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Spectral-kinetic studies of $SrAlF_5$ doped by trivalent rare-earth ions

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ABSTRACT

Photochemical properties of Ce^{3+}:$SrAlF_5$ and Ce^{3+},Yb^{3+}:$SrAlF_5$ single crystals together with spectroscopic and kinetic characteristics of several optically nonequivalent impurity centers and energy transfer between them are described. It is shown that co-activation by Yb^{3+} ions effectively suppresses color centers in Ce,Yb:SAF crystals. It was found out that in Ce,Yb:SAF crystals Yb ions exist simultaneously in 2+ and 3+ valent state. Three types of optically nonequivalent luminescent centers corresponding to the doublets in luminescence spectrum centered at 290, 305 and 370 nm (Ce^{I}, Ce^{II}, Ce^{III}, respectively) have been observed. Analysis of luminescence spectra and decays leads to the conclusion that there is no energy transfer between either cerium centers or from Ce^{3+} to Yb^{2+} apart from the Ce^{III} center which luminescence is slightly quenched by Yb^{2+}.

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1. Introduction

Searching for and investigating new perspective active media for UV tunable solid state lasers is an urgent task, because modern technology requires as short wavelength of laser emission as possible [1]. $SrAlF_5$ (SAF) crystals doped by rare-earth (RE) ions, especially Ce^{3+} [2], is a very promising laser material for the UV/VUV spectral ranges due to broad band-gap (12.3 eV [3,4]) and chemical composition similar to the most effective UV laser active media $LiCaAlF_5$ and $LiSrAlF_5$. SAF crystals doped by iron-group ions are known as rather effective IR laser material [7]. It could also be doped by of RE^{3+} ions and despite their rather low segregation coefficient it is still enough for practical applications in quantum electronics devices based on interconfigurational transitions of active ions.

Because of Goldschmidt's rule trivalent RE^{3+} ions dominantly substitute for divalent strontium lattice cations which in this crystal exist in four nonequivalent positions [8]. Besides charge compensation is required. Thus several optically nonequivalent RE^{3+} centers formation are expected. Presence of the few types of optically nonequivalent active centers in one medium especially if the energy exchange between them is ineffective will inevitably lead to the dissipation of excitation energy and puts into question the use of this material for laser applications [9].

In literature there are a lot of contradictory assignments of the $SrAlF_5$ crystalline space group [5,6]. According to the most common viewpoint this crystal belongs to the noncentrosymmetric space group $I4$ and there are a lot of studies in favor of ferroelectric effect existence in this material [5]. In such case active medium with controllable spectral properties is the one of interest. But on the other hand more recent theoretical and experimental studies report centrosymmetric space group $I4_1/a$ for $SrAlF_5$ which eliminates any ferroelectric behavior [6]. Therefore optical and spectroscopic properties studies of this crystal still remain an urgent task.

Another problem in the way of the search for the new materials for UV laser applications is that the most fluoride crystal hosts are subjected to the strong coloration under pumping condition (for example, [10]).To level this effect coactivation with Yb^{3+} ions has been successfully applied before [10]. The disadvantage of such approach are that: (a) doping by RE^{3+} ions generally leads to the formation of crystal lattice defects serving as the nuclei for color centers formation; and (b) Ce^{3+} ions 5d–4f luminescence could be significantly quenched by ytterbium ions, thus optimization of activator ions concentrations should be performed.

This paper reports on spectral, kinetic and photochemical properties of Ce:SAF and Ce,Yb:SAF single crystals of several optically nonequivalent RE impurity centers and energy transfer between them.

2. Experimental details

The tetragonal Ce:SAF and Ce,Yb:SAF crystals with doping concentration ~0.5 at% in the melt were grown in Ar-atmosphere in carbon crucibles using Bridgman–Stockbarger technique. The melt contained a slight excess of AlF_3 above the stoichiometric composition. High-purity chemicals (>99.99%) of commercially available SrF_2, AlF_3, CeF_3, and YbF_3 powders were

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utilized. Polished disk-shaped samples of 1.2 mm thickness and 7 mm in diameter with optical axis in disk plane were prepared.

Absorption and luminescence spectra were registered by CCD spectrometer Stellarnet EPP2000-HR with detector integration time more than 1 ms and deuterium and halogen lamp as light sources. Luminescence spectra were studied using site-selective laser spectroscopy technique. As a pumping source the 4th harmonic radiation of YAG:Nd laser (266 nm) and its stimulated Raman scattering component emission (217, 238, 300 nm) from H_2 Raman-cell were used. Laser radiation pulse duration did not exceed 7 ns. These wavelength conform well to the 4f–5d absorption bands of Ce^{3+} ions in SAF crystals and absorption band belonging to Yb^{2+} ions in Yb:SAF [8]. Luminescence decays were registered by photomultiplier tube FEU-87 with a rise time about 6 ns and digital oscilloscope Aktakom AOC-2282.

3. Results and discussion

Polarized absorption spectra of both Ce:SAF and Ce,Yb:SAF crystals registered at 77 K are shown on Fig. 1. Generally absorption spectra of Ce:SAF contain several broad bands localized at about 232, 244, 249, 265, 307 nm, but due to several nonequivalent Ce^{3+} centers formation it's impossible to assign these bands to specific transitions. One observes though that in the view of obtaining laser action Ce:SAF and Ce,Yb:SAF are suitable for such pumping sources as Nd:YAG (ω_4=266 nm) and KrF (ω=248 nm) as indicated on Fig. 1b.

Fig. 2 shows transmission of Ce:SAF and Ce,Yb:SAF crystals during irradiation of the samples by high-energy (~0.8 J/cm^2) emission of Nd:YAG laser's 4th harmonic (266 nm). It demonstrates effective suppression of color centers absorption in Ce:SAF

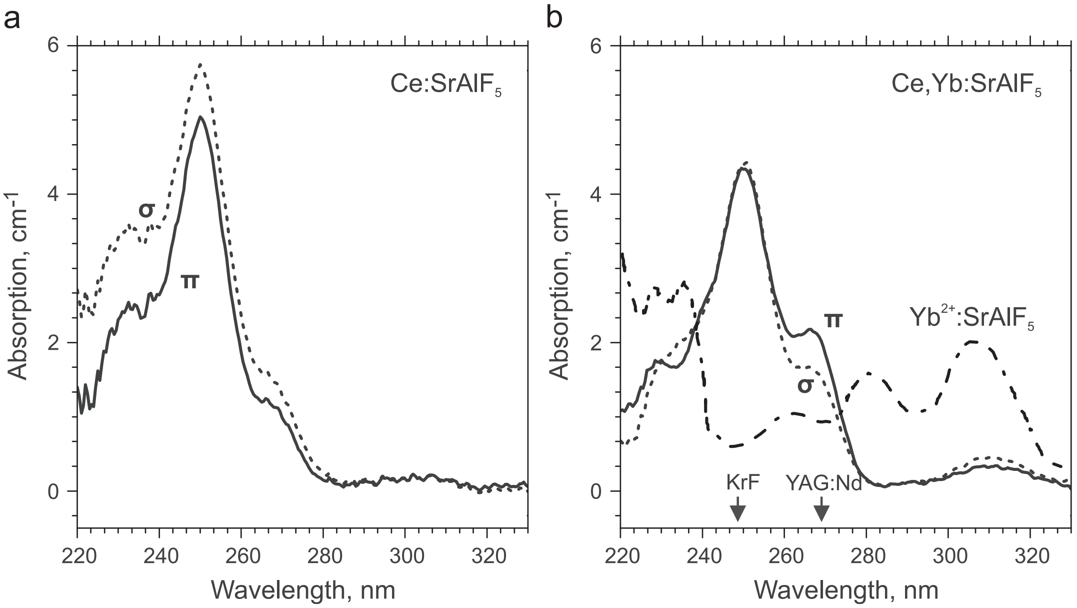

Fig. 1. Polarized absorption spectra of Ce^{3+}:$SrAlF_5$ (a) and Ce^{3+},Yb^{3+}:$SrAlF_5$ (b); π-polarized absorption spectrum (solid) and σ-polarized absorption spectrum (dashed). Non-polarized absorption spectrum of Yb^{2+}:$SrAlF_5$ crystal (dot-dashed curve) is given from [8]. The arrows show suitable pumping source wavelengths: YAG:Nd (λ_4=266 nm) and KrF (λ=248 nm).

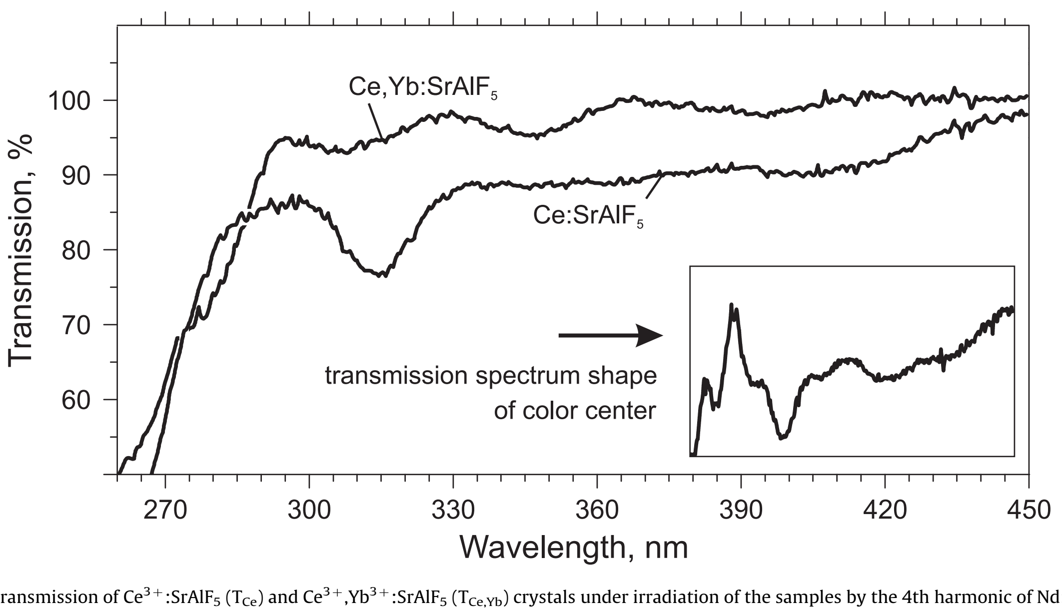

Fig. 2. π-polarized transmission of Ce^{3+}:$SrAlF_5$ (T_{Ce}) and Ce^{3+},Yb^{3+}:$SrAlF_5$ ($T_{Ce,Yb}$) crystals under irradiation of the samples by the 4th harmonic of Nd:YAG laser emission (266 nm) with a flux~0.8 J/cm^2 at the room temperature. (Inset) suggested transmission (in the same scale) spectrum shape of color centers suppressed due to Yb^{3+} ions co-doping is shown (taken as the ratio T_{Ce}/ $T_{Ce,Yb}$).

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