$F$_4$:Ce$^{3+}$+Yb$^{3+}$ active medium with different reflection coefficients R of output mirror](image-url)
5. Conclusion
As a result, we show that the LiY\(_{0.3}\)Lu\(_{0.7}\)F\(_4\):Ce\(^{3+}\) and LiY\(_{0.3}\)Lu\(_{0.7}\)F\(_4\):Ce\(^{3+}\)+Yb\(^{3+}\) crystals are promising active media of UV spectral range with low-threshold lasing (30-90 mJ/cm\(^2\)). Due to crystal-chemical approach (additional doping by ions Yb\(^{3+}\)) we obtained the effect of suppression of spurious photodynamic processes. Intracavity losses were reduced, and as a result, significantly reduced the lasing threshold. The active medium LiY\(_{0.3}\)Lu\(_{0.7}\)F\(_4\):Ce\(^{3+}\)+Yb\(^{3+}\) combines properties of saturable amplifier and oscillator with small saturation energy, and promising to generate pulses of ultrashort duration in the UV spectrum.

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