



Spectroscopic properties of UV active medium $\text{Ce}^{3+}:\text{LiSr}_{0.8}\text{Ca}_{0.2}\text{AlF}_6$



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ABSTRACT

The aim of this work is phase composition and near UV spectroscopic studies of UV active media in fluoride crystals with colquiriite structure, such as $\text{Ce}^{3+}:\text{LiSr}_{0.8}\text{Ca}_{0.2}\text{AlF}_6$. Colquiriite structure mixed crystals show higher segregation coefficient of Ce^{3+} activator ions than common LiCaAlF_6 hosts. An important result is based on the fact that this enhancement was achieved for two types of Ce^{3+} centers in a multisite $\text{Ce}:\text{LiSr}_{0.8}\text{Ca}_{0.2}\text{AlF}_6$ system. Thus, it provides a higher gain coefficient for the 5d–4f transitions of Ce^{3+} ions and it spans a wider continuous wavelength tuning range between 280 and 320 nm for tunable $\text{Ce}:\text{LiSr}_{0.8}\text{Ca}_{0.2}\text{AlF}_6$ laser systems.

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1. Introduction

Since the discovery of direct UV laser oscillation from Ce^{3+} doped LiCaAlF_6 (LICAF) crystals [1], the gain medium has been investigated for various applications [2–4]. Significant vibrational broadening of the 5d–4f transitions of Ce^{3+} ions for extended laser tunability was used in LIDAR systems for remote atmospheric control [5]. Higher cross-sections of dipole interconfigurational transitions provide a lower threshold for laser oscillation. Coupled to the appropriate pumping source this system exhibited tunability from 281 to 316 nm [5], with more than 60 mJ output energy from the oscillator [6] at 290 nm and high repetition rate, up to 20 kHz [7] with fast and robust operation. Up to now, microchip UV lasers were investigated [8] and lasing was achieved with 24% slope efficiency from a 2.2 mm long $\text{Ce}:\text{Na}:\text{LICAF}$ crystal with a Ce ion dopant concentration of 3.5% in the melt, and 2 μJ pumping energy at 266 nm from a microchip $\text{Nd}:\text{YVO}_4$ laser device. Interesting results have been achieved too in the field of short pulses. UV laser pulses of 22 ps at FWHM were obtained [9] by using synchronous pumping with 70 ps laser pulses. Also, laser pulse trains have been achieved with 800 ps at FWHM for a single pulse, from a low Q cavity [10]. Generation of short laser pulses is not a trivial task as mode-locking or Q-switching techniques must be employed and control of optical cavity's Q-factor was obtained by modulating optical cavity transients [11,12].

Any further progress of Ce-doped colquiriite lasers is associated with improvement spectroscopic and photochemical properties of

the crystals and increment of dopant's concentration. It is widely accepted that Ce^{3+} activators in $\text{Ce}:\text{LICAF}$ matrixes are able to substitute sites occupied by Ca^{2+} , which has an ionic radius similar to that of Ce^{3+} (1 Å and 1.03 Å, respectively) [2]. However, it is difficult to increase the Ce^{3+} ion concentration in $\text{Ce}:\text{LICAF}$ crystals because the valences of Ce^{3+} and Ca^{2+} ions are different. An increment of dopant's concentration has the consequence of enhancing crystal lattice defect generation, as the crystal struggles to preserve its electric neutrality. Consequently, the segregation coefficient of Ce^{3+} ions in $\text{Ce}:\text{LICAF}$ crystals is quite small [13,14]. Moreover, Ce^{3+} ions doping produces “seed centers” for color center generation. It is known that by varying the chemical structure of the compound, by adding a set of crystal lattice cations in the matrix, one can improve the optical quality of the crystals by increasing the isomorphic capacity of the solid solution [15].

By growing mixed $\text{LiSr}_{1-x}\text{Ca}_x\text{AlF}_6$ crystals with varying x, is possible to optimize the chemical content for improving laser properties. In earlier works [16,17], a series of $\text{LiSr}_{1-x}\text{Ca}_x\text{AlF}_6$ crystals activated with Ce^{3+} ions exhibited a higher absorption coefficient for Ce^{3+} ions, which could be an advantage for activator ions concentration optimization with subsequent laser action [17,18]. Furthermore, it should be noted that multicenter Ce^{3+} segregation in colquiriite crystals plays a significant role for laser action while the distribution of Ce^{3+} ions concentration among different types of impurity sites in mixed crystals is unclear. Indeed, the ionic distribution depends not only on the Ca/Sr ratio but on the crystal growth conditions also and crystals grown by the Czochralski method have a lower segregation coefficient of Ce^{3+} ions in comparison to the Bridgman growth technique.

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