Ultrashort pulsed UV lasers based on the Ce$^{3+}$:LiCaAlF$_6$ and LiLuYF$_4$:Ce$^{3+}$ crystals

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Abstract. Here we report on UV laser oscillation on colquiriite structure fluoride mixture crystals of Ce$^{3+}$:LiCaAlF$_6$ (Ce:LiCAF) and Ce$^{3+}$:LiLuYF$_4$ (Ce:LiLuYF) active media. We report on 800 ps pulse train lasing from Ce:LiCAF in composite cavity and single pulse lasing of 400 ps duration from Ce:LiLuYF in convenient Fabri-Perrot cavity and discuss conditions of pulse shortening in UV active medium by photodynamic processes.

An important problem of quantum electronics is the generation of laser radiation in the form of ultrashort pulse in UV spectral range. The family of cerium-doped laser crystals [1] offer an attractive all-solid-state route providing valuable tunable laser oscillation in the UV directly with simplications and attractive properties of solid-state laser (beam divergence, energy distribution) which is problematic for nonlinear converters. Here we report on UV laser oscillation on colquiriite structure fluoride mixture crystals of Ce$^{3+}$:LiCaAlF$_6$ (Ce:LiCAF) and Ce$^{3+}$:LiLuYF$_4$ (Ce:LiLuYF) active media which can be pumped at around 266 nm, and so can utilize frequency-quadrupled Nd-based pump lasers [2].

For the first time lasing on the Ce:LiCAF crystal was reported by Dubinskii [3]. Broad gain band of Ce:LiCAF crystal from 280 to 315 nm allows amplification and generation of femtosecond pulses. Using the 4-pass amplifier based on the Ce:LiCAF crystal Zhenlin [4] report about amplification of femtosecond pulse of 115 fs duration to 370 times at a wavelength of 290 nm.

Mathematical modeling conducted for the first time in [5] showed significant reduce of the generated pulses duration, with proper selection of the parameters of the laser cavity and the pump level. The conditions of reduced pulse duration are high exceeding of the population inversion in the active medium over the lasing threshold and large ratio of the pump pulse duration to the lifetime of photons in the cavity [5]. However, the color centers (CC) formation under pump radiation in the active medium was not considered in this paper while CC have absorption bands at the lasing wavelength and affect at its evolution.

Effect of CC on the characteristic of the Ce:LiCAF lasers were discussed earlier [6] and it has been shown that with increasing pump energy total intracavity losses decrease due to CC. This was explained by the color centers formation under the pump radiation and their subsequent destruction by the laser radiation. Color centers absorption is getting lower with higher amount of laser radiation remaining in the cavity [6].

In the present work modeling of the laser oscillation was carried out considering CC formation and the mechanism of laser pulse shortening due to CC was investigated.
The model contains conventional four-level scheme, the conduction band of the crystal and the CC level (fig. 1).

**Figure 1.** The level scheme of the UV active medium corresponding to four-level lasing scheme and formation of color centers. Crucible also participates in the formation of a temperature gradient. The wall thickness of the crucible should be minimal (in order 2-3mm) to distort the temperature field as little as possible.

In accordance with this scheme system of differential equations were written (1-8), which are solved by computer software Matlab. Lasing simulation results are shown in Figure 2.

$$\frac{dn_1(t)}{dt} = N_{pump}(t)\sigma_{14}\left(n_4(t) - n_1(t)\right) + \frac{n_2(t)}{t_{21}}$$  \hspace{1cm} (1)

$$\frac{dn_3(t)}{dt} = q(t)\sigma_{32}\left(n_3(t) - n_2(t)\right) - \frac{n_1(t)}{t_{21}} + \frac{n_3(t)}{t_{32}}$$  \hspace{1cm} (2)

$$\frac{dn_4(t)}{dt} = \frac{n_4(t)}{t_{43}} - q(t)\sigma_{32}\left(n_3(t) - n_2(t)\right) - \frac{n_4(t)}{t_{32}} - \sigma_{ESA}N_{pump}\left(n_4(t) - n_{CB}(t)\right)$$  \hspace{1cm} (3)

$$\frac{dn_{CB}(t)}{dt} = N_{pump}(t)\sigma_{ESA}\left(n_4(t) - n_{CB}(t)\right) - \frac{n_{CB}(t)}{t_{CB}} - \frac{n_{CC}(t)}{t_{CBCC}} + \sigma_{CC}n_{CC}(t)\left(q(t) + \frac{n_3(t)}{t_{32}}\right)$$  \hspace{1cm} (4)

$$\frac{dn_{CC}(t)}{dt} = \frac{n_{CB}(t)}{t_{CBCC}} - \sigma_{CC}n_{CC}(t)\left(q(t) + \frac{n_3(t)}{t_{32}}\right) - \frac{n_{CC}(t)}{t_{CCCB}}$$  \hspace{1cm} (5)

$$\frac{dq(t)}{dt} = q(t)\left(\sigma_{S2}\left(n_3(t) - n_2(t)\right) - \sigma_{CC}n_{CC}(t)\right)c - \frac{q(t)}{t_c}$$  \hspace{1cm} (6)

$$N_{Ce} = n_1 + n_2 + n_3 + n_4 + n_{CC} + n_{CB}$$  \hspace{1cm} (7)

Where $n_1 - n_4$, $n_{CB}$, $n_{CC}$ – corresponding population on Ce$^{3+}$ energy levels, CC and conduction band; $N_{pump}$ and $q$ – pump and lasing photon flux density; $\sigma_{14}$ and $\sigma_{ESA}$ – absorption cross section and excited state absorption cross section on pump wavelength; $\sigma_{32}$ and $\sigma_{CC}$ – gain cross section and CC absorption cross section on lasing wavelength; $\frac{1}{t_{21}}, \frac{1}{t_{32}}, \frac{1}{t_{43}}, \frac{1}{t_{CB}}, \frac{1}{t_{CBCC}}, \frac{1}{t_{CCCB}}, \frac{1}{t_{CC}}$ – probabilities of the corresponding transitions; $c$ – speed of light; $L$ – cavity length; $N_{Ce}$ – concentration of Ce$^{3+}$ ions.
The simulation results considering photodynamic processes

In the simulation procedure the parameters of the active medium and short cavity were considered corresponding to low Q-factor. As a result, we have obtained the case of single ultrashort pulse generation with duration of 400 ps. According to the CC populations graph (black curve named "losses") it is clear that CC in the bulk this medium work as saturable absorber (SA). Namely pump radiation induces the CC formation, which leads to higher losses thus forms the SA. When laser radiation appears it starts bleaching the CC and this process corresponds to saturation of SA. This leads to sharp increase in Q-factor of the cavity and determines conditions for the ultrashort pulses generation.

Laser experiment scheme: 1 – pump source (Nd :YAG 266 nm / Raman-shifter 300 nm); 2 – active medium (Ce:LiCAF / Ce:LiLuYF); 3, 4 – flat mirrors (R=25% on lasing wavelength), cavity length 1.5 cm; 5 – extra mirror of the cavity.

In order to test this mechanism of laser pulse shortening the experiments were carried out in a short low-Q cavity with Ce:LiCAF and Ce:LiLuYF crystals as active elements. The experimental scheme is shown in Figure 3.

We have carried out experiments on generation of subnanosecond pulses on Ce:LiCAF crystal with 266 nm 10 ns pumping form Nd:YAG laser. In these experiments we have not achieved stable generation of single ultrashort pulse at any pump energy but using composite cavity we have achieved stable “pulse-train” generation. Experimental results are presented in fig.4a.
The composite laser cavity consists of a short length low Q-factor pulse-seeding laser cavity and a feedback laser cavity. The pulse-seeding laser cavity provides only spiking mode of laser oscillation. Regenerative amplification was organized by aligning the axes of seeding cavity and long feedback cavity [1,2], and as a result we see a stable ultrashort “pulse-train” with pulse duration at about 800 ps and a period defined by the design of the cavity. It is important that length of the external cavity should be tuned so that the second pulse of “pulse-train” occur before generation of the second stochastic pike. In this case the lasing pulse is circulating inside the external laser cavity harvests the inversion and regenerative amplification prevents stochastic laser oscillation.

In a laser based on Ce:LiCAF crystal the mechanism of laser pulse shortening due to CC is not working, possibly due to the low level of losses associated with the CC formation. However, losses in the Ce:LiLuYF crystal associated with CC may exceed the same losses in the Ce:LiCAF crystal 5 times [6,7].

In experiments using LiLuYF crystal in low-Q short cavity we have obtained a single ultrashort pulse generation of 400 ps duration with pump radiation at the wavelength 300 nm obtained from Raman-shifter pumped by Nd:YAG laser. Therefore, we have proved the possibility of use of photodynamic processes for intracavity losses modulation. Internal UV pump induced losses in active medium were utilized to shorten the laser pulse.

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