

Optical and gain properties of series of crystals $\text{LiF–YF}_3\text{–LuF}_3$ doped with Ce^{3+} and Yb^{3+} ions

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Abstract

Here, we present first results of systematic studies of host cation variation impact on spectral-kinetic, photochemical and gain properties of Ce^{3+} -doped LiYF_4 (YLF), LiLuF_4 (LLF) and $\text{LiY}_{1-x}\text{Lu}_x\text{F}_4$ family crystals. 5d–4f luminescence decay of Ce^{3+} ions studies, together with pump–probe experiments, indicate that previously reported twice higher luminescence quantum yield in LLF compared with that of YLF crystals is provided by more efficient upper lasing level feeding due to recombination and higher color center destruction rate in LLF against YLF crystals. Namely, it is responsible for higher energetic characteristics of laser based on Ce^{3+} :LLF crystals. Strong and wide pump-induced absorption band centered at 310 nm is observed in Ce^{3+} :YLF. This band is shifted to blue and its intensity goes down with Lu content. We have evaluated free charges recombination rate, excited state absorption cross-section for Ce^{3+} ions and some other photodynamic processes related microparameters. Fitting results indicate that pump-induced color centers lifetime decreases with Lu-content in $\text{LiYF}_4\text{–LiLuF}_4$ mixture and it can be associated with more efficient color center bleaching by Ce^{3+} ions 5d–4f fluorescence.

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1. Introduction

UV and VUV quantum electronic devices, based on 5d–4f transitions of trivalent rare-earth ions in wide band gap dielectric crystals, offer some important advantages against the traditionally used ones. However, most media of this type pertinent for UV applications experience problems related to photodynamical processes resulting mainly in solarization effect, which comes after active ion multiphoton stepwise ionization by intense UV pumping [1]. Impurity ions photoionization not only withdraws energy from inversed population in the active media, but pump-induced color center absorption contributes to the total losses and increases laser oscillation threshold leading to ineffective and transient laser action [2,3]. Now, it is obvious that activator ions and host lattice interaction is

the key factor in photochemical conversions under intense UV pumping and, therefore, in laser performances.

Host lattice structure and cationic set determine dopant's energy levels, structure and positions relatively to host bands and therefore host cation change impact on both spectral-kinetic and photochemical properties of activated crystals. It is most appreciable for $4f^n\text{–}4f^{n-1}5d$ interconfigurational transitions of rare-earth ions since 5d-electronic orbitals are expanded far away from the core and thus very sensitive to ligand environment [4]. It is very well demonstrated by numerous studies of optical properties of crystal families doped by Ce^{3+} ions (see e.g. $\text{CaF}_2\text{–SrF}_2\text{–BaF}_2$ [5], $\text{LiYF}_4\text{–LiLuF}_4$ [6]). Thus, cationic set modification seems to be a great opportunity to manage active media basic optical and photochemical properties and to achieve a success in realization of an effective UV laser action based on $4f^{n-1}5d\text{–}4f^n$ interconfigurational transitions. This can be achieved due to two reasons. First, by choosing appropriate host cationic set we can obtain crystal with wider band gap and therefore reduce an

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activator ion stepwise two-photon ionization probability. Second, host cationic set modification induces changes in impurity energy levels position relatively to the host band structure. For instance, in case of close magnitude of ionization potential of substituted and dopant ions, the impurity ground state have to be situated close to the host valence band. Therefore, activator-ion ionization energy in that host will be larger. Switching from Ce:LiSrAlF₆ to Ce:LiCaAlF₆ crystals, vanishing solarization effect [7,8], and switching from Ce:LiYF₄ (YLF) to Ce:LiLuF₄ (LLF) crystals, drastically improving photochemical stability and laser performances, are good illustrations of this approach [6].

2. Background: comparison of spectral, photochemical and laser properties of Ce³⁺-doped LiYF₄ and LiLuF₄ host lattices

Everything said above can be implemented to LiYF₄ and LiLuF₄ homologous crystals of scheelite structure family. These crystal hosts doped with Ce³⁺ ions are well-known UV active media and both show detrimental solarization effect under intense UV pumping. However, Ce:LLF active medium displays better photochemical stability under pumping conditions and higher laser characteristics against Ce:YLF [6] (Fig. 1). Besides, it is known that the most prospective way to reduce the solarization effect is to engage processes of free charges recombination for example by codoping with appropriately chosen ions [9,10]. For example, Ce:LLF single crystal's codoping by Yb³⁺ ions furthers antisolarant effect and gives the record value of laser slope efficiency (Fig. 1b). As a matter of fact, Yb³⁺ ions have great value of electron affiliation that is responsible for effective free electrons trapping [11].

The differences between the two host lattices start with the red shift of luminescence and absorption bands of Ce³⁺ interconfigurational transitions spectra in LLF in comparison to YLF crystals. Although it is not big ($\sim 340\text{ cm}^{-1}$), it is enough to provide better fit of absorption spectrum with laser wavelength of the most popular excimer lasers, which can be used for pumping. Rise of absorption coefficient at

248 nm (KrF-laser) allows lowering Ce³⁺ ion content and thus reducing the microdefect amount in the crystals, which are responsible for solarization intensity. Besides, due to the red shift of the Ce³⁺ ions 4f–5d absorption band, pumping with XeCl laser emission at 308 nm becomes available. This can also be a step towards reducing impurity ions multiphoton ionization probability and avoiding degradation of optical properties of active medium during laser action.

The second, preliminary investigations indicate that Ce³⁺ ions energy states are localized closer to the top of valence band in LLF compared with YLF crystals. The gap between Ce³⁺ ions ground state and top of valence band is 3.3 eV for YLF and 2 eV for LLF crystal hosts [12]. Thus, Ce³⁺ ions photoionization threshold has to be shifted to the shorter wavelengths in LLF in comparison to YLF.

The most essential distinction between these active media is twice higher luminescence quantum yield of Ce³⁺ emission in LLF against Ce³⁺:YLF. This difference stayed unclear till recently because YLF and LLF have identical structures and similar optical properties and phonon spectra.

Concerning Yb-coactivation, in high-doped samples with more than 50% substitution of host cations in Ce:LLF or Ce:YLF crystals by Yb³⁺ ions Ce³⁺ 5d–4f luminescence is totally quenched [2]. Whereas lean Yb³⁺ ions codoping does not change dramatically spectral properties of Ce³⁺ ions, but effectively suppress UV pump-induced solarization [9,13].

Following Ref. [10], basic spectral-kinetic characteristics, photochemical and gain properties of series of LiF–YF₃–LuF₃ mixture crystals doped with Ce³⁺ and Yb³⁺ ions were systematically investigated in order to evaluate the Lu³⁺ and Yb³⁺ codoping effect.

3. Luminescence decay investigation

Competition between processes of free charges trap by host defect sites and impurity ions plays a key role in luminescence properties and photochemical stability of Ce-doped crystals. The mechanism of this pump-induced

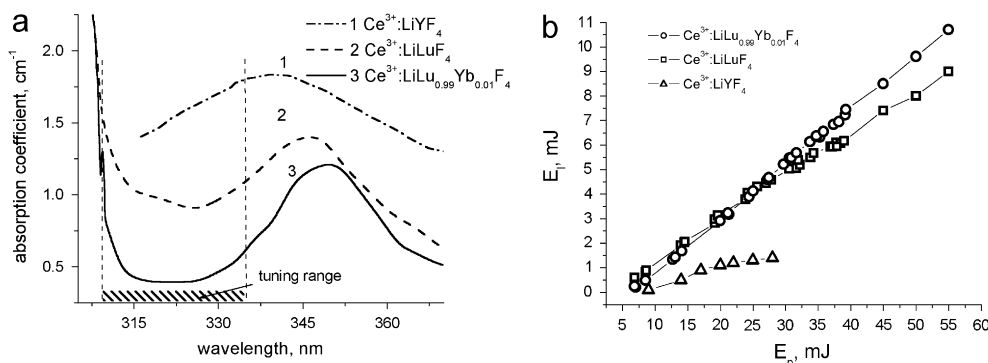


Fig. 1. (a) Absorption spectra of KrF laser pumping-induced color centers and (b) output laser energy at 311 nm vs. pumping energy for Ce³⁺ ions doped LiYF₄, LiLuF₄ and LiY_{0.95}Yb_{0.05}F₄ crystals.

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