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A new technique of the excited-state photoionization studies in $Ce:LiYF_4$ and $Ce:LiLuF_4$ crystals

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ABSTRACT

The new method of photodynamic processes investigations in UV active media based on the analysis of the activator ions fluorescence decay is proposed. The excited-state photoionization spectra from 5d-state of Ce^{3+} ions in $LiYF_4$ and $LiLuF_4$ are determined. Photoelectrons and holes recombination and trapping cross-sections are estimated. The terminal state of excited-state photoionization is attributed to the 6s configuration of Ce^{3+}, located in the conduction band of the host material.

1. Introduction

Search for the solid-state active media for UV and VUV lasers has shown that the most technically simple method of obtaining tunable UV laser action is to use parity allowed $4f^{n-1}5d$–$4f^n$ (5d–4f) interconfigurational transitions of trivalent cerium ions, doped into wideband dielectric crystals [1,2]. However, the majority of solid-state active media experience degradation of optical and laser properties under UV excitation due to the various photodynamic processes (PDP) [3]. The origin of these processes is excited state absorption (ESA) of pumping radiation, accompanied by the ionization of activator ions (excited-state photoionization—ESPI [4]), and color centers formation. Study of characteristics of such intense photodynamic processes in activated crystals is an urgent task, since it allows to determine the necessary conditions for eliminating the harmful influence of PDP and, eventually, to create effective solid-state active media of UV and VUV spectral ranges.

An important parameter, characterizing the effectiveness of PDP, is the ESPI cross-section at pumping wavelength. Knowing the ESPI spectrum it becomes possible to determine the positions of excited states of impurity centers relative to the bands of the host, which allows to select suitable excitation conditions for Ce^{3+}-doped crystals to avoid PDP negative effects. Currently several experimental approaches are used to investigate the ESA. Standard practice is to use the pump-probe technique [5]. Photoconductivity measurements are also proved to allow performing the direct studies of ESPI, because they are based on the registration of free charge carriers' appearance in the energy bands of the host material [6,7]. However, registration of the ESPI cross-section spectral distribution may be complicated by the overlapping of several processes: ground and excited-state absorption of the impurity ions and fundamental absorption of the crystalline matrix. Therefore, it is not always possible to obtain ESPI spectra directly from either kind of these investigations, which leads to the necessity to resort to indirect measurements followed by thorough analysis of the collected data.

Pump-induced PDP mentioned above will inevitably influence the decay of impurity centers fluorescence. Earlier we have proved that in $Ce:LiLuF_4$ and $Ce:LiYF_4$ crystals decays recombination component appears for the longer time, when excited by radiation resonant to 4f–5d transitions of Ce^{3+} ions [8]. Therefore, an alternative approach to establish ESPI spectra becomes possible, because the spectral dependence of the weight of recombination component in the impurity ion fluorescence decay is determined by the ESPI cross-section, and, therefore, depends on the excitation wavelength. The simplicity and effectiveness of this method makes it a useful instrument in comprehensive studies of activated materials for various applications in optical spectral range.

2. Materials

In this work $LiLuF_4$ and $LiYF_4$ fluoride crystals doped with Ce^{3+} ions have been studied. The crystals were grown by means of the Bridgman–Stockbarger technique in graphite crucibles. The concentrations of impurity ions were 0.5 mol% in the melt. All crystalline samples were cut and polished to a cylindrical shape of 5 mm diameter and 2 mm thickness.

$Ce:LiLuF_4$ and $Ce:LiYF_4$ crystals of scheelite-type structures are well-known active media of the UV spectral range [9,10]. However, there are different types of photodynamic processes arising in these media under UV excitation [11].

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