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Superfluorescence from $NdCl_3$

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Abstract

The fluorescence and superfluorescence properties of $NdCl_3$ were studied as a function of pump power and temperature. At low temperatures, we observed superfluorescence from $NdCl_3$ at 694.4 nm under excitation at 355 nm. The threshold dependence upon temperature is presented and analyzed.

1. Introduction

It is well known that cooperative effects involving many ions can deeply modify the nature of spontaneous emission [1]. One such effect is superfluorescence (SF), that is, cooperative spontaneous emission involving many ions [2–6]. Similar to superradiance (SR) and spontaneous emission amplified by stimulated emission (ASE), SF is also characterized by spectral narrowing, lifetime shortening and some kind of threshold in the output versus pumping power. Initially, SF begins as spontaneous emission with intensity proportional to the pump power but, as the coherence establishes, the intensity scales rapidly and follows a square law versus pumping power. The distinctions among SF, SR and ASE have been introduced and discussed in Ref. [3].

Although many lanthanide-doped host lattices and lanthanide compounds have shown the laser effect, there are few reports [7] on superfluorescence for this kind of material. In this Letter, we studied the fluorescence and SF properties of $NdCl_3$ as a function of the pump power and temperature. A noticeable result of our study is the appearance of a threshold above which all emission lines but 694.4 nm are quenched and a short nanosecond pulse (shorter than the laser pulse used) is emitted. Detailed studies have shown that the observed effect is superfluorescence. The threshold dependence on temperature and onset time versus pumping power are also presented and analyzed.

2. Experiment

The sample of $NdCl_3$ used in the present study was grown by the Bridgman technique. The thickness of $NdCl_3$ is about 1 mm with two unpolished non-parallel faces so that no optical feedback is likely to occur. In the experiment, the sample was mounted in a quartz tube which was sealed under 0.6 atm of He and placed in a liquid-helium optical cryostat (Oxford International) with a regulated,

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heated gas system allowing the temperature to be varied between 10 and 300 K. The excitation sources are a pulsed YAG:Nd (Quantel 20 W) laser with a repetition rate of 30 Hz and a duration of 10 ns and a dye laser pumped by the YAG:Nd laser. The emission was analyzed with a Jobin-Yvon HR-1000 monochromator with dispersion of 0.8 nm/mm and detected by a Stanford SR-510 lock-in amplifier. The luminescence decay curve was measured with a Lecroy 9350 M oscilloscope (500 MHz) interfaced with a computer.

3. Results and discussion

Below about 230 mW of incident power with the sample at 12 K, the emission spectrum of $NdCl_3$ in the range 550–700 nm under excitation at 355 nm consists of about 20 sharp lines which fall into five groups (Fig. 1a). They correspond to $^4G_{5/2} \rightarrow {}^4I_{9/2}$, $^4D_{3/2} \rightarrow {}^4F_{3/2}$, $^4D_{3/2} \rightarrow {}^4F_{5/2}$, $^2H_{9/2}$, $^4G_{5/2} \rightarrow {}^4I_{11/2}$ and $^4D_{3/2} \rightarrow {}^4F_{7/2}$, $^4S_{3/2}$ transitions respectively, with $^4G_{5/2} \rightarrow {}^4I_{9/2}$ the strongest ones. All the above assignments were checked and confirmed by selective excitation and transient measurements. Above this threshold, recorded spectra peaked only at 694.4 nm and the peak, attributed to $^4D_{3/2} \rightarrow {}^4S_{3/2}$ transitions, clearly shows a spectral narrowing effect (Fig. 1b). As a consequence of this process, all the excited ions were transferred to the $^4S_{3/2}$ state which, according to the energy level diagram of Nd^{3+} [8], should enhance greatly the infrared emission of the sample. Such effect is under study. Output intensity versus incident pumping power was also measured. The result is shown on a log–log diagram (Fig. 2). Above the threshold, the best fit of the experimental results gives a slope of 2.05, showing that the emission follows a square law versus pumping power. It should be pointed out that, in all experiments conducted, no lenses were used to focus the pumping laser. Considering that the 355 nm (28 169 cm^{-1}) excitation is not very efficient [9] and the laser spot

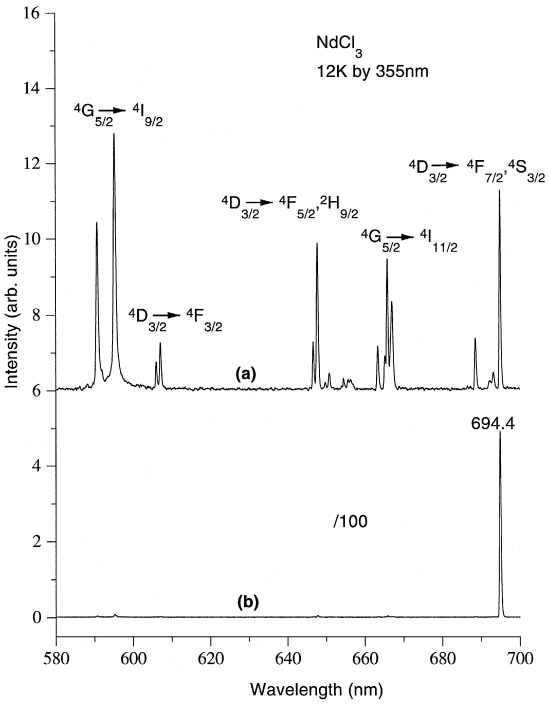

Fig. 1. Emission spectra of $NdCl_3$ under 355 nm excitation at 12 K. (a) Incident power 30 mW and (b) incident power 340 mW. Note the scale change for the latter.

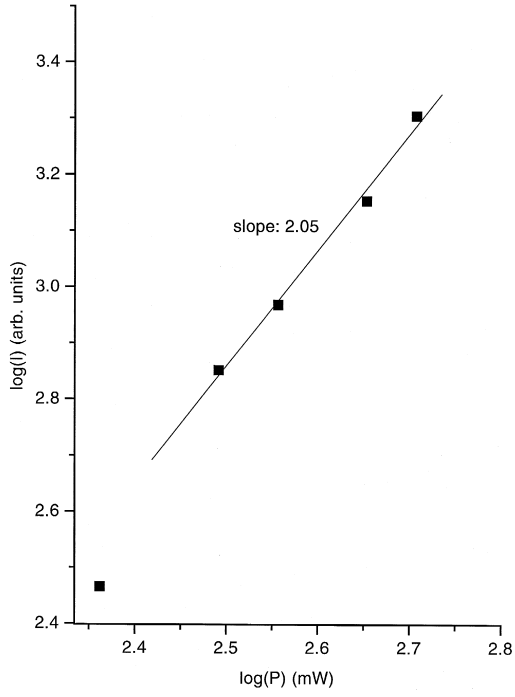

Fig. 2. Fluorescence intensity at 694.4 nm (in a.u.) versus incident power. Only the results above the threshold are shown in the figure.

was about 6 mm in diameter, the actual threshold is quite low.

The temporal behavior obtained under a quasi-continuous pulse of 10 ns duration and 30 Hz repetition is shown in Fig. 3a before the threshold (150 mW of incident power) and in Fig. 3b after the threshold (510, 450, 360, and 310 mW of incident power). Fitting curve (a) gives the lifetime of the $^4D_{3/2}$ level of Nd^{3+} in $NdCl_3$ at 12 K as 52 ns, which is much shorter than that found in $LaCl_3$:Nd^{3+} (10.5 μs) [10]. Above the threshold, the emission shortened to a pulse of a few nanoseconds, obviously shorter than the pump laser pulse. Due to the limitation of the experimental apparatus, we cannot determine the exact delay time of this short pulse. However, results presented in Fig. 3b show that the onset time becomes shorter as the pump power increases.

The emission of short pulses above a pumping threshold has already been reported by several authors [11–13]. These observations have been interpreted in terms of stimulated emission [11,12] or laser-like behavior [13]. We know that stimulated emission differs from SF in two aspects: (i) the flux dependence: the ASE has a linear output power dependence along with the incident power while SF would rather follow a quadratic law, and (ii) the propagation direction: the stimulated emission has a collimation of the output beam while SF has not. Our analysis of the emitted energy as a function of pump power (Fig. 2) and the observation of uncollimated emission light support a SF behavior. The temporal dependence shown in Fig. 3 confirmed the conclusion.

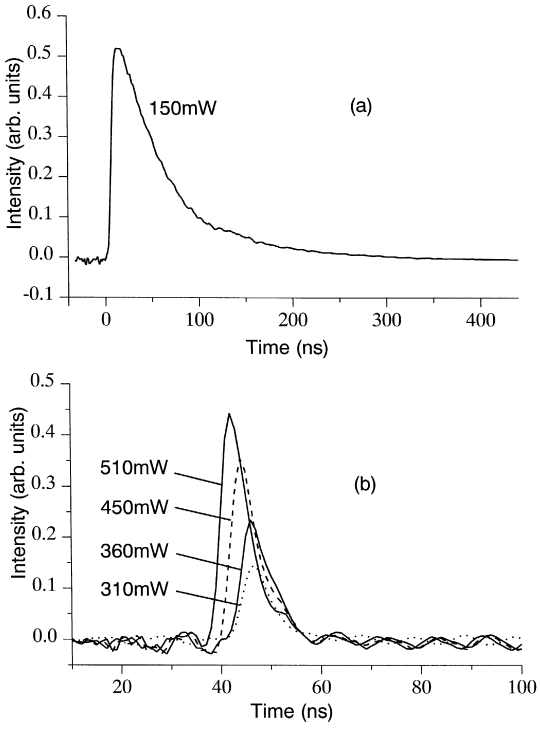

Fig. 3. Temporal behavior at 694.4 nm under different incident power excitation. (a) Below and (b) above the threshold.

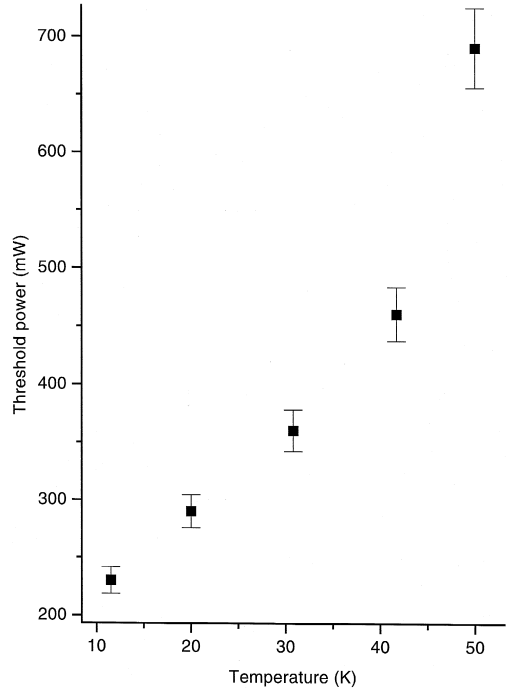

Fig. 4. Threshold dependence on temperature.

Fig. 4 presents the threshold dependence upon the temperature. The threshold increases with increasing temperature. At the beginning, the increase is not large, from 230 mW at 12 K to 290 mW at 20 K, but as the temperature rises, the increasing rate becomes more and more important: at 41.7 K, the threshold is 460 mW and reaches 690 mW at 50 K, the highest power available with our apparatus. The phenomenon can be explained as follows: a larger cross-relaxation rate at higher temperature [14] induces a weaker emission and a shorter lifetime. In

fact, we did find that the lifetime of the radiative level $^4D_{3/2}$ becomes pronouncedly shorter at room temperature compared to 12 K.

4. Conclusion

From square-law behavior, spectral narrowing, reduction of decay time (from 52 ns to a few nanoseconds), and no observation of a propagation direction of the emission light, we have shown that superfluorescence at 694.4 nm was obtained in the $NdCl_3$ crystal under 355 nm excitation at low temperature.

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