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Electronic excitation energy transfer processes in Er:YAG under variable pump duration

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ABSTRACT

For the first time, the luminescence decay dependences of $^4I_{13/2}$ and $^4I_{11/2}$ energy levels on pulsed excitation duration are studied with and without pinhole technique in erbium-doped YAG single crystals. Luminescence decay investigations of $^4I_{13/2} \rightarrow {}^4I_{15/2}$ (1565 nm) and $^4I_{11/2} \rightarrow {}^4I_{15/2}$ (970 nm) transitions are carried out on 5 at. % and 40 at.% doped samples. Time-resolved measurements of luminescence decay have demonstrated non-exponential behavior depending on excitation duration as well as impurity concentration, thus, indicating the presence of various energy transfer processes. In order to eliminate the influence of radiation trapping effect on luminescence decay from $^4I_{13/2}$ and $^4I_{11/2}$ levels the pinhole technique is used. Qualitative comparisons of luminescence decay curves are made to estimate the influence of radiation trapping effect. The influence of non-radiative energy transfer processes on population redistribution of $^4I_{13/2}$ and $^4I_{11/2}$ energy levels is evaluated. A theoretical model for non-radiative transfer of electronic excitation energy in YAG:Er^{3+} crystals is proposed, in the frame of which the constants of rate equations are determined. The experimental and theoretical investigations are made for luminescence corresponding to both $^4I_{13/2} \rightarrow {}^4I_{15/2}$ and $^4I_{11/2} \rightarrow {}^4I_{15/2}$ transitions. The following non-radiative energy transfer processes are taken into account: $^4I_{13/2} \rightarrow {}^4I_{15/2} + {}^4I_{13/2} \rightarrow {}^4I_{9/2}$, $^4I_{11/2} \rightarrow {}^4I_{15/2} + {}^4I_{11/2} \rightarrow {}^4F_{7/2}$, $^4S_{3/2}(^2H_{11/2}) \rightarrow {}^4I_{9/2} + {}^4I_{15/2} \rightarrow {}^4I_{13/2}$, $^4I_{11/2} \rightarrow {}^4I_{15/2} + {}^4I_{13/2} \rightarrow {}^4F_{9/2}$.

1. Introduction

Recently, there has been an increased interest on erbium-doped yttrium aluminum garnet (YAG:Er^{3+}) lasers operating at wavelengths near 1.5 μm and 3 μm due to their wide applications in optical communications systems, medicine, remote sensing and ranging, etc. Understanding the mechanisms of energy transfer processes in impurity subsystem of YAG is extremely important for further laser pumping improvement and effective generation of coherent radiation in near- and mid-infrared regions.

Various radiative and non-radiative energy transfer processes (ETP) occurring in the impurity subsystem affect the luminescence properties of crystal such as the luminescence decay law, quantum yield, etc. In particular, the resonant radiative ETP leads to the phenomenon of radiation trapping (RT) and, as a result, to the lengthening of luminescence lifetime compared to true lifetime of corresponding energy level [1–6]. The RT phenomenon in Er^{3+} impurity subsystem is investigated in [4–6], where the ways minimizing the influence of RT by implementation of special excitation and detection schemes are suggested.

This can be achieved by using a pinhole technique [1,2,6] which can suppress the luminescence from sample areas excited by the radiative resonant transfer of laser pumping. On the other hand, the presence of non-radiative ETPs in crystals can change the luminescence decay time in both directions. Processes of non-radiative ETPs in Er^{3+} doped crystals are investigated in [7–10] where various mechanisms of ETPs are proposed to describe the non-radiative redistribution of excitation energy between Er^{3+} ion energy levels, and corresponding energy transfer coefficients are determined.

The theoretical basis for investigation of non-radiative and radiative ETPs is developed in [11–22]. In particular, in [12–18], the radiative ETPs are discussed where the authors mention two different ways of theoretical description of these processes based on the Holstein-Biberman integral-differential equations [12–15] and the Milne diffusion equation [16–18]. In [11,19–22], the probabilities of elementary acts of non-radiative ETPs induced by a variety of mechanisms and interactions of impurity ions with each other are calculated, as well as the methods of summing the ETP elementary act probabilities over the donor and acceptor subsystems are discussed. In [23–25], investigations of non-radiative ETPs in YAG:Er^{3+} are carried out by taking into account the following channels of excitation energy transfer: $^4I_{13/2} \rightarrow$

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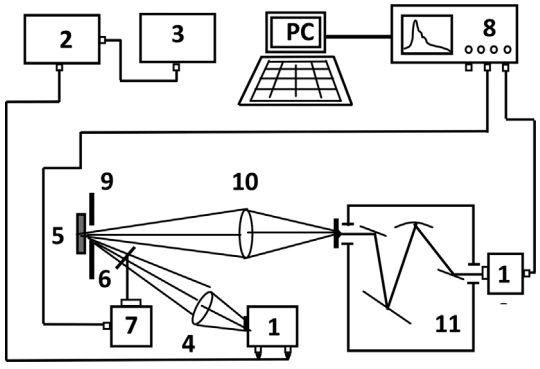

Fig. 1. The experimental setup.

$^4I_{15/2} + {}^4I_{13/2} \rightarrow {}^4I_{9/2}$, $^4I_{11/2} \rightarrow {}^4I_{15/2} + {}^4I_{11/2} \rightarrow {}^4F_{7/2}$ and $^4S_{3/2} \rightarrow {}^4I_{9/2} + {}^4I_{15/2} \rightarrow {}^4I_{13/2}$. Although the excitation pulse intensity is changed in [24,25], the excitation duration is kept constant.

In this work, the ETPs for different concentrations of impurity ions in YAG:Er^{3+} crystals are investigated with the purpose of estimation of ETP influence on population dynamics of $^4I_{13/2}$ and $^4I_{11/2}$ levels, determination of constants of non-radiative energy transfer, and evaluation of the RT phenomenon influence on luminescence characteristics under consideration. Wherein, the luminescence from both $^4I_{13/2}$ and $^4I_{11/2}$ levels is considered to increase the calculation accuracy.

2. Experimental

Time-resolved spectroscopic investigations of luminescence decay on the $^4I_{13/2} \rightarrow {}^4I_{15/2}$ and $^4I_{11/2} \rightarrow {}^4I_{15/2}$ transitions were carried out on two plane-parallel, optically polished samples made of bulk YAG:Er^{3+} crystals with 5 at.% and 40 at.% impurity ion concentrations. The experimental setup used for luminescence measurements is schematically shown in Fig. 1. 1 W modulated laser diode (1) operating at 442 nm was used as an excitation source. It was powered by Thorlabs LDC 210 laser diode current controller (2) providing adjustable output current. The excitation pulse durations are modulated via generator (3) which allows changing pumping durations in the wide 20–10000 μs range with acceptable modulation properties, i.e. pulse rise and fall time, modulation depth. Laser excitation was directed to the samples (5) by focusing lens (4). The thin glass plate (6) was used to reflect a small part of the excitation to the photodetector (7) for monitoring the excitation pulse as well as triggering the data acquisition process (8). In order to estimate the influence of RT on the luminescence decay kinetics, measurements with pinhole technique, as well as without, were carried out.

For this purpose, a thin pinhole was mounted directly on the crystal surface (9). The sample excitation and luminescence detection were carried out through the same crystal side as shown in Fig. 2. It is shown by many authors, including our team [1–3,6], that increase in the

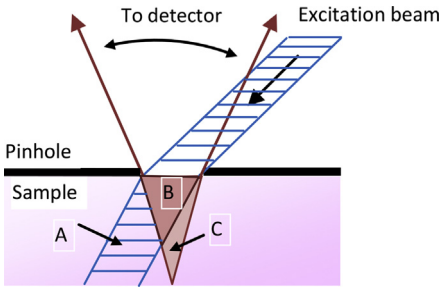

Fig. 2. Excitation and detection scheme of YAG:Er^{3+} crystals: A –crystal region directly excited by laser; B – luminescence from directly irradiated region; C – luminescence from region excited via radiative transfer of excitation.

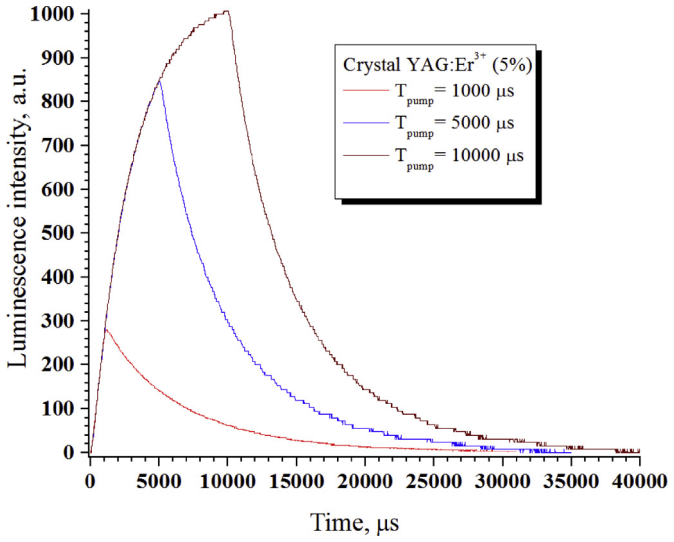

Fig. 3. 1565 nm luminescence of YAG:Er^{3+} 5% crystal registered without pinhole under different pumping durations.

registered decay time of impurity ion luminescence due to RT phenomenon depends on the ratio of luminescence portion detected from region directly excited by laser (region B in Fig. 2) to that from region excited via radiative transfer of excitation (region C in Fig. 2). This approach let us practically eliminate the RT influence on the registered luminescence signals.

During the experiment pinholes with diameters from 200 to 1000 μm are used. The sample's luminescence was collected via collimating lens (10) and directed to the monochromator MDR-3 (11). Output luminescence signals were detected using photomultiplier tube and germanium photodiode (12). The time-resolved registration of luminescence was carried out at 970 nm and 1565 nm wavelengths near the peaks of the samples' luminescence bands. These radiations correspond to the transitions from $^4I_{11/2}$ and $^4I_{13/2}$ excited states to $^4I_{15/2}$ ground state of the Er^{3+} ion, respectively.

As an example, the initial registered view of 1565 nm luminescence for YAG:Er^{3+} 5% crystal under several pumping durations is presented in Fig. 3.

Hereinafter, only decay parts of luminescence curves are used, so that the beginning of time (t = 0) is coupled with starting point of luminescence decay. Luminescence decays at both 1565 nm and 970 nm wavelengths under 100–10000 μs pumping durations are registered with pinholes technique as well as without. In Fig. 4, the 1565 nm luminescence decay of YAG:Er^{3+}(40%) under 100 μs excitation is shown for both measurement methods. Each curve is normalized by its maximum value.

It should also be noted, that measurements made by using pinhole result in shorter average luminescence lifetime at 1565 nm compared to that without pinhole which is explained by the RT impact on lifetime measurements for a given concentration.

In Fig. 5 (a, b), dependences of 1565 nm and 970 nm luminescence decay curves on excitation duration registered with 200 μm pinhole are shown for 5 at.% doped YAG:Er^{3+} sample. Processing of registered signals, especially, at 970 nm wavelength, showed that above certain excitation duration ($\tau_{pump} > 500$ μs) the luminescence decay curves cannot be approximated by a single exponential function. It can be seen that increasing the excitation duration makes the luminescence decay at 1565 nm faster and, in contrast, slows down it at 970 nm.

The luminescence decay curves at 1565 nm and 970 nm registered with 200 μm pinhole for 40 at.% doped YAG: Er^{3+} sample are shown in Fig. 5 (c, d). In this case, none of the decay curves, especially at 1565 nm wavelength, can be approximated by a single exponential function for the whole range of excitation durations (Fig. 5 (c)). Note,