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Pump-probe studies of absorption saturation and optical gain in Ce:LiCaAlF$_6$ ultraviolet active medium

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Abstract. Aim of this work was to investigate nonlinear absorption of pump radiation in UV spectral range of Ce^{3+}:LiCaAlF$_6$ crystalline active medium. We were able to evaluate excited state absorption cross-section at pump wavelength (266 nm) and recombination probabilities by means of computer simulation.

1. Introduction

Ultraviolet (UV) spectral range appears to be fruitful in many applications as precise materials processing, analytic equipment and remote atmosphere control. But question of providing stable and reliable lasers here is still opened. From this point of view Ce^{3+} based active media experience interest because it is always prospective to achieve laser oscillation in required spectral range directly in order to manage characteristics of laser radiation by design of laser cavity [1].

Pump-probe spectroscopy appears to be one of prospective ways to investigate the potential of novel laser active media as this method allows quantitative estimation of its abilities in laser oscillation. At the same time implementing computer simulation methods to pump-probe results can give important information on dynamic processes in active media [2]. Further computer simulation of active media can give estimates of power and temporal characteristics of potential laser oscillators on the basis of investigated material.

Aim of this work was the investigation of nonlinear absorption of pump radiation and optical amplification in UV spectral range of Ce^{3+}:LiCaAlF$_6$ crystalline active medium and interpretation of experimental results on the basis of computer simulation with absorption of pump radiation from excited lasing level included in the model.

2. Experimental technique and samples

Pump-probe experiments were performed with Nd:YAG laser from Lotis TII company as a pump source. We used quadruple (266 nm) radiation to pump our Ce^{3+}:LiCaAlF$_6$ samples. Probe radiation was 289 nm lasing from Ce:LiCaAlF$_6$ laser by Ultraviolet Solutions LLC which, in order to eliminate problem of synchronizing pump and probe pulses, was pumped by the same 266 nm radiation. Delay between pump and probe pulses was controlled with use of Tektronix oscilloscope and fast detectors from Alphalas. Absorption coefficient and optical gain coefficient dependences on pump radiation flux were achieved with use of optical attenuators. Samples of Ce^{3+}:LiCaAlF$_6$ crystals were Brewster-cut plane-parallel plates 5 mm thick with two laser grade polished windows for longitudinal scheme of pump-probe experiment. Ce^{3+} ions content was 1 at.% in the melt. Samples were prepared from material grown in Kazan Federal University.

3. Pump-probe experiments

In the figure 1 an absorption saturation of Ce:$LiCaAlF_6$ crystal of 266 nm pump radiation is shown. Although absorption is traditionally decaying with pump flux, in our case the saturation appears at fluxes higher than it should be for interconfigurational 4f-5d transitions of Ce^{3+} ions characterized by high probabilities. Optical gain coefficient dependence on pump radiation flux for 289 nm probe radiation in Ce:$LiCaAlF_6$ crystal is shown in figure 1b.

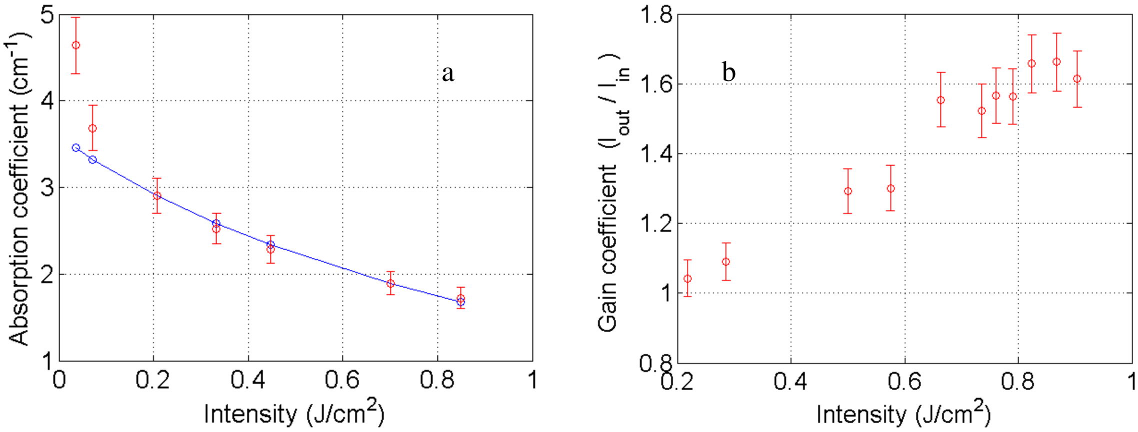

Figure 1. Absorption coefficient at 266 nm (a) and gain coefficient (b) at 289 nm dependence of Ce^{3+}:$LiCaAlF_6$ crystal on 266 nm pump radiation flux.

We can see that gain is increasing linearly with pump radiation flux and do not exhibit any saturation. Such dependence is characteristic for most of laser active media and evidences absence of processes of crystals optical properties degradation. Experimental errors in figure 1b are due to strong dependence of the gain coefficient on matching of pumped and probed volumes of the crystal.

4. Ce:$LiCaAlF_6$ active medium model and computer simulation

Ce^{3+} based active media amplify UV light due to interconfigurational 5d-4f transitions forming four-level scheme of lasing. But 266 nm photons used as a pump and ultraviolet photons of laser radiation often cause high probability of excited state absorption transition terminating in conduction band of the host lattice [3]. In order to interpret correctly the results of pump-probe experiments and to determine parameters of dynamic processes in Ce:$LiCaAlF_6$ active medium we have used a well known stochastic model of Ce^{3+} active medium shown in figure 2 [4,5].

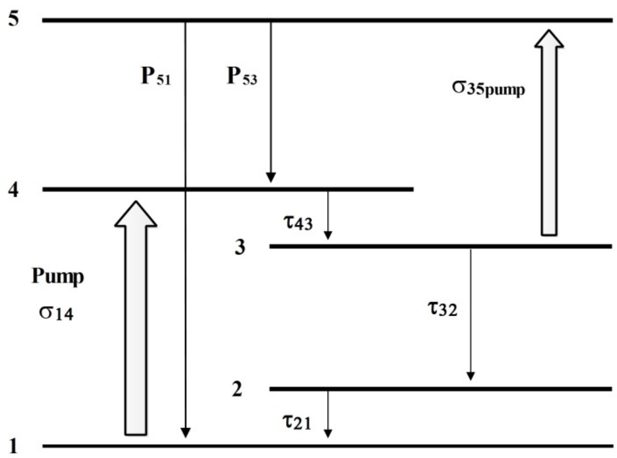

Figure 2. Stochastic model of Ce:LiCaAlF$_6$ active medium with four-level scheme of lasing based on 5d-4f transitions of Ce^{3+} ions and a level of host lattice conduction band at which excited state absorption of pump radiation transition terminate.

Our model included population inversion processes of pump radiation absorption from the lowest 4f state to excited 5d state with cross-section σ_{14} and fast thermalization at 5d state (probability $1/\tau_{43}$), processes of excited state depopulation by luminescence (probability $1/\tau_{32}$) and fast nonradiative transition to the lowest 4f state (probability $1/\tau_{21}$). Also σ_{35} is a cross-section of excited state absorption of pump radiation. Processes of relaxation of the system from conduction band are described here by probabilities P_{51} and P_{54} [5]. They correspond to recombination processes at the site of Ce^{3+} ion and leave it in the ground state and excited state, respectively. These processes make our system closed. We did not include color centers related processes in our model since we did not evidence any degradation of optical properties which could speak for accumulating of color centers absorbing incident radiation. Kinetic equations corresponding to our model are as follows:

$$\frac{d}{dt}n_1 = -Pump(t)\cdot\sigma_{14}\cdot n_1 + \frac{n_2}{\tau_{21}} + Pump(t)\cdot\sigma_{14}\cdot n_4 + P_{51}\cdot n_5 \quad (1)$$

$$\frac{d}{dt}n_2 = -\frac{n_2}{\tau_{21}} + \frac{n_3}{\tau_{32}} \quad (2)$$

$$\frac{d}{dt}n_3 = -\left[Pump(t)\cdot\sigma_{35} + \frac{1}{\tau_{32}}\right]\cdot n_3 + \frac{n_4}{\tau_{43}} + Pump(t)\cdot\sigma_{35}\cdot n_5 \quad (3)$$

$$\frac{d}{dt}n_4 = Pump(t)\cdot\sigma_{14}\cdot n_1 - \left[Pump(t)\cdot\sigma_{14} + \frac{1}{\tau_{43}}\right]\cdot n_4 + P_{54}\cdot n_5 \quad (4)$$

$$\frac{d}{dt}n_5 = Pump(t)\cdot\sigma_{35}\cdot n_3 - [Pump(t)\cdot\sigma_{35} + P_{54} + P_{51}]\cdot n_5 \quad (5)$$

Here $n_1 - n_5$ are populations at corresponding states, *Pump*(*t*) – photon flux of pump radiation given by Gaussian temporal distribution. At any moment of system evolution the sum of populations of five levels was considered to be equal to Ce^{3+} ions content N_{Ce}.

Optimization procedure consisted in search for minimal difference between experimental data on absorption and gain coefficients and data derived from computer simulation by varying parameters of dynamic processes σ_{35}, P_{51}, P_{54}. Also we varied Ce^{3+} ions content N_{Ce} as LiCaAlF$_6$ host lattice have several sites where Ce^{3+} ions could be incorporated and distribution of these sites is unknown. Parameters of spontaneous emission and gain cross-section are known from work [6], probabilities of nonradiative transitions $1/\tau_{43}$ $1/\tau_{32}$ are very high and were fixed at the level of 10^{12} s^{-1}.

Absorption coefficient computer simulation results are shown in figure 1a. Dynamic processes parameters corresponding to these results are presented in the table 1.

Table 1. Dynamic processes parameters evaluated from Ce:$LiCaAlF_6$ active medium absorption saturation by means of computer simulation

Parameter	Value
Pump radiation (266 nm) excited state absorption cross-section, cm^2	$(3,2\pm0,8)\cdot10^{-18}$
Probability of free charge carrier recombination at the Ce^{3+} site, s^{-1}	$(6,3\pm1,3)\cdot10^{8}$
Probability of free charge carrier recombination at the Ce^{3+} site terminated at excited state of Ce^{3+}, s^{-1}	$(3,7\pm1,2)\cdot10^{8}$
Ce^{3+} ions content, cm^{-3}	$(4,5\pm0,5)\cdot10^{17}$

One can see poor agreement of experimental and simulated absorption coefficients at lower pump fluxes. We explain this by the influence of pump-induced color centers which add their absorption significantly when lower pump flux do not bleach them. Excited state absorption cross-section of Ce^{3+} ions derived by us at 266 nm wavelength in $LiCaAlF_6$ has a value which is typical for Ce^{3+} doped crystals [5,6] but it is not higher than a gain cross-section, and this is proved by stable UV laser action for this active medium with 266 nm pumping. Probabilities of free charges recombination at Ce^{3+} site both have values of the same order as spontaneous 5d-4f emission of Ce^{3+} which is fast enough to make no influence on loss accumulation in the active medium but causes loss in laser efficiency when part of excitation is wasted to induce free charge carriers in conduction band and do not take part in creating the population inversion due to high rates of stimulated emission transitions. Also we were able to determine Ce^{3+} ions content for sites absorbing at 266 nm. The obtained value corresponds to low segregation coefficient of Ce^{3+} ions in $LiCaAlF_6$ crystals which leads to absorption coefficient of about 5 cm^{-1} at 266 nm for 1 at. % of Ce^{3+} in the melt.

5. Conclusion

In this work the results of nonlinear absorption and optical gain studies in Ce:$LiCaAlF_6$ active medium are presented. With the help of computer simulation of four-level scheme of Ce^{3+}-based active medium with excited state absorption included in the model we were able to evaluate parameters of excitation losses from excited 5d level of Ce^{3+} at pump wavelength and parameters of free charge carriers recombination. We have revealed that laser action efficiency is affected by excited state absorption in Ce:$LiCaAlF_6$ but remaining laser oscillation could be stable due to the absence of losses accumulation effect. Our results and computer simulation methodic could be implemented in further works on design of laser oscillators on the basis of Ce:$LiCaAlF_6$ active medium.

Acknowledgments
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