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The experimental evidence of the amplified spontaneous emission of Yb^{3+} ions in $LiYbF_4$ crystal

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ABSTRACT

The experimental evidence of the amplified spontaneous emission of Yb^{3+} ions in $LiYbF_4$ crystal, which partially stipulates up-conversion processes in Yb-sensitized phosphors, doped by rare-earth ions are presented for the first time. To do that the spatial distributions and the spectra of Yb^{3+} ions luminescence along the excitation radiation propagation through the sample were studied simultaneously. The laser diode radiation (1 W, $\lambda=932$ nm) was used for the luminescence excitation and its surface power density was varied by shifting of the position of the laser beam waist within the sample (similar to Z-scanning).

1. Introduction

The upconversion (UC) luminescence of the rare-earth ions doped crystals is typically caused by a combination of the numerous multiphoton and cooperative processes: a) cross-relaxation, b) excited-state absorption of excitation and/or fluorescence radiation, c) energy transfer and energy migration etc [1–3].

The co-doping of the crystalline materials by Yb^{3+} ions with appropriated optical excitation allows enhancing UC efficiency and even realizes the effective UC pumped lasing [4–8]. Yb^{3+} ions sensitized rare-earth ions doped nanoparticles also demonstrate high effective UC luminescence and it is widely used for many various applications [9–12]. Usually the role of Yb^{3+} ions in the UC processes is considered to be sensitizer only.

To study UC luminescence, the spectral and kinetic luminescence characteristics versus the pump radiation power are traditionally measured at stable focusing of the laser beam on a surface of a sample. In this case, the spatial distribution of the excitation and luminescence power inside the sample is ignored. The recent comprehensive studies demonstrate the necessity of taking into account the spatial distribution of excitation radiation and UC emission inside the sample and assume that there is a hidden amplified spontaneous emission (ASE) within Yb^{3+} ions assemble. The hidden ASE plays a dominant role in the excitation of the up-conversion luminescence of Tm^{3+} ions in Yb^{3+}-doped YF_3 [13,14] and Ho^{3+} ions in $LiYbF_4$ single crystals [15].

Here we present the results of study of spatial distributions and the spectra of Yb^{3+} ions luminescence along the excitation radiation propagation through the sample using Z-scan technique [16,17] and demonstrate the experimental evidence of the amplified spontaneous emission due to $^2F_{5/2}$-$^2F_{7/2}$ transitions of Yb^{3+} ions in $LiYbF_4$ crystal.

2. Material and methods

A $LiYbF_4$ crystal grown using the Bridgman-Stockbarger technique in Kazan Federal University was cut in the form of a parallelepiped with dimensions $\sim 3\times4\times10$ mm. The c-axis of the crystal sample was oriented along the shortest side and it was perpendicular to the z-axis of the experimental setup (Fig. 1). The Yb^{3+} ions luminescence was excited by the radiation of a semiconductor laser diode (LD) with a maximal power of 1 W at $\lambda=932$ nm. All the experiments were performed at a temperature of 300 K. The luminescence spectra were recorded with StellarNet spectrometer (spectral resolution 0.5 nm). To avoid an overloading of the spectrometer with intensive excitation radiation, the luminescence spectra were taken perpendicularly to the laser beam. The longitudinal cross sections of the luminescence were photographed with a ToupCam VCMOS14000KPA MT9F002 camera, attached to measuring microscope (magnification 25/50). The luminescence of Yb^{3+} ions was separated with an infrared filter (cutoff wavelength $\lambda>0.95$ μm).

The spatial distributions and the spectra of Yb^{3+} ions luminescence along the excitation radiation propagation through $LiYbF_4$ sample were studied simultaneously using the Z-scan technique. The standard Z-scan technique is based on the registration of intensity of focused laser beam passing through the

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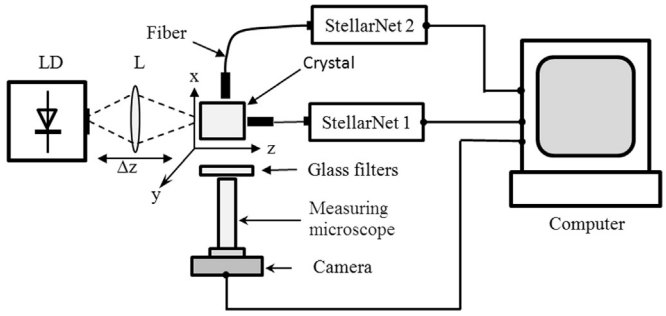

Fig. 1. The experimental setup. LD — the laser diode (1 W at λ=933 nm); StellarNet — fiber coupled CCD spectrometer (spectral resolution better than 0.5 mm) attached to the computer; L — the lens (f=50 mm); Camera — ToupCam VCMOS14000KPA MT9F002; Crystal — the $LiYbF_4$ single crystal.

sample versus the position (Δz) of the beam waist within the sample [16,17]. The laser beam waist was shifted with a movable convex lens (focal length 5 cm) along the axis z relative to the front surface of the sample. We took the photographs of the longitudinal cross-sections of Yb^{3+} luminescence and its spectra simultaneously at the various Δz. The values $\Delta z < 0$ correspond to the laser beam waist position ahead of the crystal, $\Delta z = 0$ means that the beam waist is situated just exactly at the front surface of the sample, and $\Delta z > 0$ corresponds to the beam waist situated inside the crystal. The diameter of the laser beam waist on the front surface of the crystal varied within $1 \div 0.1$ mm range, which provides the surface excitation power density within the range of $\rho \approx 0.1 \div 10$ kW/cm^2.

3. Results

The typical Yb^{3+} ions non-polarized luminescence spectrum (a), its intensities at 1030 nm and the laser beam surface power density dependencies versus the position of the laser beam waist (b) are presented in Fig. 2.

The luminescence spectra are strongly distorted because of the reabsorption. Nevertheless, the red tail of luminescence spectra situated around 1030 nm reflects the relative population of excited manifold $^2F_{5/2}$ of Yb^{3+} ions, which must be proportional to the excitation power density. The surface power density ρ on the front surface of the sample depended on the laser beam waist position Δz and reached the maximum value at Δz=0 mm (Fig. 2b). At the same time, the Yb^{3+} ions luminescence intensity in $LiYbF_4$ crystal did not follow ρ and it even decreased when the density ρ increased. Moreover, it reached the minimum value at Δz=0.4 mm and its position partially depended on the laser beam power (Fig. 2b). It looked *abnormal*. Taking into account that the spectrometer registers the luminescence from the whole sample, absorption saturation of excitation radiation could not explain this one. However, on the other hand, it could be easily made clear if the impact of the ASE of Yb^{3+} ions, which "ate up" the population of $^2F_{5/2}$ excited manifold was considered. In this case the Δz=0.4 mm corresponded to the critical length of the sample with the maximum of the ASE effect. Whereas the ASE had directionality along z-axis, the measured luminescence intensity across that direction dropped at Δz=0.4 mm and then it slowly grew due to ASE reabsorption and re-excitation of Yb^{3+} ions situated into the sample at $\Delta z > 1.0$–1.2 mm.

The direct observation of the Yb^{3+} ions ASE along the laser beam was impossible because of overloading of the spectrometer with intensive excitation radiation, but we obtained additional evidence from analyzes of longitudinal cross-section photographs of the Yb^{3+} ions luminescence in the sample. The horizontal Z (on to the z-axis) and vertical Y (on to the y-axis) profiles were calculated for the different values of Δz (Fig. 3). The widths of these profiles were estimated by the technique described in [7].

As the laser beam waist penetrated deep into the sample at about Δz=0.3–0.4 mm, the narrow and long channel with the highest luminescence intensity appeared on the photos. The channel could be easy observed on vertical Y profile and its width was less than 0.1 mm. This channel is noted as vertical Ych profile. In case of the laser radiation waist situated within the sample there was only one pixel on the y-axis with the maximal intensity on the all photos. It was located on the laser beam axis and its y-position did not depend on Δz. In order to confirm the ASE effect, the vertical Ych profile (Fig. 4), horizontal Z profile and the line profile lying on the axis of channel with maximal intensity (Zaxs profile) were analyzed (Fig. 5).

As it can be easily seen in the Fig. 4a the wave front of Ych profile is the almost plane between $\Delta z < -0.1$ mm and $\Delta z > 1$ mm and becomes Gaussian-like at Δz =0.4 mm. It indicates that the ASE is drastically enhanced between Δz=0 mm and Δz=1 mm and becomes maximal at the critical length Δz=0.4 mm. It is can also be seen in the Fig. 4b, which demonstrates the narrowing of Ych profile at the same Δz. In contrast, the total vertical Y profile of the Yb^{3+} ions luminescence dependence versus Δz has the minimum width at about Δz=0.1–0.2 mm and mainly reflects the behavior of the laser beam cross-section on the front surface of the sample.

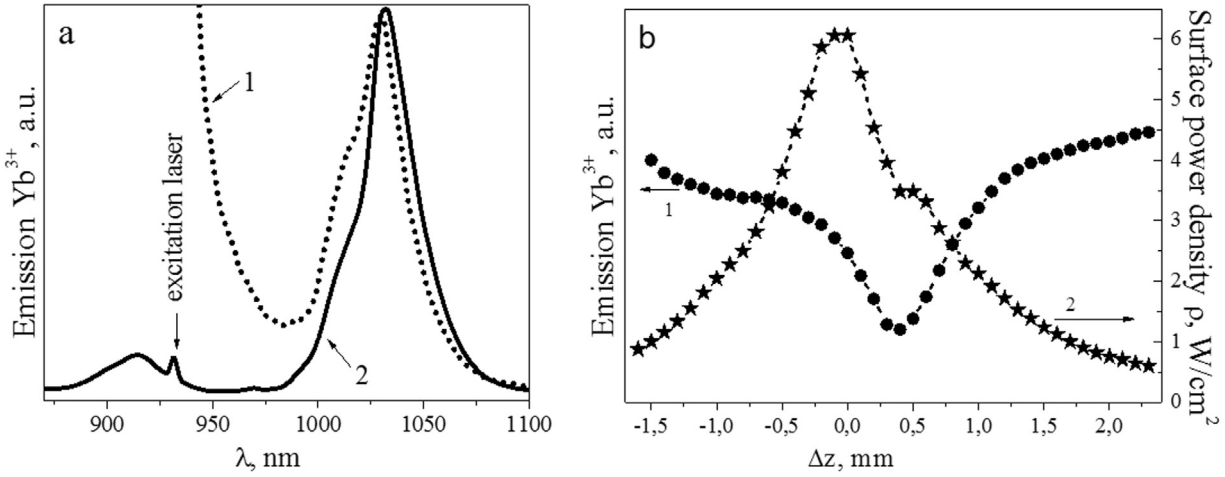

Fig. 2. The non-polarized $^2F_{5/2} \rightarrow {}^2F_{7/2}$ luminescence spectra of Yb^{3+} ions in $LiYbF_4$ crystal recorded along the laser beam (1) and across one (2) (Fig. 2a). The Yb^{3+} ions luminescence peak intensity at λ_{em}=1030 nm (1) and the power density of exciting radiation on the front surface of the sample (2) dependences versus laser beam waist position Δz (Fig. 2b). Excitation wavelength λ_{ex}=932 nm. T=300 K.