

# Intracavity losses investigation of $\text{LiCaAlF}_6:\text{Ce}^{3+}$ laser

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## ABSTRACT

We have discovered that total intracavity losses, being dependent on color centers amount, go down for higher pump energies for  $\text{LiCaAlF}_6:\text{Ce}^{3+}$ . This dependence is explained by the formation by the pump radiation and destruction of color centers due to laser radiation. The more energy remains in the lasing cavity, the lower the color centers absorption. Such dependencies were investigated for active medium crystals grown by different methods. Influence of growth conditions on active media characteristics is discussed.

As a result we have shown for the first time that the crystal  $\text{LiCaAlF}_6:\text{Ce}^{3+}$  internal losses depend on the pump energy. Method has been worked out to determine the intracavity losses of the laser, which allows evaluation of prospects of its practical use in the most correct way.

Solid-state UV active media, intracavity losses, UV lasers.

## 1. INTRODUCTION

For the development of techniques and technologies, to date, the use of lasers of ultraviolet (UV) spectral range is required. The use of short wavelength coherent radiation defines a new level of quality and efficiency in the field of precision materials processing and in many other areas. However, application in industry is currently limited due to lack of market reliable and inexpensive in use lasers of UV range. The most promising active medium for such devices is analyzed in this work  $\text{LiCaAlF}_6:\text{Ce}^{3+}$  crystal, as the active medium based on it allows obtaining stable practically important characteristics of the laser<sup>1</sup>.

Commercial applications of lasers include increasing production and quality control of active media, calculation and construction of an optimal cavity. It is particularly important for active media of UV spectral range, as they are characterized by photodynamic processes caused by defects in the crystal matrix and the energy scheme of states of active ions. These processes are induced by the pump radiation and lasing<sup>2</sup>, resulting in cavity Q factor dependency on the energy of the pump radiation<sup>3</sup>.

In the development of new quantum electronics devices for the UV range one should take into account the dynamic processes associated with ionization of impurity centers and the formation of color centers.

## 2. THE SCHEME OF SOLID-STATE TUNABLE UV LASER

The oscillator with double-pass amplifier, seen at work, is shown in Figure 1.

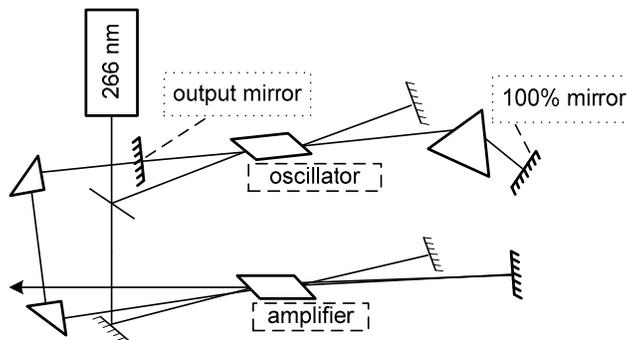


Figure 1. Scheme of the laser oscillator with an amplifier.

The pump radiation is divided by a semireflecting mirror into two beams to pump the active medium of the oscillator and amplifier. The scheme provides the return of pump radiation to the crystal for more efficient use of the pump. The oscillator is formed by two flat mirrors. Since one of the major benefits of  $\text{LiCaAlF}_6:\text{Ce}^{3+}$  active medium is the possibility of obtaining tunability in the range of 281-305 nm inside the oscillator a dispersive element is set - a prism for a smooth adjustment of the wavelength of lasing. The objectives of the proposed unit are to provide a channel for probe of the pump-probe experiments, as well as providing coherent radiation excitation laser spectroscopy with a spectral resolution of 1 nm.

### 3. THE ACTIVE MEDIUM WITH THE OPTIMAL LASING CHARACTERISTICS

To construct the optimal cavity one must use as an active element the crystal of best quality, i.e. has the lowest ratio of losses. Earlier the laser experiment interpretation technique was proposed which allows evaluating correctly the total intracavity losses with photoinduced processes involved<sup>7</sup>.

We have treated several sets of data on laser characteristics, previously obtained in different laboratories. The first treated the experimental data were obtained by authors for  $\text{LiCaAlF}_6:\text{Ce}^{3+}$  crystal, grown in the laboratory of crystal growth of Physics Department, Kazan (Volga Region) Federal University. The sample was prepared for the longitudinal pumping schemes, a cylinder length of 2 mm with polished parallel ends, which were to form an angle whose value is chosen from the conditions of Brewster incidence of laser radiation on the surface. The crystal was grown by the Bridgman-Stockbarger method. The cavity was formed by two flat mirrors spaced 10 cm apart. Output couplers were varied with different reflection coefficients  $R = 66, 58, 50, 45\%$ . The pump is the radiation of 4-th harmonic with a wavelength of 266 nm laser YAG: Nd, pump pulse duration was 8 ns, pulse repetition rate of 10 Hz. The pump radiation was focused into a beam with radius of 1 mm.

Figure 2 shows the dependence of the radiation energy from the lasing energy of the pump radiation for cavities with different Q value.

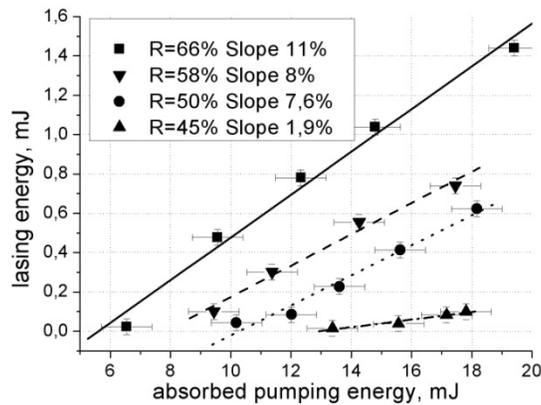


Figure 2. The dependence of the lasing energy on the pump energy for cavities with different reflectances of output coupler for crystal  $\text{LiCaAlF}_6:\text{Ce}^{3+}$ .

Dependence of dynamic losses calculated from experimental data on laser action, on the energy of the pump radiation is shown in Figure 3.

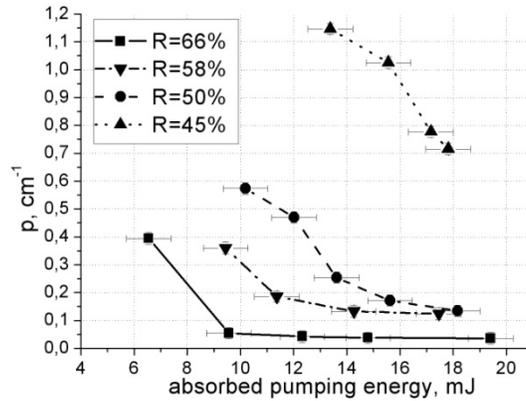


Figure 3. Dependence of dynamic losses on the pump energy for cavities with different reflectivity output coupler for crystal  $\text{LiCaAlF}_6:\text{Ce}^{3+}$ .

Dependences presented in Figure 3 show that with increasing pump energy total intracavity losses decrease. This is explained by the formation by the pump radiation color centers and their subsequent destruction by laser radiation. The more laser radiation energy remains in the cavity, the lower the color centers absorption.

The next set of lasing experimental data on laser was taken from <sup>8</sup> where the crystal  $\text{LiCaAlF}_6:\text{Ce}^{3+}$  sized 10x10x10 mm was studied, which was grown by the Czochralski method. Pumping was carried out at a wavelength of 263.3 nm, pulse duration was 140 ns, pulse repetition rate of pumping 100 and 500 Hz. The pump radiation was focused into a beam area  $S = 0.3 \cdot 10^{-3} \text{cm}^2$ . Lasing was observed at a wavelength of 289 nm. The cavity was formed by two flat mirrors, the cavity length was 6.7 cm, the reflection coefficient of the output mirror of 50% <sup>8</sup>. Dependence of the laser radiation energy on the pump energy is shown in Figure 4.

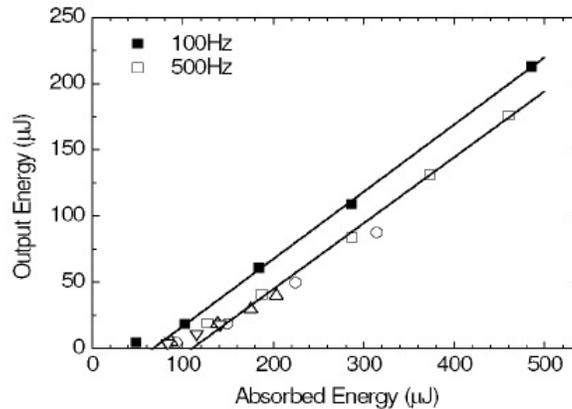


Figure 4. Dependence of the laser radiation energy of the pump energy for the crystal  $\text{LiCaAlF}_6:\text{Ce}^{3+}$ ; repetition rate pumping: ■ - 100 Hz, □ - 500 Hz <sup>8</sup>.

The related dependence of the total intracavity losses of energy for the lasing pulse repetition rate of pumping 100 and 500 Hz is shown in Figure 5.

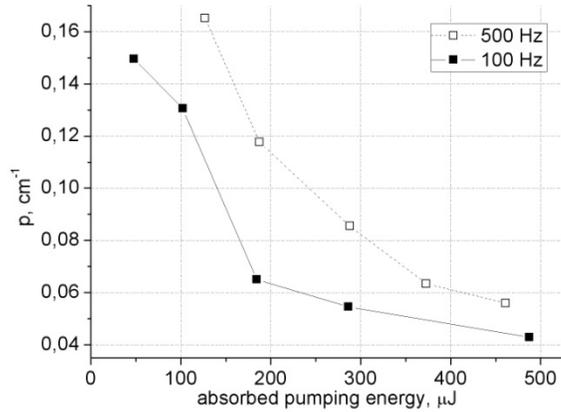


Figure 5. Dependence of the total intracavity losses of the pump energy for the crystal  $\text{LiCaAlF}_6:\text{Ce}^{3+}$ ; pulse repetition rate of pumping: ■ - 100 Hz, □ - 500 Hz.

The figure 5 shows that losses decrease with an increase of pump energy, indicating bleaching of color centers by laser radiation. Also it is shown that with increasing pulse repetition rate losses increase, indicating the accumulation of color centers during the experiment. Influence of the lifetime of color centers on lasing active medium was noted even in <sup>6</sup>. We found that the losses associated with the absorption of color centers depends on the period of repetition.

There have also been studied data on the lasing experiments <sup>9</sup> where two crystals  $\text{LiCaAlF}_6:\text{Ce}^{3+}$  and  $\text{LiSrAlF}_6:\text{Ce}^{3+}$  were investigated. These crystals were grown by the Czochralski method. Pumping was carried out at a wavelength of 266 nm, pulse duration 10 ns, repetition rate of 10 Hz. The pump radiation was focused into a beam radius of 68 microns. Lasing was observed at a wavelength of 292 nm. The resonator was formed by two flat mirrors, the distance between the mirrors of 11 cm. Reflectivity of output coupler was 50% <sup>9</sup>. Dependences of the laser energy on the pump energy are shown in Figure 6a and b for crystals  $\text{LiSrAlF}_6:\text{Ce}^{3+}$  and  $\text{LiCaAlF}_6:\text{Ce}^{3+}$  respectively <sup>9</sup>.

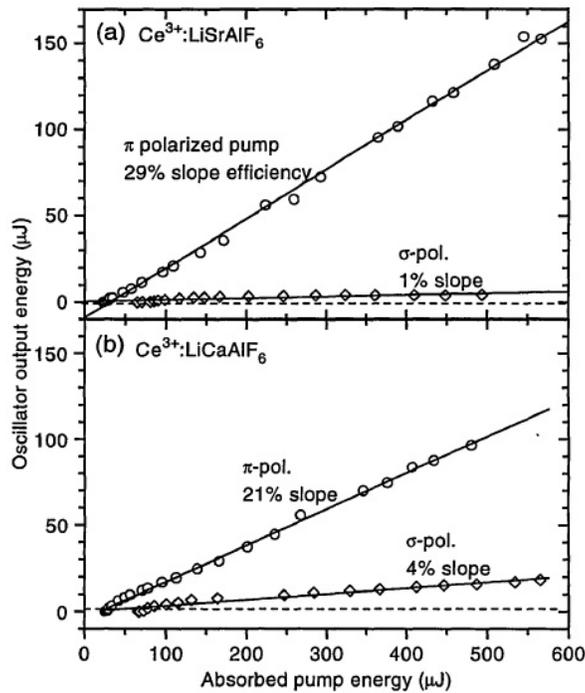


Figure 6. The dependence of laser radiation energy on the pump energy for the crystal  $\text{LiSrAlF}_6:\text{Ce}^{3+}$  (a) and  $\text{LiCaAlF}_6:\text{Ce}^{3+}$  (b) for different polarizations: ○ -  $\pi$  polarization, ◇ -  $\sigma$  polarization <sup>9</sup>.

Figure 7 shows the recalculated total intracavity losses of the lasing energy for these crystals.

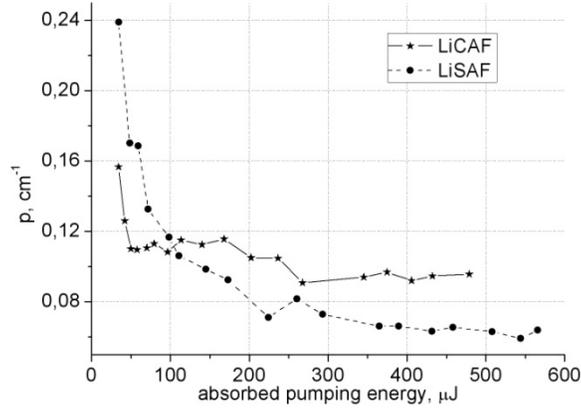


Figure 7. The dependence of the total intracavity losses of the pump energy for the crystals  $\text{LiSrAlF}_6:\text{Ce}^{3+}$  and  $\text{LiCaAlF}_6:\text{Ce}^{3+}$ .

From figures 6 a and b also can be concluded that color centers had been forming in the crystals during the experiment, which subsequently fade with increase of laser radiation energy. The value of the losses factor at the threshold of lasing is higher for crystal  $\text{LiSrAlF}_6:\text{Ce}^{3+}$ . Indeed, it is known that the active elements on the basis of this crystal are much more solarizable by the radiation of the pump. In this case, as shown by the dependence in figure 7, with increasing pump energy, color centers are bleaching by the lasing emission, but the loss is obviously due to color centers are reduced in a wide range of energy values of the pump radiation. For a crystal  $\text{LiCaAlF}_6:\text{Ce}^{3+}$  losses no longer depend on the energy of the pump radiation, even at low pumping. However, the losses rate remains high, compared with the crystal  $\text{LiSrAlF}_6:\text{Ce}^{3+}$ , which is explained by the better optical quality of the latter. It is known that growing crystal  $\text{LiCaAlF}_6:\text{Ce}^{3+}$  requirements for the growth are much higher.

From the data presented it can be concluded that the dynamic losses associated with the formation of color centers under the action of pump radiation and their destruction by laser radiation are common to all crystals  $\text{LiCaAlF}_6:\text{Ce}^{3+}$  and  $\text{LiSrAlF}_6:\text{Ce}^{3+}$ , regardless of their growth conditions. However, the data presented, we can conclude that the Czochralski method for growing crystals provides smaller losses on the color centers, i.e. the number of defects, than in crystals grown by the Bridgman-Stockbarger. This is evidenced by lower losses at high pump energies.

#### 4. OPTIMIZATION OF LASER PUMPING

For best pumping of an active medium, i.e. the creation of its population inversion, it is necessary to find the optimal parameters, such as the length of the crystal and the intensity of the pump radiation.

For the four-level scheme (figure 8) we can write the system of differential equations and considering the stationary case we get:

$$dN_1/dt = -(I\sigma/h\nu)N_4 + N_2/t_{21} = 0, \quad (4.1.1)$$

$$dN_2/dt = N_3/t_{32} - N_2/t_{21} = 0, \quad (4.1.2)$$

$$dN_3/dt = N_4/t_{43} - N_3/t_{32} = 0, \quad (4.1.3)$$

$$dN_4/dt = (I\sigma/h\nu)(N_1 - N_4) - N_4/t_{43} = 0, \quad (4.1.4)$$

$$N_1 + N_2 + N_3 + N_4 = N_t. \quad (4.1.5)$$

Solving this system of equations for  $\Delta N/N_t$ , we obtain:

$$\Delta N/N_t = 1/(1 + (I\sigma/h\nu)(t_{32} + t_{21} + 2t_{43})), \quad (4.2)$$

$$I_s = h\nu/\sigma(t_{32} + t_{21} + 2t_{43}). \quad (4.3)$$

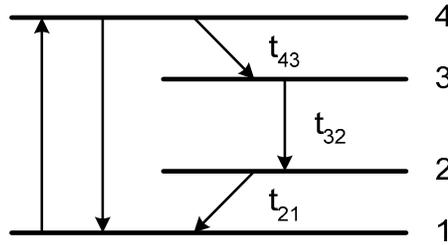


Figure 8. Four - level circuitry.

By Bouguer law in a medium with an absorption coefficient  $\alpha$  for the light intensity depending on the propagation depth, we have:

$$I(x)=I_0 \exp(-\alpha x), \quad (4.4)$$

where  $I_0$  - the initial intensity of electromagnetic waves,  $x$  - coordinate axis along the direction of propagation. When applying the amendments to the saturation, we have:

$$I(x)=I_0 \exp\left(-\frac{\alpha_0}{1+I(x)/I_s} x\right), \quad (4.5)$$

where

$$I_s = h\nu/2\sigma\tau, \quad (4.6)$$

is a parameter that depends on the properties of the medium and the frequency of the incident radiation. As discussed above the pump radiation after a single passage through the crystal returns back to the crystal by a mirror. We write the intensity distribution of the crystal for this case:

$$I(x)=I_0 \exp\left(-\frac{\alpha_0}{1+I(x)/I_s} x\right) + I(L) \exp\left(-\frac{\alpha_0}{1+I(x)/I_s} (L-x)\right), \quad (4.7)$$

where  $L$  - length of the crystal.

The intensity distribution of pump radiation was simulated on a computer. Parameters  $L$  and  $I_0$  have been optimized for the case of the closest to uniform distribution of pump intensity on the crystal, the maximum of its value was close to the threshold of destruction of the crystal. The calculation was performed for the activator ions content  $c = 1,5 * 10^{18} \text{ cm}^{-3}$ , which corresponds to the initial absorption coefficient  $\alpha_0 = 5,01 \text{ cm}^{-1}$ . The simulation results are presented in Figure 9 and 10.

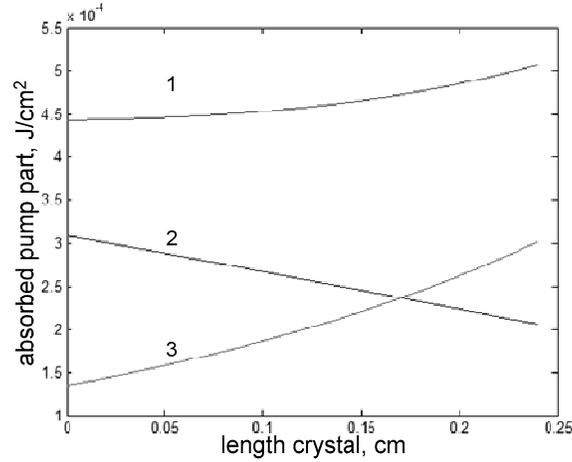


Figure 9. The distribution of the absorbed part of pump energy along the length of the crystal. 2 - density of the absorbed part of pump energy for direct passage along the crystal, 3 - to reverse the passage, 1 - the total density of the absorbed part of pump energy.

Thus the optimum parameters were found: the length of the crystal  $L = 0,24$  cm,  $I_0 = 0,72$  J/cm<sup>2</sup>. Calculated for the initial absorption coefficient  $\alpha_0 = 5,01$  cm<sup>-1</sup> and the saturation intensity  $I_s = 0.54$  J/cm<sup>2</sup>.

As a result of absorption of pump energy in the medium, a population inversion  $\Delta N = N_3 - N_2$ :

$$\Delta N = I_{\text{abs}} / h\nu, \quad (4.8)$$

where  $l$  - length of the crystal,  $h\nu$  - photon energy of the pump,  $I_{\text{abs}}$  - absorbed part of the pump. This inversion corresponds to an initial gain:

$$k_0 = \Delta N \sigma, \quad (4.9)$$

where  $\sigma$  - cross-section of the stimulated transition  $3 \rightarrow 2$ . Thus we get:

$$k_0 = \frac{I_{\text{abs}} \sigma}{h\nu}. \quad (4.10)$$

To calculate the parameters of the initial gain was  $k_0 = 4,2$  cm<sup>-1</sup>.

## 5. OPTIMAL FEEDBACK

In the case of losses, depending on the laser energy inside the cavity, the use of known formula for determining the optimal losses on laser output  $K_r$  (5.1) leads to incorrect results.

$$K_r^{\text{opt}} = \sqrt{k_0 \rho} - \rho, \quad (5.1)$$

$$K_r = \frac{1}{2L} \ln \left( \frac{1}{R} \right), \quad (5.2)$$

here  $\rho$  - intracavity losses,  $k_0$  - initial gain,  $l$  - length of the cavity,  $R$  - reflection coefficient of the output coupler of the cavity.

To determine the optimal parameters of the cavity a mathematical model was constructed of four-level scheme (Figure 12) of the laser oscillation with losses, depending on the laser radiation energy. To simplify the calculation was made for

the case of one longitudinal mode in the cavity considered at a wavelength of 293 nm, and the spatial distribution of radiation intensities of the pump and lasing radiation in the cavity and active medium is neglected.

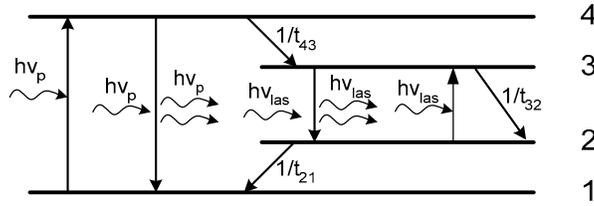


Figure 12. Four-level scheme of lasing.

The system (5.3) represents the system of differential equations corresponding to this model with embedded losses (5.4) depending on the pump energy.

$$dN_1/dt=(N_4-N_1)\sigma_{14}U+N_2/t_{21}, \quad (5.3.1)$$

$$dN_2/dt=(N_3-N_2)\sigma_{32}q-N_2/t_{21}+N_3/t_{32}, \quad (5.3.2)$$

$$dN_3/dt=(N_2-N_3)\sigma_{32}q-N_3/t_{32}+N_4/t_{43}, \quad (5.3.3)$$

$$dN_4/dt=(N_1-N_4)\sigma_{14}U-N_4/t_{43}, \quad (5.3.4)$$

$$dq/dt=c(N_3-N_2)\sigma_{32}q-q/t_q, \quad (5.3.5)$$

$$N_1+N_2+N_3+N_4=N. \quad (5.3.6)$$

$$t_q=1/(K_r+\rho)c, \quad (5.4)$$

$$\rho=A/(1+Bq). \quad (5.5)$$

Where  $N_1 \dots N_4$  - the population of the energy levels,  $N$  - content of the activator ions,  $q$  - the number of lasing photons in the cavity,  $\sigma_{14}$ ,  $\sigma_{32}$  – cross-sections of stimulated transitions  $1 \rightarrow 4$  and  $3 \rightarrow 2$ , respectively.  $t_{21}$ ,  $t_{32}$ ,  $t_{43}$  - lifetimes of the energy levels of 2, 3 and 4, respectively,  $1/t_{ij}$  - corresponding to the probability of transition  $i \rightarrow j$ . To explain the formulas (5.4) in Figure 13 are shown schematically the bottom of the conduction band (CB) and the ground state of color centers (CC).

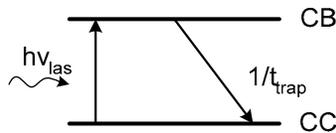


Figure 13. Scheme for the energy of conduction band and the color centers.

In the following the differential equation and the normalization condition for this scheme:

$$dN_{CC}/dt=N_{CB}/t_{trap}-N_{CC}\sigma_{CC}q, \quad (5.6)$$

$$N_{CC}+N_{CB}=N_C, \quad (5.7)$$

where  $N_{CC}$  and  $N_{CB}$ , respectively, the population of color-center and the conduction band,  $N_S$  their sum, which in general depends on the pump energy,  $1/t_{\text{trap}}$  - the probability of electron capture by traps from the conduction band,  $\sigma_{cc}$  - absorption cross-section of energy lasing color centers. This model does not take into account the electron transition from the level of color centers back into the conduction band, since the probability of this process is much less than the probability of the considered processes. Solving the equation (5.5) in the stationary approximation we obtain:

$$N_{CC} = \frac{N_S}{t_{\text{trap}} \sigma_{CC} q + 1}. \quad (5.8)$$

Solving this equation for  $\rho \sim N_{CC}$ , we obtain:

$$\rho = A / (1 + Bq), \quad (5.9)$$

where the parameters  $A$  and  $B$  vary in the course of the procedure of approximation of experimental data.

As a result of constructing the model on the computer and data approximation of lasing parameters of photodynamic processes were determined. Then by extrapolating the results to higher pumping and output coupler losses the point of the maximum laser energy and therefore optimum for output coupler losses was estimated  $K_r^{\text{opt}} = (1/21) \ln(1/R) = 0.3 \text{ cm}^{-1}$ .

## 6. CONCLUSION

The method of calculating the optimal parameters of laser cavity for the  $\text{LiCaAlF}_6:\text{Ce}^{3+}$  UV active medium was proposed, which provides accounting losses depend on the intensity of the pumping and lasing.

With the proposed method the optimum parameters were calculated for the tunable wavelength oscillator on the basis of the crystal  $\text{LiCaAlF}_6:\text{Ce}^{3+}$  with two-pass amplifier, including the length of active medium, the intensity of pump radiation, the output coupler losses.

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