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Photon migration at multiple frequencies in $YLiF_4:Nd^{3+}$ crystal

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ABSTRACT

A spatially resolved microluminescence technique was used to measure the spatial distribution of emitted light and photon propagation in Nd^{3+}-doped $YLiF_4$ crystal excited at 514 nm. The spatial distribution of the luminescence was studied and the energy transfer processes among neodymium ions were discussed. We measured the photon diffusion length for specific wavelengths. It was found that re-absorption by ground and excited states plays a crucial role to the propagation. With this information, the microluminescence technique has played a useful role in the investigation of energy transfer processes in rare-earth doped systems.

1. Introduction

Optical energy transfer (ET) is a very common process responsible for the spread of excitation of energy in several systems. It accounts, for example, for the basic phenomena of antenna–electron transfer properties in biological means such as photosynthesis [1,2]. In particular, in solid state laser materials it is responsible for detrimental increase of thermal loading, which decreases the output laser power [3]. Thus, the knowledge of its dynamics is of fundamental importance to the design of optical devices. Therefore, the study of these processes still attracts the attention of the researchers. Usually the temporal behavior of the photoluminescence (PL) is used to check the influence of ET in the dynamics of luminescent systems [4]. However, a great amount of information can also be obtained from the spatial distribution properties of the PL [5]. It has been shown, for example, by using a microluminescence (ML) technique, that there is an optimum ion concentration corresponding to the highest photon diffusion length inside the sample [6]. Tentatively, this information was not clear in the lifetime behavior, and we believe that these techniques are complementary and can be used to explore the ET process in rare-earth doped systems. This kind of spatial measurement has been shown to be an important method of determining the photon migration lengths in glasses [6] and has the important property of being able to detect the spatial migration of energy without spectral diffusion. The sample used in this work, a neodymium-doped yttrium lithium fluoride crystal ($YLiF_4:Nd^{3+}$), has advantages for diode pumping due to its long fluorescence lifetime, typically $\sim$520 μs, which enhances the energy storage inside a laser cavity. The crystal $YLiF_4$, on the other hand, is an attractive host material because of the wavelength match of the laser transition with neodymium glass amplifiers [7], its natural birefringence, and its relatively weak thermal lens in the polarization corresponding to 1053 nm operation [8]. The study of the spatial photon migration, at distinct emission wavelengths, can bring more information about the energy transfer inside this laser crystal. At the same time, ET was observed between different levels of the Nd^{3+} ions. As the majority of ET measurements are realized by time-resolved measurements, in this work we are able to show that pretty similar results can be obtained using the spatial resolved photon migration. To our knowledge this is the first time that such study has been performed in a laser crystal, allowing good knowledge about the energy transfer/migration processes.

2. Experimental

The spatial profile of the emitted luminescence on the surface was obtained by the ML technique [9]. Here, a 514 nm excitation beam was focused by a microscope objective ($\times 10$) onto a microsized spot on the crystal surface. The incident intensity was controlled by neutral density filters. After that, the generated luminescence was collected by the same objective and directed to the spatial scanning system. The scan was performed on the image plane using an optical fiber mounted over a translation stage, as shown in Fig. 1. Usually the image formed is magnified by one order of magnitude. This allows a nice enhancement in the

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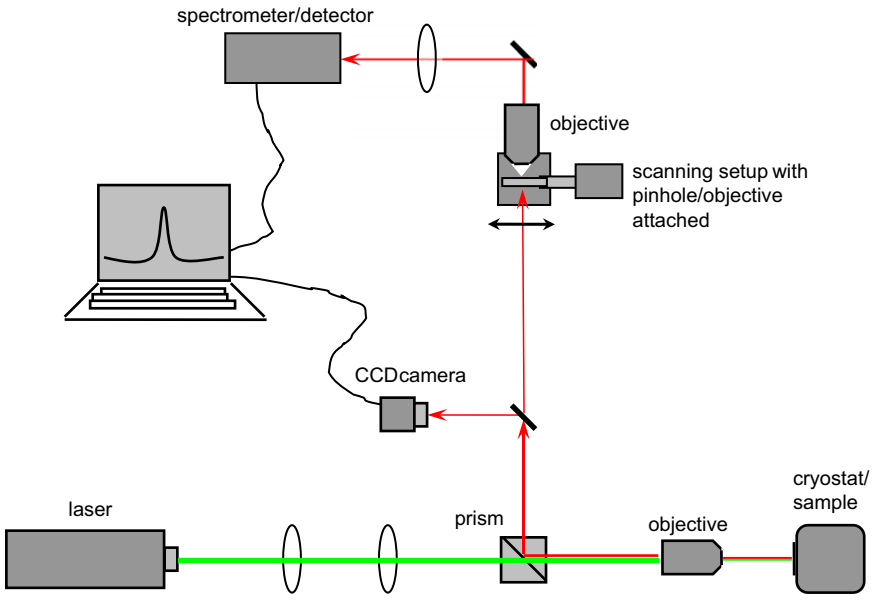

Fig. 1. Experimental setup for the spatially resolved microluminescence technique.

spatial scan resolution. In this work, a 2.0 μm spatial resolution was achieved using a 50 μm diameter optical fiber. Finally, the luminescence was chopped, filtered (with an interference filter to eliminate the laser signal) and analyzed by a spectrometer attached to a refrigerated germanium photodetector. It is worth mentioning that the scanned luminescence was centered in the excitation laser spot to ease the data analysis. A lock-in amplifier enhances the signal detection. Spatial migration of energy was observed at long distances (tens of microns) at low temperatures inside a cryostat at 77 K. The details of this process are discussed with a diffusion theory.

In the ML technique, the incident light excites the rare-earth ions inside a small surface area of the sample and the energy migrates out from this area by photon diffusion processes. The quantum photon transport problem was quantified in terms of the local photon density, n, inside the sample. By considering a diffusive transport for photons this can be accomplished by formulating the problem with the diffusion equation. After excitation, in a time subsequent to the intrinsic lifetime, the spatial energy distribution on the surface follows the equation:

$$L_{ef}^2 \frac{1}{r}\frac{d}{dr}\left(r\frac{dn}{dr}\right) - n = -G_0\,\delta(r) \tag{1}$$

where $L_{ef} = \sqrt{2D\tau}$ is the effective diffusion length that describes the mean distance the photon travels before it is quenched, D is the diffusion constant, τ is the effective lifetime, G_0 is a constant related to the laser excitation intensity, and $\delta(r)$ is the spatial distribution of the excitation beam. This $\delta(r)$ distribution was assumed since the excitation area is typically much smaller than the diffusion area. This equation can be solved analytically providing

$$n(r) = aK_o\left(\frac{r}{L_{ef}}\right) \tag{2}$$

where a is a constant and K_0 is the zeroth-order modified Bessel function. The observed photoluminescence intensity is proportional to the concentrations of excited ions. Since n is proportional to the excited ion concentration, it can be read directly from the luminescent spatial profile, which is a linear function of the excited ion concentration. Thus, Eq. (2) can be used to fit this profile and evaluate L_{ef}.

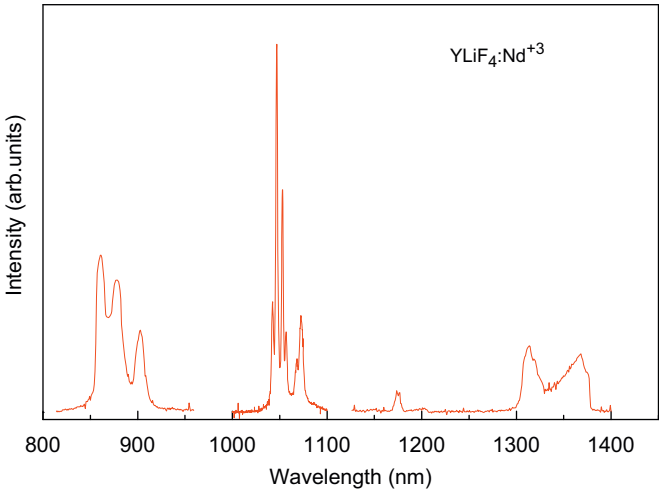

Fig. 2. Fluorescence spectrum of a $YLiF_4$ crystal doped with Nd^{3+} ions.

The studied sample was a 2 mm thick $YLiF_4$ crystal 0.5 mol% Nd^{3+}-doped. All the measurements were performed with the electric field parallel to the c optical axis (π excitation) and at 77 K. The sample was cleaved and polished in order to improve the surface quality.

3. Results and discussion

By the optical pumping the electrons are excited from the $^4I_{9/2}$ ground level to the $^4G_{7/2}$ upper level. After a short lifetime, they decay to the lower excited state $^4F_{3/2}$. The photoluminescence spectrum can be seen in Fig. 2. Here, the lines around 878 nm are originated from $^4F_{3/2} \rightarrow {}^4I_{9/2}$ transition, 1046 nm from $^4F_{3/2} \rightarrow {}^4I_{11/2}$, and 1313 nm from $^4F_{3/2} \rightarrow {}^4I_{13/2}$ manifolds, respectively. In Fig. 3 we have plotted the luminescence scanning profiles corresponding to the several emission lines of the Nd ions. Using these luminescence profiles we are able to obtain the diffusion length constants by fitting the data points with Eq.2. The obtained values of L_{ef} are summarized in Table 1. It is clear that the mean free path for photon propagation is independent of

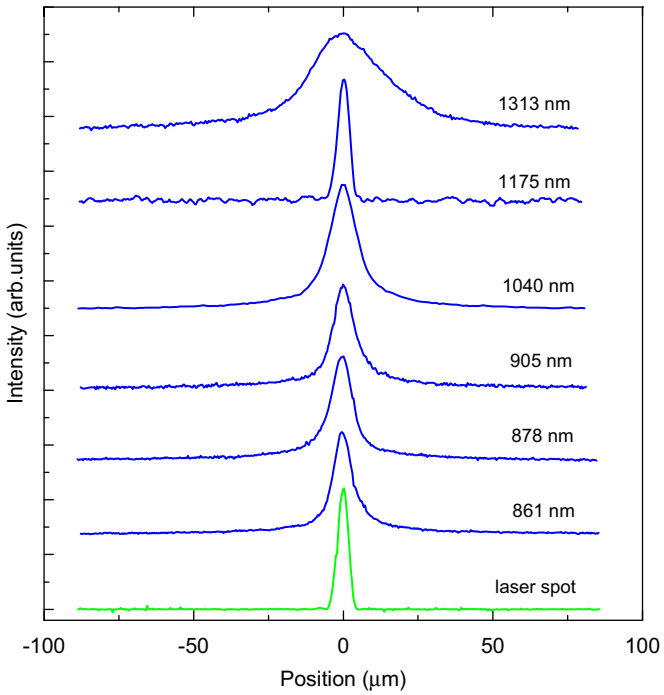

Fig. 3. Luminescence intensity profiles measured on the surface of the Nd^{3+}-doped crystal.

Table 1
Emission line, λ_{em}, and its respective diffusion length, L_{ef}.

λ_{em} (nm)	L_{ef} (μm)
861	7.8
878	8.6
905	8.9
1175	3.6
1313	32
1046	100

the luminescence intensity for a given photon wavelength. For instance, although the luminescence intensity is higher at 861 nm compared to 1313 nm, photons at the line 1313 nm have traveled further making L_{ef} larger than the photons at 861 nm. In order to test the dependence of L_{ef} with the incident power, several measurements were performed at the 1046 nm line. The averaged power ranged from 0.8 to 23.5 mW, roughly two orders of magnitude. As can be seen in Fig. 4, no significant change in the effective diffusion length has been detected inside the instrumental error. This indicates that this parameter is nearly independent of the photon density, and is in good accordance with the used assumptions in our diffusion theory. The only effect noticed in the same figure was an increasing signal-to-noise ratio.

As L_{ef} measures the distance traveled by the photon before quenching, the 1046 nm photon has, by far, the less quenched wavelength. The quenching mechanism, in this case, can be provided by photon re-absorption inside the crystal. The $^4F_{3/2} \rightarrow {}^4I_{9/2}$ emission is strongly absorbed by the ground state of nearby Nd ions [10]. This should shorten the photon propagation arising from this transition. On the other hand, Fornasiero et al [11] have demonstrated that the $^4F_{3/2} \rightarrow {}^4I_{11/2}$ emission has a negligible excited state absorption (ESA). Thus, the well-known laser transition around 1046 nm has the largest L_{ef}. Finally, the $^4F_{3/2} \rightarrow {}^4I_{13/2}$ emission manifolds have a small effective emission cross-section due to ESA [11]. A short L_{ef} can also be seen in Table 1. All these data corroborate the mean free paths found in this work, since the less absorbed photons have traveled larger paths.

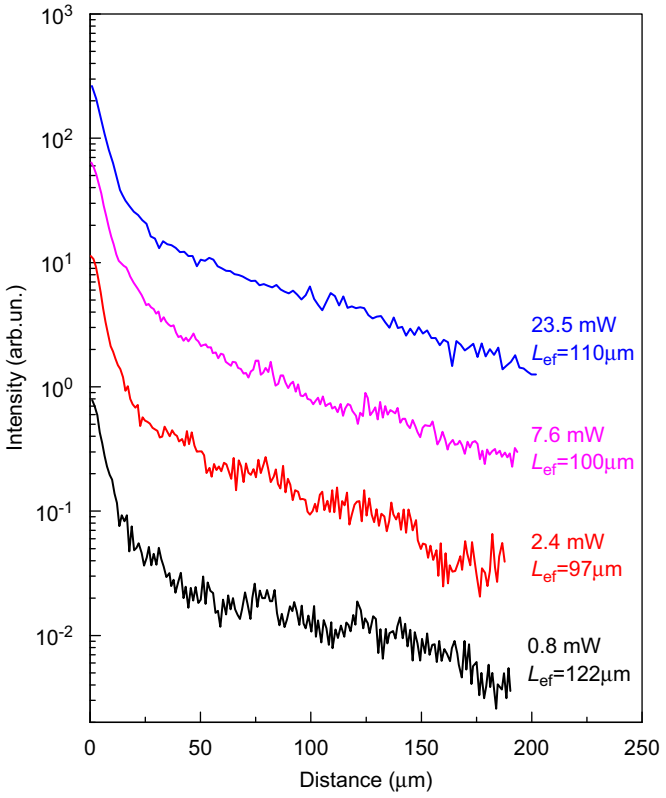

Fig. 4. Profiles in log scale of the luminescence intensity on the surface of the Nd^{3+}-doped crystal as a function of the excitation power. The curves were shifted only to make clear for visualization.

For all the above presented data, regarding the mechanism of luminescence quenching at different wavelength, it is quite possible that trapping play the main role in the quenching processes, although the results reported here do not give direct information. The technique presented here provides complementary data to the transient ET investigations. Indeed, in measuring spatial photon diffusion we are not limited by low signal-to-noise measurements from short lifetimes in time-resolved transient measurements.

4. Conclusions

In conclusion, a spatially resolved microluminescence technique has been used to measure the spatial distribution of emitted light together with the photon propagation in Nd^{3+}-doped $YLiF_4$ crystal excited at 514 nm. The spatial distribution of the luminescence was measured and the energy transfer processes among Nd^{3+}-ions were discussed. The diffusion lengths were measured for specific wavelengths. It was found that re-absorption by ground and excited states have played a crucial role in the photonic propagation. All the obtained data were in accordance with the literature, also suggesting that the ML technique was very useful for the investigation of ET transfer processes in rare-earth doped systems.

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