# Determining the Upconversion Luminescence in a Diode-pumped Nanocrystaline $Nd^{3+}$:$YVO_4$ Random Laser

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**Abstract:** Random lasing in a nanocrystaline $Nd^{3+}$:$YVO_4$ powder is demonstrated for the first time. Lasing action is demonstrated by a method that analyzes the decay kinetics after long-pulse excitation. This method permits to measure the fractional contribution of each component produced in the random laser as a function of the pump intensity, namely spontaneous and stimulated emission as well as upconversion.

**OCIS codes:** 140.0140, Lasers and laser optics; 290.4210, Multiple scattering;290.1350, Backscattering.

## 1. Introduction

Random lasers, which refer to lasing in disordered media, have several interesting features that cannot be achieved with conventional lasers, such as simultaneous emission of several different wavelengths at the same time and emission at new extremely low gain lines [1].

Only few reports exist on diode pumped random lasers [2]. This interesting application which is specially suited for neodymium (Nd) doped materials requires generally that the pump pulse duration is of the order of the spontaneous decay time. As opposed to the usual short pulse pumping technique, this quasi-continuous pumping regime presents qualitative changes in the emission kinetics of the random lasers' stimulated emission. Mosk *et al* presented a detailed theoretical and experimental work on the relaxation oscillations in long-pumped random lasers for a liquid dye containing scatters, concluding that in order to observe relaxations oscillations the linewidth of the laser cavity must be smaller than of the laser medium, a condition which is fulfilled for a dye emission (where the emission linewidth is much larger than the characteristic escape rate of photons from the random laser volume which acts as the cavity) [3]. Noginov *et al* show that upconversion influences the decay time from a Nd doped sample, where a strong pumping density influences the $^4F_{3/2}$ kinetics [4]. However, to our knowledge, no explanations have been performed on the effects of upconversion in the interesting regime of long pump pulses.

The random lasers characteristic relaxation oscillations are normally used to demonstrate in the time domain that the system undergoes laser action [5]. However, the observation of these spikes not only depends on the pump power and the detector type, but also on the kind of nanopowder. Here we demonstrate an alternative method to demonstrate that random emission is occurring.

In this work we demonstrate for the first time lasing action in Nd:$YVO_4$ nanopowder as well as a method to determine quantitatively the upconversion rate and the contribution of the spontaneous emission in the samples backscattering cone as a function of pump power.

## 2. Experiment

The nanopowder was obtained by grinding a 1.66 mol% $Nd^{3+}$:$YVO_4$ laser crystal. The concentration was determined by the EDX technique. The powder morphology was determined using SEM (Cambridge Instruments, Model 360) and the laser diffraction technique (CILAS – 825 nm) as shown in Fig.1.

The mean diameter of the nanoparticles was 390 nm. A compressed sample with flat surfaces and dimension of Φ5x1 $mm^3$ was pumped by a quasi-continuous 110 W laser diode bar, operating at 809 nm with 3 Hz repetition rate, 100 μs pulse width and 1 μs pulse decay time. The excitation beam was focused to a square shape with area of 5.33 $mm^2$ and the samples backscattered luminescence was separated from the pump excitation by a beam splitter and analyzed using a fast oscilloscope and a spectrometer. We choose a QCW laser diode as pump source, because the wavelength can be tuned to be resonant to the $Nd^{3+}$ absorption peak and pulse duration can match $Nd^{3+}$:$YVO_4$ lifetime well (for 1.66 mol% about 60 μs). Therefore, enough gain can be introduced by relatively low peak pump power and QCW operation.

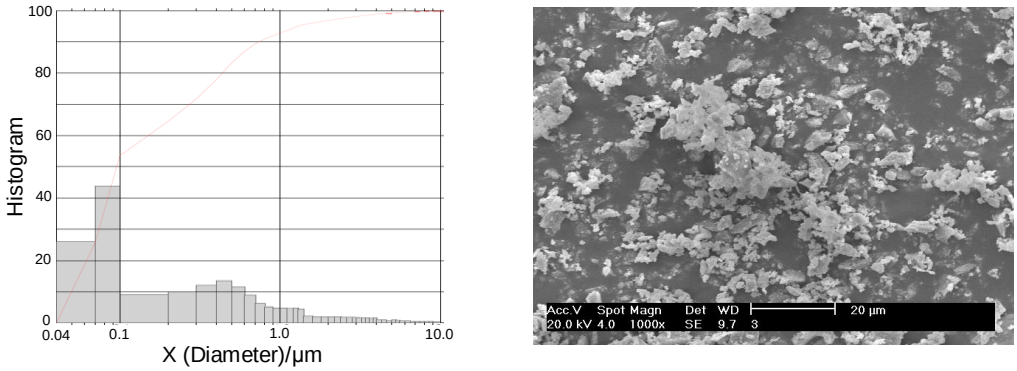


**Fig. 1.** Left: Number particle distribution versus the diameter of the Nd:$YVO_4$ nanopowder sample, obtained by the laser diffraction technique, range: 0.04 - 10µm. Right: Scanning electron microscopy (SEM) of the surface of this sample.

In Fig. 2 the experimental setup is shown. Two cylindrical divergent lenses were used for a modal conformation of the pump beam, so that by inserting a 20 mm spherical lens at 77 mm from the diode bar, a near square shape focus of the excitation beam could be achieved, with area of 5.33 $mm^2$. The samples backscattered luminescence was separated from the pump excitation by a beam splitter and analyzed using a spectrometer (Ocean Optics, Model HR2000 – resolution: 0.11 nm) and a fast oscilloscope (Lecroy, resolution 1 ns). Time resolution was limited by the Germanium photodetector with 1.5 µs fall time.

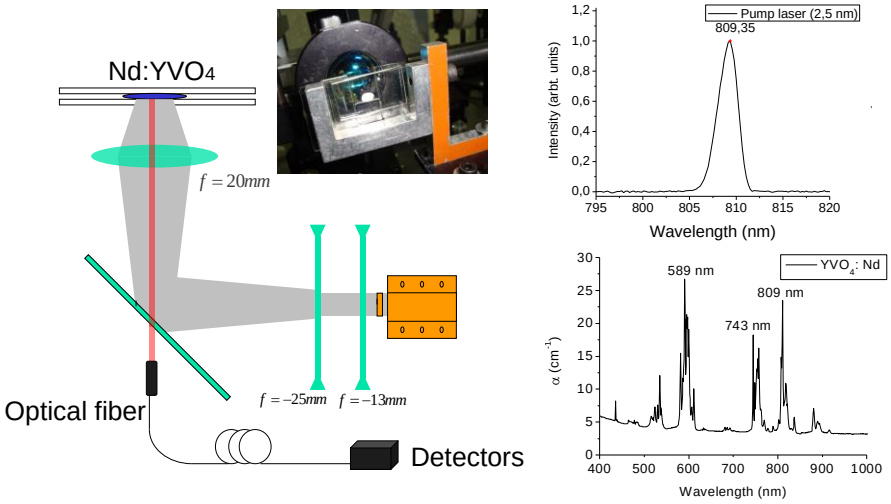


**Figure 2.** Experimental setup. The samples luminescence was separated from the pump excitation by a beam splitter. Inset: detail of the sample which is confined by glass slabs in the front and the back. Upper right: the emission from the diode bar which was set to maximum absorption peak of Nd. Lower right: absorption spectra of the sample.

## 3. Results

At low pump intensity (0.8 mW/$mm^2$), several fluorescent emissions from $Nd^{3+}$ transitions were visible, as seen in Fig. 3a. Increasing the pump power gradually, a threshold pump intensity was observed, at which a sharp emission line at the $^4F_{3/2} \rightarrow {}^4I_{11/2}$ transition (1064.12 nm) appeared whose spectral width decreased as a function of pump power, from 1.30 nm to 0.48 nm (inset in Fig. 3a). The IR-pulse decay was fitted with a double exponential function which permitted to calculate the fraction of spontaneous and stimulated emission. Figure 3b shows the decay in log-log scale and the fitting parameters at 100 W of diode peak pump power.

Two decay components were measured, a fast stimulated emission component with a time constant of 6.5 µs and a spontaneous emission component with a time constant of 58 µs.

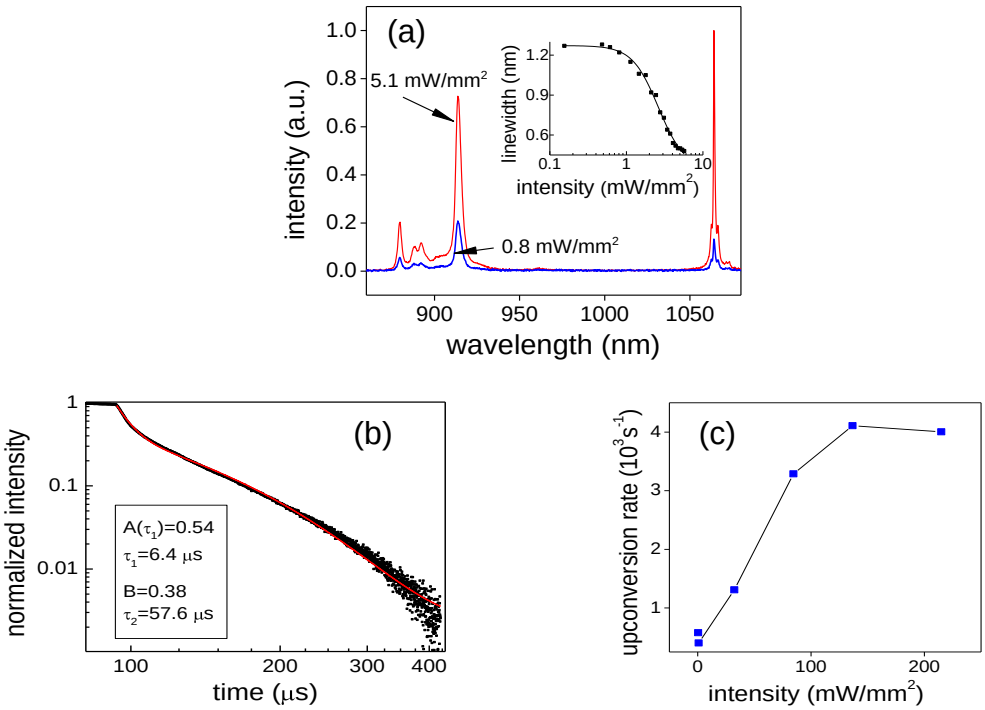


**Fig. 3:** (a) Emission spectra as a function of average pump intensity. The inset shows the observed linewidth narrowing of the $^4F_{3/2} \rightarrow {}^4I_{11/2}$ emission. (b) Laser pulse decay and fit with two exponentials for a pump intensity of 5.1 mW/mm$^2$. (c) Saturation of the upconversion rate as a function of peak pump intensity.

## 4. Discussion

The analysis allows determining the fractional contribution of each component produced in the random laser as a function of the pump intensity because there is a direct relation between the laser emission broadening and the contribution of the spontaneous emission to the laser signal captured in the detector field of view. With decreasing contribution of the spontaneous emission at higher pump intensities, the laser emission narrows. The fraction A/(A+B) corresponds to stimulated emission. The up-conversion mechanism ($^4F_{3/2}$, $^4F_{3/2}$) $\rightarrow$ ($^4I_{15/2}$, $^2G_{7/2}$) that contributes to the upper laser level depopulation was well observed by the non-exponential decay curve of the spontaneous emission as a function of the peak pump intensity when laser action is not present. The best luminescence fitting was obtained using the Inokuti-Hirayama model [6] where the energy-transfer parameter $\gamma(s^{-1/2})$ was taken and the decay time constant ($\tau_2$) was obtained by the integration of the normalized decay. The up-conversion rate (Fig. 3c) was taken using the relation $W_{up}(s^{-1}) = 1/\tau_2(I) - 1/\tau_2(0)$, where $\tau_2(0)$ is equal to 58 μs including the cross-relaxation ($^4F_{3/2}$, $^4I_{9/2}$) $\rightarrow$ ($^4I_{15/2}$, $^4I_{13/2}$). This value is expected for Nd = 1.66 mol % as obtained with $\tau = 100\mu s\ [1- (Nd/Nc)^2]$ where Nc is 2.56 mol%.

## 5. References


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