

As manuscript



Nurtdinova Larisa Alvertovna

**LASING IN $\text{LiY}_x\text{Lu}_{1-x}\text{F}_4\text{:RE}^{3+}$ (RE=Ce, Yb) CRYSTALS WITH
MANAGEABLE PHOTODYNAMIC PROCESSES**

Specialty 01.04.05 – optics

AUTHOR'S ABSTRACT

of Thesis for the degree of the candidate of physical and mathematical sciences (PhD)

KAZAN – 2015

Research was conducted at the Department of Quantum Electronics and
Radiospectroscopy of Kazan Federal University

Scientific adviser:

Tagirov Murat Salihovich, Doctor of Physical and Mathematical Sciences, Professor, Head of the Department of Quantum Electronics and Radiospectroscopy of the Institute of Physics of Kazan Federal University

Official opponents:

Naumov Andrei Vitalievich, Doctor of Physical and Mathematical Sciences, Associate Professor of "Optics", Institute for Spectroscopy RAS (ISAN), Troitsk, Head of the Department of Molecular Spectroscopy, Deputy Director for Research.

Lobkov Vladimir Sergeevich, PhD, Kazan E. K. Zavoisky Physical-Technical Institute (KPhTI) of the Kazan Scientific Center of the Russian Academy of Sciences (KFTI KSC RAS), Kazan, Head of the Laboratory of Ultrafast Molecular Processes.

Reviewing organization:

N.P.Ogarev Mordovia State University, Saransk

The defence will take place in October 15th 2015 at 15.20 during the meeting of the Dissertation Committee Д212.081.07 at the Institute of Physics (aud. 212), 16a Kremlevskaya st., Kazan, Russia, 420008.

Thesis is available for examination at the Nikolay Lobachevsky Scientific Library of Kazan Federal University (35 Kremlevskaya st., Kazan, Russia). The electronic version is available on the official websites of the Higher Attestation Commission of the Ministry of Education and Science of Russian Federation (vak2.ed.gov.ru) and Kazan Federal University (kpfu.ru).

Abstracts were distributed in August 7th 2015.

Scientific Secretary of the Dissertation Committee,
Doctor of Physical and Mathematical Sciences,
Professor,

Kamalova D.I.

GENERAL DESCRIPTION OF WORK

Relevance of the research topic

Ultraviolet (UV) lasers today find applications in many fields of science and technology. The most popular applications include environmental monitoring (as a part of lidar systems [1,2]), diagnostics of combustion processes in internal combustion engines [3], manufacture of semiconductor devices [4], precision machining [5], optical communication [6], photolithography [7], as well as medicine (in dermatology [8], cosmetology [9,10], eye surgery [11]) and biology [12]. The advantages of solid-state tunable lasers, capable to directly generate UV radiation, over the commercially available parametric oscillators and other sources of laser radiation, based on nonlinear conversion of laser wavelength, lie in their simplicity, reliability, durability and ability to increase output power by adding multiple amplifier stages while maintaining small size.

Search for UV active media based on optical transitions of the impurity ions is currently limited to the fluoride crystals (wide bandgap), activated by rare earth ions, trivalent cerium (the simplest energy level scheme) in particular. Lasers, based on cerium-activated fluoride crystals, generate radiation in the range of 280-340 nm. However, the first attempts to obtain lasing in these media have demonstrated, that intense pumping and lasing radiation induce in these materials various photodynamic processes (FDP) such as absorption from the excited state (ESA) of the impurity ions, photoionization of the impurity centers, formation and destruction of color centers (CC), and so on. These processes greatly contribute to the losses of laser generation, and the very possibility of lasing depends on the ratio between the cross-sections characterizing these processes and the cross-section of small-signal gain in the active medium. From this perspective, an understanding of the mechanisms, involved in their occurrence and behaviour, in the energy exchange between the localized levels of rare-earth ions and diffuse energy bands of the crystal lattice cations, and finding ways to avoid or to reduce the related losses, or even use them for good, is an **urgent task**.

Previously a crystallochemical approach [13] has been proposed to increase the stability of the optical properties of active media with respect to UV irradiation. It consists in variation of chemical composition of the medium (e.g., coactivation by Yb and Lu ions [14]) in order to arrange for additional channels of recombination for the photo-induced free carriers, that would compete with the processes of their capture by the lattice defects (formation of CC). Moreover, it is known that dynamic balance between the processes of formation and destruction of CC can be tipped in favor of

either of these processes varying experimental conditions. Thus, in [15] it was reported, that slope efficiency of laser oscillation in $\text{LiLuF}_4:\text{Ce}^{3+}$ crystal reaches its maximum when temperature of the active element is about 273 K. It has also been demonstrated [16], that to improve energy characteristics of the $\text{LiSAF}:\text{Ce}^{3+}$ laser additional irradiation of the active medium at 532 nm, bleaching the CC and thereby reducing losses in the channel of lasing, should be performed. Thus, a combination of crystallochemical and photophysical approaches looks rather promising when creating new active media for UV lasers, as well as of developing and deepening the extent of fundamental scientific knowledge in this field.

The Thesis was **aimed** at the development of a new and efficient solid-state active medium for quantum electronics of UV spectral range on the basis of scheelite-type fluoride crystalline materials doped by Ce^{3+} ions with manageable photodynamic processes.

Scientific novelty

1. For the first time **comprehensive studies** of PDP in $\text{LiY}_x\text{Lu}_{1-x}\text{F}_4:\text{Ce}^{3+}, \text{Yb}^{3+}$ ($\text{LYLF}:\text{Ce}, \text{Yb}$) crystals by means of optical and laser spectroscopy and photoconductivity have been **performed**; **spectral characteristics and values** of the essential parameters of photodynamic processes in these materials have been **estimated**; the nature of the ESA transition of Ce^{3+} ions has been **established**; the **position** of Ce^{3+} ions ground state relative to the bottom of the conduction band in $\text{LiY}_x\text{Lu}_{1-x}\text{F}_4:\text{Ce}^{3+}$ ($x = 0..1$) crystals has been **determined**; the most extensive to date **model** of PDP in the investigated active media has been **proposed**.

2. For the first time optimal **chemical composition** and optimum **conditions for lasing** in $\text{LiY}_x\text{Lu}_{1-x}\text{F}_4:\text{RE}^{3+}$ ($\text{RE} = \text{Ce}, \text{Yb}$) active medium has been **revealed**.

3. For the first time **experiments** to investigate the effect on the laser characteristics of pumping pulses repetition rate, chemical composition of the samples, temperature of the active element and application of additional irradiation of the pumped volume of the active element at wavelengths falling well within the bands of photoionization and CC absorption in $\text{LiY}_x\text{Lu}_{1-x}\text{F}_4:\text{RE}^{3+}$ ($\text{RE} = \text{Ce}, \text{Yb}$) active medium have been **conducted**; the potential of crystallochemical and photophysical approaches (temperature, irradiation and chemical composition of the active element) for the purpose of lasing performance manipulation by tipping the balance between various photodynamic processes in the active medium has been **demonstrated**.

Practical value

Optimal range of $\text{Y}^{3+} / \text{Lu}^{3+}$ ions concentrations and set of the optimum pumping conditions (pumping wavelength, decreased temperature and application of additional laser irradiation of the active element at 532 nm) for $\text{LiY}_x\text{Lu}_{1-x}\text{F}_4:\text{RE}^{3+}$

(RE = Ce,Yb) active media are determined, for which maximal for the particular pumping source and resonator slope efficiency and tuning range of laser generation are achieved. Developed comprehensive approach of photodynamic processes studies, combining methods of optical, laser spectroscopy and photoconductivity, allows to build the most complete to date model of photodynamic processes in the active medium, in order to assess its prospects as a UV laser active medium and to establish the optimal conditions for its excitation. The prospects of using photophysical approach for the management of photodynamic processes in active media, which can be applied in various devices of quantum electronics (laser element, Q-switch, saturable absorber, etc.), have been demonstrated.

Author defends

1. Photoconductivity arising in fluoride crystals with scheelite-type structure LYLf doped with Ce^{3+} ions in the range of 240-300 nm with a maximum at 270 nm is due to the ESA between 5d and 6s energy levels of Ce^{3+} ions.

2. Optimum conditions for lasing excitation on 5d-4f transitions of Ce^{3+} ions in fluoride crystals with scheelite-type structure LYLf are π -polarized laser pumping with a wavelength >290 nm, which corresponds to the lowest probability of Ce^{3+} ions photoionization.

3. $\text{LiY}_x\text{Lu}_{1-x}\text{F}_4$: Ce, Yb ($x = 0.5-0.3$, concentration of ions Ce^{3+} 0.3-1%) crystal is an efficient active medium for UV laser based on the 5d-4f transitions of Ce^{3+} ions.

4. Decreasing the temperature of the active element and application of additional laser illumination of its pumped volume at wavelengths falling well within the photoionization or CC absorption bands allows to reversibly vary the slope efficiency and the tuning range of lasing by tipping the balance between the processes of formation and destruction of CC.

Evaluation of the results

The main results of the Thesis have been personally reported and discussed at the following international and local conferences and symposia: XI, XVI and XVII All-Russian Youth Scientific School "The Coherent Optics and Optical Spectroscopy" (Kazan 2006, 2008 and 2013); International Conference on Excited States of Transition Elements (ESTE 2010), Wroclaw & Piechowice, Poland, 4-9 September, 2010; 11th Europhysical Conference on Defects in Insulating Materials (EURODIM 2010), Pecs, Hungary, 2-16 July, 2010; XV International Feofilov symposium on spectroscopy of crystals activated by rare-earth and transition metal ions, Kazan, Russia, 16-20 September, 2013; 18th International Conference on Dynamical Processes in Excited States of Solids (DPC'13), Fuzhou, China, 4-9

August, 2013; 17th International Conference on Luminescence and Optical Spectroscopy of Condensed Matter (ICL2014), Wroclaw, Poland, 13-18 July, 2014.

Publishing of the results

The main results are published in nine papers in the peer-reviewed journals [A1-A9]. Seven of these publications are in the international journals [A1, A3-A6, A8, A9], one publication is in the Russian periodical [A2], included in the list of scientific journals recommended by VAK, and one publication is in the international electronic journal [A7]. The results are also represented in the multiple conferences proceedings [A10-A26].

Personal contribution of the author

The most part of the presented in the Thesis experimental data was obtained in the laboratory of Quantum Electronics and Radiospectroscopy named after S.A. Altshuler of the Institute of Physics of Kazan Federal University. Photoconductivity studies using microwave technique as well as VUV spectroscopy results were obtained at the Claude-Bernard University Lyon 1 (France) in the Laboratoire de Physico-Chimie des Matériaux Luminescents (LPCML). All the experimental work (with the exception of excitation and luminescence spectra and decays under the synchrotron excitation that were implemented by K. Ivanovskikh), computer programming, numerical calculations and simulations, analysis of the experimental data and writing of the papers were personally done by the author of the Thesis. The author was actively involved in setting goals and objectives, planning of the experiments, and preparation of the samples for the research.

The structure and volume of work

Thesis consists of introduction, four chapters, conclusion, a list of abbreviations, list of references and the author's bibliography. The total volume of the Thesis (in Russian) is 192 pages, including 67 figures and 1 table. List of references contains 190 sources.

The **first chapter** provides an overview of the published works in the field of photodynamic processes and lasing in cerium-doped UV active media, used by the author for the analysis and interpretation of experimental data. The overview consists of five sub-sections, summarizing the amount of knowledge on the processes of excited-state absorption and photoionization of the impurity centers in Ce-activated active media, spectral characteristics of various types of CC in Ce-activated LiYF₄ (LYF) and LiLuF₄ (LLF) crystals, lasing and its parameters obtained in these active media, as well as on the width of the band gap in double fluoride scheelite-type

crystals and existing models of photodynamic processes induced in the active media by intensive UV pumping.

The **second chapter** presents the results of spectral and kinetic studies of the objects.

LYLF:Ce,Yb crystals were grown by the Bridgman-Stockbarger technique in the laboratory of Quantum Electronics and Radiospectroscopy of the Institute of Physics of Kazan Federal University by S.L. Korableva. After the orientation of single crystals using polarization microscope they were shaped as flat disks with a thickness of about 0.5-2 mm with 2 polished surfaces. The optical axis of the crystal was lying in the disk plane. Absorption, luminescence and luminescence excitation spectra of the samples in the UV spectral range were registered. It was found that increasing the concentration of Y^{3+} ions in the mixture leads to the monotonous shift of the peaks near 295 and 245 nm in the absorption spectra and both of the luminescence peaks towards shorter wavelengths, while the peaks of the absorption bands near 205, 195 and 185 nm shift towards longer wavelengths. Such a shift is due to increased repulsion between 5d sublevels when crystal field increases, which in turn is caused by the decrease in the lattice constant following the replacement of one cation of the lattice (Y^{3+}) by another (Lu^{3+}) with a smaller ionic radius.

Analysis of the recorded excitation spectra of luminescence and luminescence decays under synchrotron irradiation of samples in the energy range 4-19 eV shows correlations between excitation spectra and spectral distributions of luminescence decays fitting parameters. Fitting was performed using following function:

$$I(t) = I_{pile-up} + \sum_i I_i \cdot \exp\left(-\frac{t}{\tau_i}\right), \quad (1)$$

where I_i and τ_i is the amplitude and lifetime of the corresponding component of the luminescence decay, $I_{pile-up}$ – pile-up component of the decay, representing contribution of the long luminescence with a lifetime >100 ns (its contribution isn't clear in the decay itself due to the experimental limitations of the particular synchrotron operating regime). It was found that excitation within 4-9 eV leads to the photoionization of Ce^{3+} ions as a result of intracenter transitions from the lowest 5d excited state of Ce^{3+} ions to the sublevels located in the conduction band of the crystal. Under the high-energy excitation energy transfer to the trivalent cerium ions occurs mainly through various types of excitons associated with electronic transitions from the levels of fluoride ions, forming valence band, to the levels of Lu, Y and Li ions, forming conduction band. In the Lu-containing materials energy transfer from the excited Lu^{3+} ions, transitioned to

excited state as a result of intracenter 4f-5d and 4f-6s transitions of Lu^{3+} ions themselves, competes with the excitonic mechanism of energy transfer.

The **third chapter** is devoted to photoconductivity measurements in $\text{LYLF}:\text{Ce}, \text{Yb}$ crystals, and the results of PDP parameters calculations. Photoconductivity measurements were carried out using two proven techniques – conventional with blocked electrodes [17] and microwave [18] under the one- and two-photon excitation by laser pulses within spectral range of 225-330 nm. Photoconductivity spectra, recorded under the one-photon excitation, shown in Figure 1. It can be seen that the photoconductivity threshold falls at the wavelength of ~ 300 nm and photoconductivity spectrum consists of a band with a maximum at ~ 260 -270 nm.

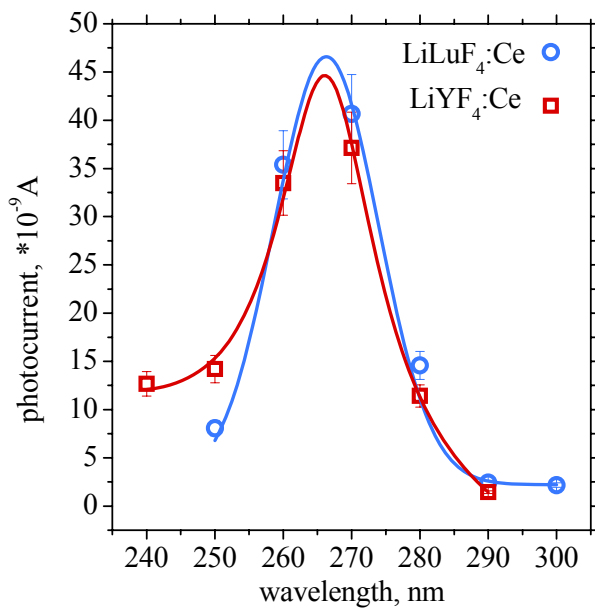


Figure 1a. Photoconductivity spectra in LiYF_4 and LiLuF_4 crystals registered by means of conventional technique.

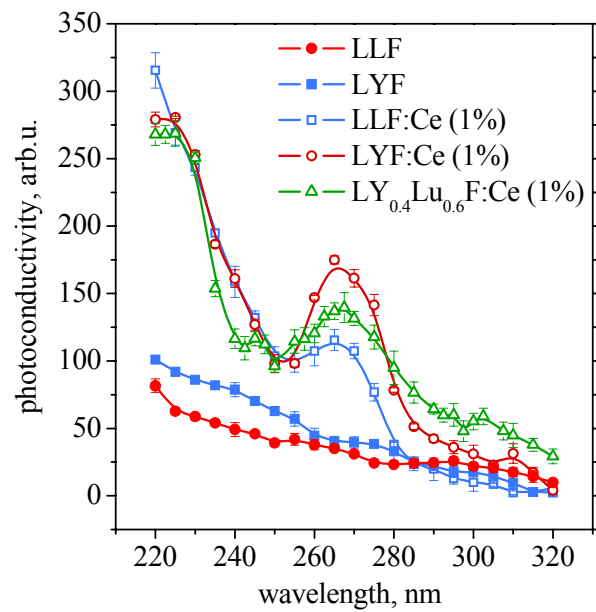


Figure 1b. Photoconductivity spectra in crystals $\text{LiY}_x\text{Lu}_{1-x}\text{F}_4$ ($x=0, 0.4, 1$), nominally pure and doped with Ce^{3+} ions, registered by means of microwave technique.

To ascertain the nature of this band, energy dependences of photoconductivity signal using both techniques were recorded. It can be seen (Figure 2) that the character of these dependences changes from square under the most long-wave excitation to linear and sublinear for the shorter excitation wavelengths. On the basis of four-level model of PDP (see. Figure 3) a system of differential kinetic equations (2) for the populations of these 4 levels was written. Steady-state (for the conventional photoconductivity measurements results) and nonsteady (for the microwave photoconductivity measurements results) solutions of the system (4) were found.

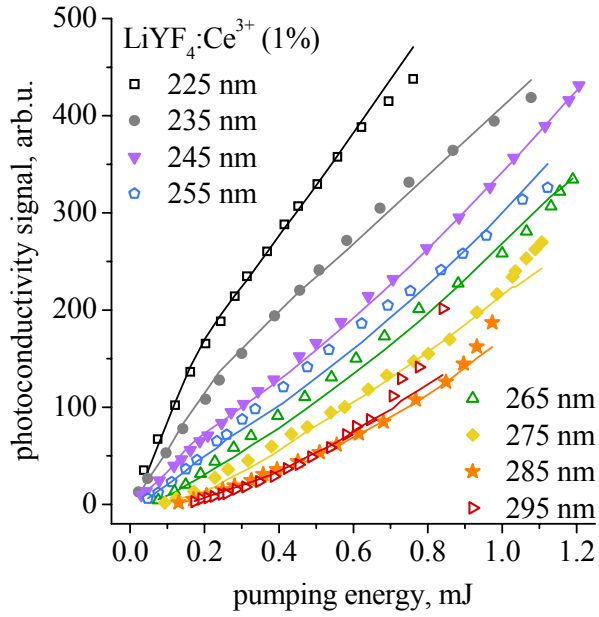


Figure 2. Energy dependences of photoconductivity excited in the range of 225-295 nm. Lines indicate the results of approximation

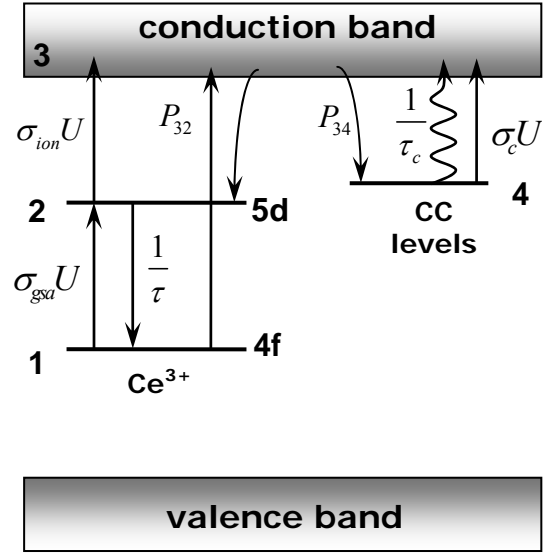


Figure 3. Four-level model of photodynamic processes

$$\begin{aligned}
 \frac{dn_1}{dt} &= -U(t, \lambda) \cdot \sigma_{gsa}(\lambda) \cdot n_1 + \frac{1}{\tau} \cdot n_2 - U(t, \lambda) \cdot \sigma_{dir}(\lambda) \cdot n_1 \\
 \frac{dn_2}{dt} &= U(t, \lambda) \cdot \sigma_{gsa}(\lambda) \cdot n_1 - \frac{1}{\tau} \cdot n_2 - U(t, \lambda) \cdot \sigma_{ion}(\lambda) \cdot n_2 + \sigma_{rec} \cdot v \cdot (n_3 + n_4) \cdot n_3 \\
 \frac{dn_3}{dt} &= U(t, \lambda) \cdot \sigma_{ion}(\lambda) \cdot n_2 + U(t, \lambda) \cdot \sigma_{dir}(\lambda) \cdot n_1 + \frac{1}{\tau_c} \cdot n_4 + \\
 &+ U(t, \lambda) \cdot \sigma_c(\lambda) \cdot n_4 - \left[\sigma_{rec} \cdot v \cdot (n_3 + n_4) + \sigma_{trap} \cdot v \cdot (n_c - n_4) \right] \cdot n_3 \\
 \frac{dn_4}{dt} &= \sigma_{trap} \cdot v \cdot (n_c - n_4) \cdot n_3 - \frac{1}{\tau_c} \cdot n_4 - U(t, \lambda) \cdot \sigma_c(\lambda) \cdot n_4 \\
 N &= n_1 + n_2 + n_3 + n_4
 \end{aligned} \tag{2}$$

where 1, 2 are ground and excited states of Ce^{3+} ions, 3 – conduction band of a crystal, 4 – CC levels; n_1, n_2, n_3, n_4 – populations of levels 1,2,3,4; $U(t, \lambda)$ – photon flux density of the exciting radiation at wavelength λ ; τ – radiative lifetime of the lowest 5d-state of Ce^{3+} ions; $\sigma_{gsa}(\lambda)$ – ground state absorption cross-section of Ce^{3+} ions; $\sigma_{ion}(\lambda)$ – ESA cross-section (cross-section of the two-step ionization) of Ce^{3+} ions; $\sigma_{dir}(\lambda)$ – Ce^{3+} one-step photoionization cross-section; $\sigma_c(\lambda)$ – absorption cross-section of color centers; $\sigma_{rec}, \sigma_{trap}$ – cross-section of recombination and trapping of the free carriers; v – velocity of free electrons in the conduction band; τ_c – the average lifetime of CC at the room temperature; n_c – concentration of traps; N – concentration of the impurity ions in the active medium.

To do this in the first case an analytical function, describing population n_3 of level 3, was derived from system (2), and in the second – the procedure of numerical solution and variation of the parameters of system (2) was implemented and the lowest value of the objective function using gradient descent method was achieved. Furthermore, since some of the parameters of system (2) are dependent and some are independent on the excitation wavelength, numerical solution and optimization procedure for the received decisions versus experimental data were carried out for all sets of data (for the various excitation wavelengths) at once. As a result of these calculations values and spectral distribution of the key parameters, which characterize PDP in the studied active media, were obtained (see. Figure 4, [1.A3]). Recombination cross-section turned out to be 2 orders of magnitude higher in LLF than in LYF ($\sim 10^{-13}$ and 10^{-15} cm² respectively). Moreover, it was necessary to include in the model a direct transition from ground 4f-state of cerium ions into the conduction band. The cross-section of such transition turned out to be 2-3 orders of magnitude smaller than the cross-section of Ce³⁺ photoionization from the excited state ($\sim 10^{-19}$ - 10^{-20} versus 10^{-17} cm²). This may mean that transition occurs in the short-wave edge of the absorption band, as it was shown in [19].

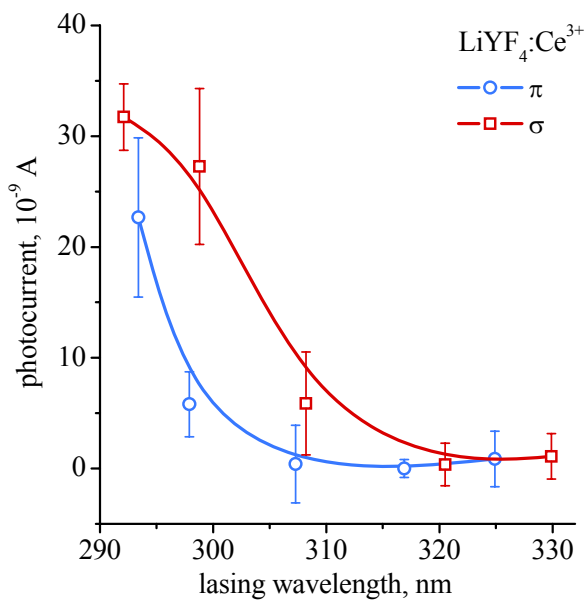


Figure 4a. Polarized photoionization spectra of Ce³⁺ ions in YLF:Ce crystal near the photoconductivity threshold

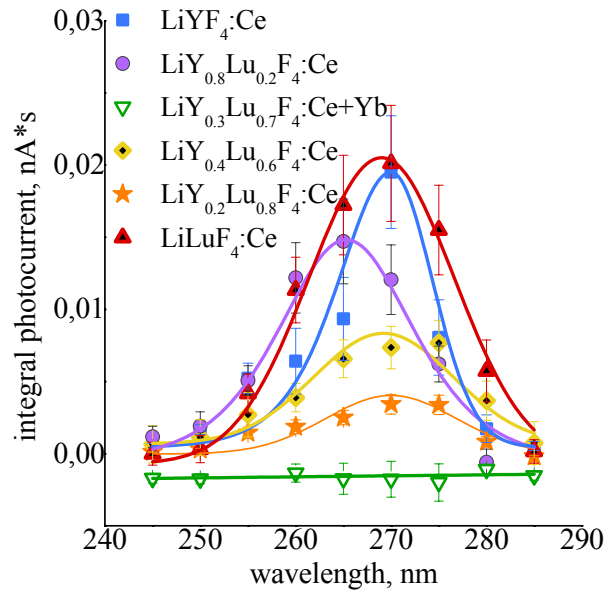


Figure 4b. π -polarized photoionization spectra of Ce³⁺ ions in the wavelength range of 245-285 nm (pumping – 300 nm)

Spectral distribution of Ce³⁺ photoionization cross-section values in the range of 240-300 nm (see. Fig. 4b) also turned out to be a band with a maximum near 265 nm and a HWHM ~ 20 nm. Since ground-state absorption spectrum and CC absorption spectrum don't reveal similar features, it is reasonable to assume, that the band in the photoconductivity spectra is due to the Ce³⁺ ions two-step photoionization

processes. This assumption was confirmed when photoconductivity spectra under the two-photon excitation were registered by means of conventional technique. For this purpose 300-nm pumping radiation was used in the ground-state absorption channel and probing radiation in the range of 240-300 nm was applied in the channel of the ESA of Ce^{3+} ions. The threshold of photoconductivity (Fig. 4a) appeared to be higher for the π -polarized light probing (~ 300 nm) than for the σ -polarized one. Excited-state photoconductivity spectrum also revealed a band, identical to the one registered in the photoconductivity spectra and calculated photoionization spectra of the trivalent cerium. In the sample, coactivated by Yb^{3+} ions, excited-state photoconductivity signal takes negative values in the entire spectral range of probing radiation. Since Yb^{3+} ions have no absorption bands in this region, the observed effect may be associated with intensifying of the recombination processes associated with the presence of ytterbium ions.

The position of the Ce^{3+} photoionization band's maximum in the spectrum is determined by the relative positions of the impurity ions' energy states and energy bands of the crystal. Since position of the band in the obtained photoionization spectra of Ce^{3+} ions, taking into account the quantum of the ground-state absorption to the lowest 5d level of Ce^{3+} , is in a good agreement with the position of the 4f-6s absorption band of Ce^{3+} ions (see. Fig. 6), it is reasonable to assume that this band in photoconductivity spectra with a maximum near 265 nm corresponds to the transition between isolated 4f and lying higher in the conduction band of the crystal 6s levels of Ce^{3+} ions. The square-law dependence of the photoconductivity signal on the excitation energy in the range of 275-305 nm reflects the two-step nature of that transition (through the lowest 5d level of Ce^{3+}).

Since contribution of the long-lived component in the luminescence decays of Ce^{3+} ions, when excited into the absorption bands of all the upper 5d states, is negligible, and lasing was obtained even under the pumping at ~ 240 nm, the upper 5d-states of ions Ce^{3+} , apparently, are positioned slightly below the edge of the conduction band of the crystal. An increase of the long-lived component's contribution begins with the excitation energies of ~ 7.2 eV, which is apparently due to the direct photoionization of Ce^{3+} ions into the conduction band. If the bandgap of LYF crystal is ~ 11.5 -12 eV then 4f state of Ce^{3+} ions is located ~ 4.5 eV above the valence band. This is consistent with the findings of Thiel based on the results of XPS studies in Tb:LYF crystal [20]. He found that the ground state of Tb^{3+} ions is situated ~ 2.9 eV above the valence band. Following the Dorenbos rule [21] the same value for Ce^{3+} ions should be ~ 4.4 eV.

Thus, photoconductivity measurements were successfully applied to the study

of PDP in LYLf: Ce³⁺ crystals in the range of 225 - 320 nm excitation wavelengths.

The **fourth chapter** describes the results of laser experiments in LiY_xLu_{1-x}F₄:RE³⁺ (RE = Ce, Yb) active media. Lasing was achieved in a flat-mirror Fabry-Perot resonator with longitudinal and transverse pumping and selective and non-selective geometry under the pulsed laser excitation (pulse duration ~10 ns, repetition rate 10 Hz) at the room and below the room temperatures. Resonator output mirror's reflectance was optimized to achieve the highest lasing power using selective and non-selective schemes (25% and 80% respectively). Maximum slope efficiency of lasing was ~22% in a non-selective geometry. Tuning of lasing wavelength in the range of about 305-335 nm was achieved, and consisted of 2 areas (see Fig. 5), corresponding to the short- and long-wavelength wings of Ce³⁺ luminescence spectrum (near 310 and 327 nm). In the range of about 317-323 nm lasing could not be observed despite the continuously positive gain throughout the entire spectral region (302-338 nm) [A8]. In the mixed LiY_{0.3}Lu_{0.7}F₄:Ce crystal, unlike LLF:Ce and the samples coactivated with Yb³⁺ ions, lasing in the long-wave wing of the luminescence spectrum could not be obtained at all.

Laser characteristics of the samples under the pumping at 300 and 290 nm were investigated. In the first case a slope efficiency of lasing didn't vary over the entire range of excitation energies near the both peaks of luminescence spectrum. Under 290-nm pumping a dip in the long-wavelength wing of the tuning characteristic was revealed (see. Fig. 5). In addition the slope efficiency of lasing at 310 nm changed when a certain level of pumping energy was achieved. For the lasing at 327 nm the dependence of lasing energy on the pumping energy saturates and exhibits "hysteresis" when the pumping energy is being increased and then decreased [A5], probably due to the processes of the long-lived CC accumulation.

Tuning characteristics of π - and σ -polarized radiation components of lasing at 310 nm under the excitation at 300 nm show, that in the long wing of the tuning range σ -polarized radiation dominates over the π -polarized one (see. Fig. 6), although the geometry of the resonator favors π -polarized lasing. Lim and Hamilton found that for Ce:LYF crystals there is an absorption band peaking near 340 nm, much more intense for π -polarized light than for σ -polarized one, which they attributed to the long-living F-type CC. Therefore, these centers effectively absorb π -polarized laser radiation of cerium ions in this spectral range.

Thus, taking into account the previously obtained excited-state photoionization spectrum of Ce³⁺ ions in the investigated active media, it can be argued that in order to achieve lasing in LYLf:Ce crystals it seems reasonable to reduce the efficiency of

PDP, choosing the source of pumping radiation with a wavelength longer than 290 nm, where the probability of Ce^{3+} ions photoionization is minimal.

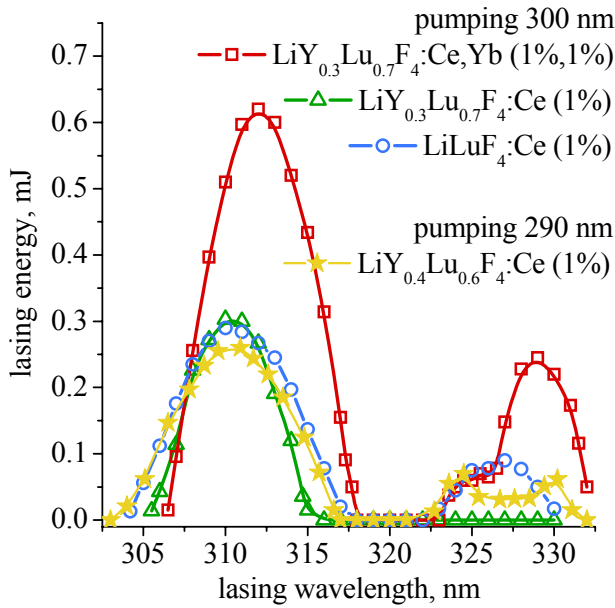


Figure 5. Tuning of lasing wavelength in LYLF:Ce,Yb crystals

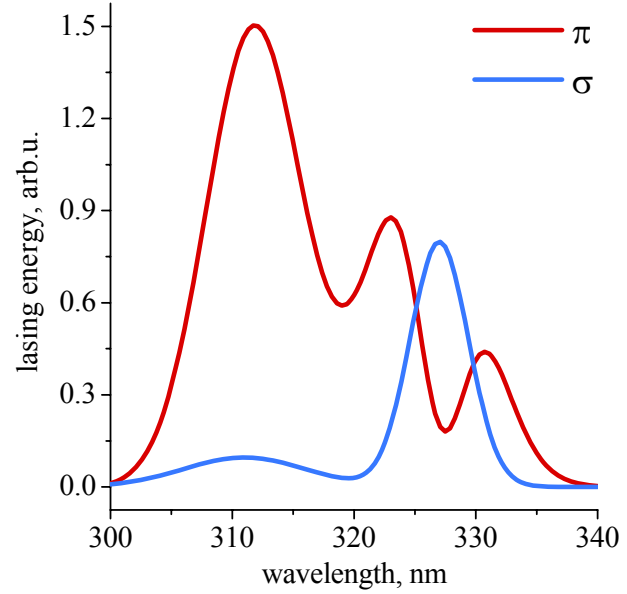


Figure 6. π and σ -polarized components of tunable lasing registered in LLF:Ce (0.1 at.%) crystal

Further experiments were conducted to obtain lasing when cooling the active element to the temperatures below 0 °C. It can be seen (Fig. 7) that cooling of the active element increases energy and slope efficiency of laser generation as well as widens the tuning range of lasing wavelength. Besides, in the mixed crystal noncoactivated with Yb^{3+} ions laser oscillation in the long-wavelength wing of tuning range was finally achieved. This, dependence of laser characteristics on the temperature of the active element can be explained by narrowing of vibrationally-broadened photoionization or CC absorption spectra, or by the decreased efficiency of thermalization of the impurity ions excited states located near the bottom of the conduction band of the crystal [22].

Finally the experiments to explore the effect of pumping pulse repetition rate and additional illumination of active element on laser characteristics were conducted. 100-time reduction of pumping pulse repetition rate didn't significantly affect either energy or slope efficiency of laser oscillation. Therefore, long-lived CC don't have enough time to be thermally bleached between the two sequential pumping pulses.

Additional illumination of laser element was performed at 532, 340 and 266 nm with energy density, comparable to the one of pumping radiation at 300 nm. Illumination wavelengths were selected in accordance with absorption spectra of various types of color centers, observed in the YLF and LLF crystals, and taking into account previously obtained photoionization spectrum of cerium ions. Radiation of the additional illumination

was generated by the same laser that was used as a source of pumping radiation, thus allowing to avoid problems with the synchronization of different sources of radiation.

Both energy and slope efficiency of lasing were greatly reduced when the illumination at 266 nm was turned on. In the mixed crystal, non-coactivated with Yb^{3+} ions, lasing was completely suppressed. 266 nm falls well near the peak of the two-step photoionization spectrum of Ce^{3+} ions, as well as in the absorption band of V_k centers. Analysis of the energy dependence of 266-nm absorption in the sample's pumped (by 300 nm) volume revealed that 266-nm radiation is absorbed mainly due to the excited-state absorption of Ce^{3+} ions [A9].

To shift the dynamic equilibrium in the active medium towards the increase of UV-induced CC bleaching processes, 340-nm and 532-nm irradiation of the pumped volume of the active element was performed. When 532-nm radiation was turned on, energy and slope efficiency of lasing increased significantly (see. fig. 7), while tuning range of the laser wavelength became continuous throughout the whole tuning range (see. Fig. 8).

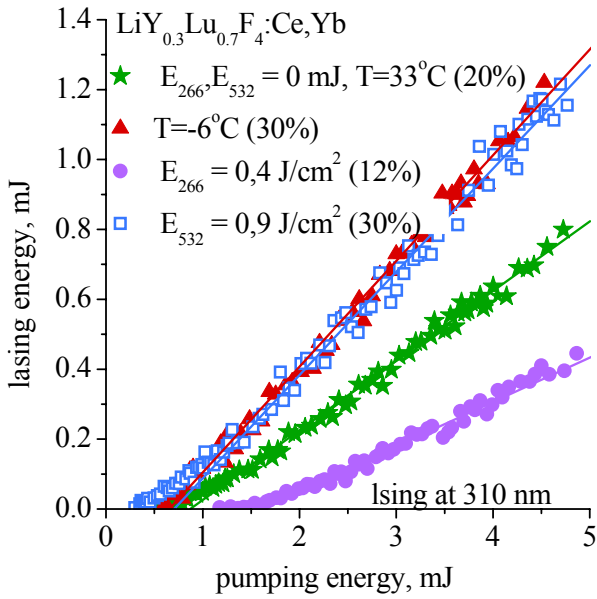


Figure 7. Slope efficiency of lasing at 310 nm in $\text{LiY}_{0.3}\text{Lu}_{0.7}\text{F}_4:\text{Ce},\text{Yb}$ crystal (impurity concentration ~ 1 at.%) for the various lasing conditions

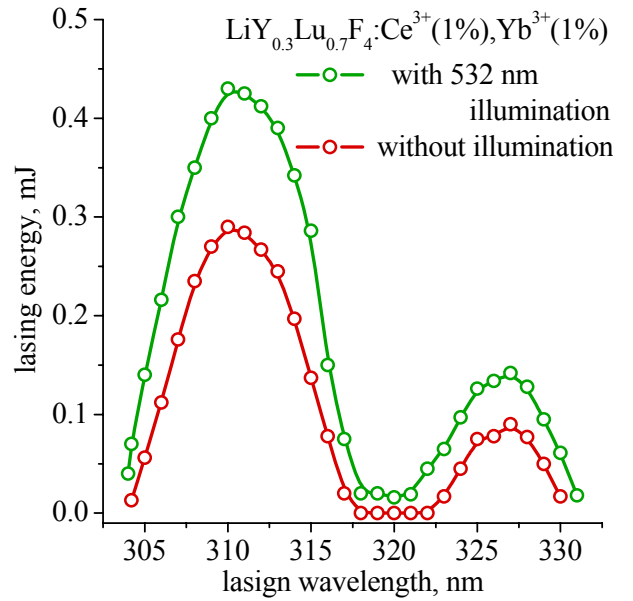


Figure 8. Tuning range of lasing with and without additional irradiation of the pumped volume of the active medium at 532 nm

Irradiation at 340nm, against expectations, didn't lead to any improvement, but to a deterioration of lasing output, although it was anticipated that 340-nm illumination would bleach the F-centers. This in turn should have resulted in return of the trapped photoinduced electrons back to the upper laser level. However, when the illumination of the active element was applied the energy of lasing in the short wing of the tuning curve decreased, and in the long wing it was completely suppressed. Since trivalent cerium ions have no absorption bands in this region, it is reasonable to assume that the observed effect is due to the absorption of CC.

Ground states of the long-lived F-centers according to [19] are the most deeper-lying in the bandgap. Absorbing 340-nm radiation, these centers are being promoted to the excited states, but not bleached. That is, captured photoelectrons cannot be released back into the conduction band and further participate in lasing. However, these F-centers' excited states lay not deeper than 3000 cm^{-1} below the bottom of the conduction band. Hence captured electrons can be nonradiatively transferred to the less deeply-lying levels of the other types of color centers (activation energy ~ 4200 and 4500 cm^{-1}), identified in [19], with the lifetimes of the order of micro-, milliseconds and seconds. These short-lived centers, apparently, absorb generated laser radiation much more efficiently than the F-centers (see. model in [A9]).

Thus, dynamic losses of laser oscillation occur mainly due to the short-lived CC. The possibility to influence processes of their formation (decreasing effectiveness of PDP processes by means of cooling of the active element) or effectively bleach them while the pumping pulse lasts (irradiation of the pumped volume of the active element at 532 nm, 340 nm and 266 nm), tipping the balance between the processes of lasing and CC formation in one direction or another, have been demonstrated here. Long-lived CC only cause the initial reduction of lasing energy by $\sim 30\%$.

Best laser characteristics were achieved in LLF:Ce sample. However, the mixed crystals LYLf:Ce,Yb ($x = 0..0.5$) demonstrated similar values of slope efficiency and width of the tuning curve. Yb-coactivated mixed samples demonstrated higher photochemical stability and, hence, better laser characteristics as compared with the non-coactivated ones. Laser characteristics in all investigated media can be substantially improved by cooling the active element to the temperatures below $0\text{ }^{\circ}\text{C}$ and by providing additional illumination of the pumped volume of the active element by laser radiation at 532 nm with approximately the same level of energy density as that of the pumping radiation.

In **conclusion**, the main results of the research are summarized:

- absorption, luminescence, excitation of luminescence and photoconductivity spectra under the UV excitation in $\text{LiY}_x\text{Lu}_{1-x}\text{F}_4:\text{RE}^{3+}$ ($\text{RE} = \text{Ce}, \text{Yb}$) crystals were **registered**;
- values and spectral distributions of the key parameters of the photo-induced processes (ionization cross section of the activator ion, probability of trapping and recombination of the charge carriers, CC absorption cross-section) are **determined**;

- it was **established** that ground 4f-state of Ce^{3+} ions in fluoride crystals with scheelite-type structure $\text{LiY}_x\text{Lu}_{1-x}\text{F}_4:\text{RE}^{3+}$ ($\text{RE} = \text{Ce}, \text{Yb}$) is located ~ 7.2 eV below the bottom of conduction band;
- the most complete to date model of photodynamic processes in the investigated media is developed; position of the 4f ground state of Ce^{3+} ions relative to the conduction band of the crystal is **established**;
- lasing in $\text{LiY}_x\text{Lu}_{1-x}\text{F}_4:\text{RE}^{3+}$ ($\text{RE} = \text{Ce}, \text{Yb}$) crystals in ultraviolet spectral range is **obtained**; optimization of the reflection coefficient of the resonator's output mirror is **performed**;
- lasing characteristics (slope efficiency, lasing threshold, wavelength tuning range), depending on the experimental conditions (temperature of the active element, wavelength of additional irradiation of the pumped volume of the active element, pumping pulse repetition rate) are **determined**;
- recommendations on the management of PDP, induced by UV radiation in scheelite-type fluoride crystals, doped with trivalent cerium, are **suggested**.

Following **conclusions** are drawn:

- electronic excitations, initiated in $\text{LiY}_x\text{Lu}_{1-x}\text{F}_4:\text{Ce}^{3+}$ crystals under excitation in the range of 4-18 eV, lead to the photoionization of Ce^{3+} ions and following energy transfer to the cerium ions through the excitons of different nature;
- photoconductivity, occurring in $\text{LiY}_x\text{Lu}_{1-x}\text{F}_4:\text{Ce}^{3+}$ crystals under the UV radiation and characterized by the band with a maximum near 265 nm, is due to the intracenter Ce^{3+} transitions between the lowest excited 5d and located in the conduction band 6s energy levels of cerium ions;
- technique to study PDP, which consists in registration of the dependencies of photoconductivity on the optical excitation energy in a certain spectral range and subsequent fitting of these dependencies, including the optimization procedure of the received solutions with respect to the experimental data, allows to evaluate the fundamental parameters of the medium and PDP;
- $\text{LiY}_x\text{Lu}_{1-x}\text{F}_4:\text{Ce}, \text{Yb}$ ($x = 0.5-0.3$, concentration of Ce^{3+} ions 0.3-1%) crystals are efficient active media for UV lasers, characteristics of which can be significantly improved by additional irradiation of the pumped volume of the active element at 532 nm or cooling the active element down to the temperatures about -20 °C;
- variation of slope efficiency and tuning range of laser generation under additional irradiation at 532 and 340 nm is due to the shift of equilibrium between the processes of formation of different types of CC, while the effect of 266 nm irradiation and cooling of the active element is due to the decrease of the efficiency of ESA.

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