

Direct confocal lifetime measurements on rare-earth-doped media exhibiting radiation trapping

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Abstract: Radiation trapping occurs in rare-earth-doped active media with strong spectral overlap of luminescence and ground-state absorption. It is demonstrated experimentally that a confocal measurement mitigates the influence of radiation trapping on the measured luminescence lifetime, hence allowing for direct extraction of the lifetime from the measured decay curves. The radiation trapping effect is largely suppressed by probing a small sample volume and rejecting the photons reemitted from the unpumped region. This non-destructive measurement method is applied to ytterbium (Yb^{3+}) activated potassium double tungstate crystalline layers with Yb^{3+} concentrations ranging from 1.2 at.% up to 76 at.% ($\sim 8 \times 10^{19} - 5 \times 10^{21} \text{ cm}^{-3}$). The measured lifetime values are comparable to the results reported for Yb^{3+} -doped potassium double tungstate powder diluted in liquid.

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1. Introduction

Measurement of the luminescent lifetime in a rare-earth-activated gain medium with strong spectral overlap of luminescence and ground-state absorption is known to be severely affected by radiation trapping [1]. A photon spontaneously emitted by an excited ion can be absorbed by another, potentially even rather distant ion in its ground state, which in turn emits a new photon, resulting in considerable lengthening of the measured luminescence lifetime. A higher concentration of active ions in the material increases the occurrence of this process and leads to the observation of longer measured lifetimes as the concentration of active ions increases [2]. Radiation trapping is particularly prominent for the $^2\text{F}_{5/2} \rightarrow ^2\text{F}_{7/2}$ transition in Yb^{3+} at $\sim 1 \mu\text{m}$ wavelength and is also apparent for transitions such as the $^4\text{I}_{13/2} \rightarrow ^4\text{I}_{15/2}$ in Er^{3+} at $\sim 1.5 \mu\text{m}$, the $^3\text{F}_4 \rightarrow ^3\text{H}_6$ in Tm^{3+} at $\sim 2 \mu\text{m}$, and the $^5\text{I}_7 \rightarrow ^5\text{I}_8$ in Ho^{3+} at $\sim 2.1 \mu\text{m}$ [1, 3]. Accurate values of the intrinsic luminescence lifetime are important for determining the emission cross-section via the Füchtbauer-Ladenburg equation [4–7] and for understanding the performance of amplifiers and lasers [8, 9].

Several methods have been suggested to minimize the impact of radiation trapping. Sandwiching a thin active sample by two undoped slabs to avoid total internal reflections, along with aperturing of the emission, was demonstrated on $\text{YAG}:\text{Yb}^{3+}$ [1] and $\text{LiNbO}_3:\text{Er}^{3+}$ [10]. Measurements on powdered samples diluted in liquid were used to determine the lifetime in $\text{YVO}_4:\text{Yb}^{3+}$ [11], $\text{YAlO}_3:\text{Yb}^{3+}$ [12], and $\text{KY}(\text{WO}_4)_2:\text{Yb}^{3+}$ [13]. The pinhole method was suggested by Kühn *et al.* as a non-destructive way of quantifying the lifetime by use of pinholes with diameters ranging from 0.5 to 2.5 mm [14]. The pinhole method has been tested on various Yb^{3+} -doped hosts [14–16]. It involves multiple measurements of the luminescence lifetime collected through pinholes of decreasing diameter. The lifetime value extrapolated to

zero pinhole diameter represents the intrinsic luminescence lifetime. The method, however, may suffer from an uncertainty of the lifetime value due to fluctuations of the measured data and the extrapolation procedure.

In order to largely suppress radiation trapping, the detection volume should be matched to the pumped volume, and this volume should be as small as possible [17]. This goal can be achieved by means of a confocal setup [18]. The use of a confocal setup for lifetime measurements on a rare-earth-doped material was reported by Brynolfsson [19]. However, his implementation was essentially a modified version of the pinhole method proposed by Kühn *et al.*, to the extent that multiple measurements using pinholes with diameters ranging from 1 mm to 10 mm were performed, followed by an extrapolation procedure.

In this work, it is experimentally demonstrated that radiation trapping in highly Yb^{3+} -doped potassium rare-earth double tungstates, $\text{KRE}(\text{WO}_4)_2:\text{Yb}^{3+}$, can be almost entirely eliminated by performing a confocal luminescent lifetime measurement. The non-destructive measurement method allows one to directly extract the lifetime from the measured decay curves without performing multiple measurements with different pinholes and subsequent data extrapolation. This procedure permits us to directly study lifetime quenching in highly-doped $\text{KRE}(\text{WO}_4)_2:\text{Yb}^{3+}$ crystalline layers exhibiting high transition cross-sections and strong spectral overlap of luminescence and ground-state absorption. Only weak lifetime quenching was observed for concentrations up to 76 at.% Yb^{3+} .

2. Measurement setup and samples

Figure 1 shows the setup used for confocal luminescence lifetime measurements. Not indicated in the figure is the pump configuration, namely a fiber-pigtailed single-mode diode laser operating at 981 nm wavelength and modulated at 233 Hz with 50% duty cycle, whose emitted light passes through an optical isolator, after which it is collimated and expanded in free space. The repetition rate and the duty cycle of the pump beam are chosen such that the unpumped duration is more than six times the measured lifetime, allowing for complete relaxation of the Yb^{3+} ions between two excitations. A half-wave-plate ($\lambda/2$ plate) and a polarizing beam splitter (PBS) are used to control the power incident upon the sample. A Si photodetector (Thorlabs DET10A) is placed into the reflected arm of the PBS to serve as a triggering source. An additional $\lambda/2$ plate is placed after the PBS to align the polarization near to the $E||N_m$ optical axis of the sample for maximum pump absorption. The pump beam then passes through a dichroic mirror (Thorlabs DSMP1000) and is focused by a $\times 16$ microscope objective (MO) (NA = 0.32) to a spot size of $\sim 7 \mu\text{m}$. The limitation of the confocal method is due, firstly, to the pump spot size, which should ideally be infinitesimally small, and, secondly, the effective discrimination of luminescence from the non-confocal region via spatial filtering. The luminescence from the sample is collected by the same MO, reflected by the dichroic mirror, and then focused by a $\times 10$ MO (NA = 0.25) through a pinhole with 10 μm diameter. The luminescence passing through the pinhole is then relayed to a second Si photodetector using a 30 mm lens. The collected signal is amplified (FEMTO DHPA-100), sent to a digital storage oscilloscope (Agilent Infinium 54845A), and averaged for 4096 times. Detection of the back-reflected pump beam indicates that the temporal resolution of the measurement system is limited to $\sim 6 \mu\text{s}$ by the switching speed of the pump laser. During the experiment, three pump filters (Thorlabs FEL1000) are placed before the $\times 10$ MO. These filters along with the dichroic mirror strongly attenuate the back-reflected pump light by ~ 100 dB. The launched pump power is maintained at < 2 mW, unless specified otherwise, to avoid stimulated emission and pump-induced heating at the focal spot. A flip mirror and a camera are used to assist in positioning the sample.

The investigated samples are $\text{KRE}(\text{WO}_4)_2:\text{Yb}^{3+}$ epitaxial layers grown onto undoped potassium yttrium double tungstate, $\text{KY}(\text{WO}_4)_2$, substrates with Yb^{3+} concentrations varying from 1.2 at.% to 57 at.%. Detailed information on the growth of these samples and the lattice engineering approach used to achieve such high Yb^{3+} concentration has been previously

reported [20, 21]. The highest-doped sample was a $\text{KLu}(\text{WO}_4)_2:\text{Yb}^{3+}$ (76 at.%) grown onto an undoped $\text{KLu}(\text{WO}_4)_2$ substrate [16, 21]. An overview of the investigated samples is given in Table 1. The samples with 47.5 at.%, 53.5 at.%, 57 at.%, and 76 at.% Yb^{3+} were lapped and polished parallel to the substrate. The other samples were not polished. The sample with 47.5 at.% Yb^{3+} was overgrown with undoped $\text{KY}(\text{WO}_4)_2$.

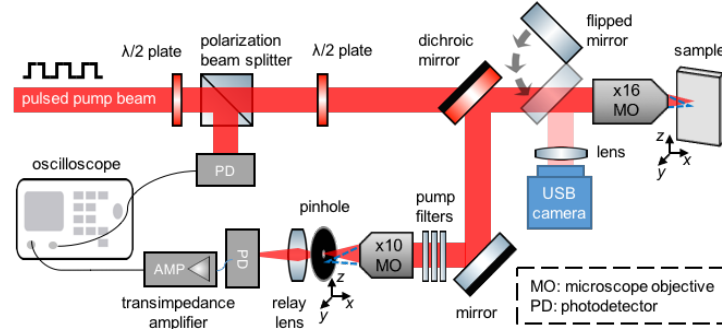


Fig. 1. Schematic diagram of the setup used for confocal luminescence lifetime measurements. The blue dashed foci in front of the two microscope objectives illustrate the discrimination of undesirable luminescence reemitted from the unpumped region of the sample from the detection system by use of a pinhole.

Table 1. Material composition and layer thickness of the Yb^{3+} -doped potassium rare-earth double tungstate epitaxial films.

Nominal composition of epitaxial layer	Substrate	Layer thickness (μm)
$\text{KY}_{0.988}\text{Yb}_{0.012}(\text{WO}_4)_2$	$\text{KY}(\text{WO}_4)_2$	36
$\text{KY}_{0.75}\text{Gd}_{0.10}\text{Yb}_{0.15}(\text{WO}_4)_2$		20
$\text{KY}_{0.67}\text{Gd}_{0.13}\text{Yb}_{0.20}(\text{WO}_4)_2$		42
$\text{KGd}_{0.462}\text{Lu}_{0.213}\text{Yb}_{0.325}(\text{WO}_4)_2$		20
$\text{KGd}_{0.447}\text{Lu}_{0.078}\text{Yb}_{0.475}(\text{WO}_4)_2$		6.7
$\text{KGd}_{0.44}\text{Lu}_{0.025}\text{Yb}_{0.535}(\text{WO}_4)_2$		6
$\text{KGd}_{0.43}\text{Yb}_{0.57}(\text{WO}_4)_2$	$\text{KLu}(\text{WO}_4)_2$	32
$\text{KLu}_{0.24}\text{Yb}_{0.76}(\text{WO}_4)_2$		124

3. Results

Figures 2(a)-2(c) depict representative measured decay curves for the samples with 1.2 at.%, 57 at.%, and 76 at.% Yb^{3+} , respectively. Evidently, the measured luminescence decay curves exhibit a single exponential decay regardless of the Yb^{3+} concentration. In the case of the sample with 1.2 at.% Yb^{3+} , the launched pump power was increased to ~ 50 mW to obtain significant luminescence. In all cases, the lifetime is obtained by fitting the time-dependent luminescence intensity by

$$I(t) = I_0 \exp(-t/\tau) + I_{\text{base}}, \quad (1)$$

where I_0 is the initial luminescence intensity upon cut-off of the pump ($t = 0$), τ is the fitted lifetime, and I_{base} represents the baseline noise level of the detected signal.

The resulting luminescence lifetime values versus Yb^{3+} concentration are shown in Fig. 3 as blue inverted triangles. For each composition, measurements on several randomly chosen positions on the sample were carried out and the resulting lifetime values were averaged. For example, lifetime values of 245 ± 3 μs , 228 ± 10 μs , and 222 ± 9 μs were measured for samples with Yb^{3+} concentrations of 1.2 at.%, 57 at.%, and 76 at.% respectively, which are comparable to the lifetime values measured in $\text{KLu}(\text{WO}_4)_2:\text{Yb}^{3+}$ thin films and extrapolated

by the pinhole method (green squares in Fig. 3) [16] and to the lifetime values measured in bulk Yb^{3+} -doped potassium double tungstate crystals ground to powder and diluted in liquid ethylene glycol (red circles in Fig. 3) [13, 22]. For comparison, measurements were performed by removing the 10 μm pinhole in the setup, i.e., in non-confocal mode. The resulting luminescence lifetimes in non-confocal mode (black triangles in Fig. 3) are elongated to 256 μs for the sample with 1.2 at.% Yb^{3+} and 325 μs for the sample with 75 at.% Yb^{3+} , clearly demonstrating the effect of radiation trapping.

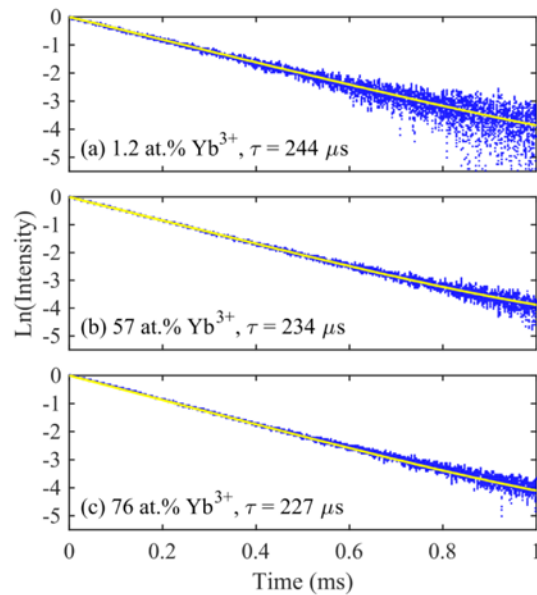


Fig. 2. Measured (blue) and fitted (yellow) luminescent decay curves of samples with (a) 1.2 at.%, (b) 57 at.%, and (c) 76 at.% of Yb^{3+} .

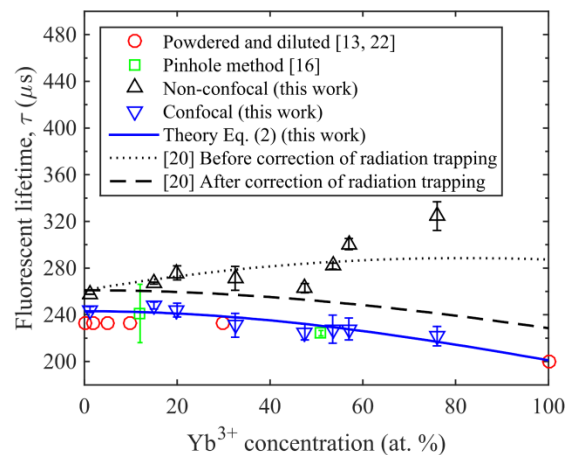


Fig. 3. Luminescence lifetimes measured without (black triangles) and with (blue inverted triangles) confocal feature. Lifetime values measured using either powdered bulk crystals of $\text{KY}(\text{WO}_4)_2:\text{Yb}^{3+}$ [13] and $\text{KYb}(\text{WO}_4)_2$ [22] (red circles), the pinhole method on $\text{KLu}(\text{WO}_4)_2:\text{Yb}^{3+}$ thin films [16] (green squares), and the modified pinhole method before (dotted line) and after (dashed line) correction of radiation trapping [20] are shown for comparison. The blue line is the fitted curve based on Eq. (2).

The setup described in this work permits the direct measurement of luminescence lifetimes thanks to the small probe volume, in combination with confocal detection, in which the 10 μm pinhole rejects the luminescence trapped and reemitted from outside the focal volume. Reported lifetime values measured on $\text{KRE}(\text{WO}_4)_2:\text{Yb}^{3+}$ with low Yb^{3+} concentration without using the pinhole method nor a powdered sample vary widely from 300 to 600 μs [2, 19, 23].

The reduction of lifetime with increasing Yb^{3+} concentration observed in Fig. 3 is ascribed to concentration quenching due to non-radiative energy migration and subsequent energy transfer to a trap, e.g. a crystal defect or an impurity ion, which follows the phenomenological expression [3, 24]

$$\tau(N) = \frac{\tau_0}{1 + (9/2\pi)(N/N_0)^2}, \quad (2)$$

where τ_0 is the measured lifetime at low Yb^{3+} concentration, N is the Yb^{3+} concentration, and N_0 is the critical concentration beyond which the lifetime quenching is severe. Applying Eq. (2) to the measured data using the confocal setup shown in Fig. 3, the fitted values of $\tau_0 = (243.2 \pm 8) \mu\text{s}$ and $N_0 = (1.69 \pm 0.6) \times 10^{22} \text{ cm}^{-3}$ are obtained. The obtained N_0 value agrees well with our previous results [20] of $\tau_0 = 261 \mu\text{s}$ and $N_0 = 2 \times 10^{22} \text{ cm}^{-3}$ measured with the modified pinhole method. The difference in τ_0 leads to the small offset between the blue and the dashed black curve in Fig. 3. The high N_0 value which is much greater than the Yb^{3+} concentration of $6.4 \times 10^{21} \text{ cm}^{-3}$ for stoichiometric $\text{KYb}(\text{WO}_4)_2$ [22] signifies that the luminescence lifetime quenching due to increasing Yb^{3+} concentration is rather weak over the entire doping range. The lifetime value extrapolated to 100 at.% Yb^{3+} doping, i.e., for $\text{KYb}(\text{WO}_4)_2$, is 201 μs , or $\sim 83\%$ of the lifetime at low Yb^{3+} concentration. The results presented in this work reaffirm our previous finding [20] that the lifetime quenching is rather weak and $\text{KRE}(\text{WO}_4)_2$ with high Yb^{3+} concentration may be utilized for amplifiers [9] and lasers [22].

4. Conclusions

In summary, it is demonstrated that confocal lifetime measurements eliminate the elongation of the measured luminescence lifetime owing to radiation trapping. Implementation of the method is shown for $\text{KRE}(\text{WO}_4)_2:\text{Yb}^{3+}$, which has large transition cross-sections and strong spectral overlap between luminescence and ground-state absorption. Rejection of the luminescence reemitted from regions outside the pumped volume by a 10 μm pinhole within the confocal setup allows for direct extraction of the luminescence lifetime without any extrapolation procedure or preparation of a powdered sample. The method is applicable even to samples with very high Yb^{3+} concentration up to $\sim 5 \times 10^{21} \text{ cm}^{-3}$, equaling 76 at.% in $\text{KRE}(\text{WO}_4)_2$, and has allowed us to directly quantify the concentration quenching of luminescence lifetime in $\text{KRE}(\text{WO}_4)_2:\text{Yb}^{3+}$. The method can be readily applied to lifetime measurements on other ground-state-luminescence transitions where radiation trapping prevails, such as materials doped with Er^{3+} , Tm^{3+} , and Ho^{3+} .

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