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RELAXATION OF HIGH LYING EXCITED STATES OF Nd³⁺ IONS IN YAG: Nd³⁺ AND IN YAP: Nd³⁺

M.F. JOUBERT, J.C. COUDERC and B. JACQUIER

*Laboratoire de Physico-Chimie des Matériaux Luminescents, Université Lyon I, Bât. 205,
43 boulevard du 11 novembre 1918, F-69622 Villeurbanne cedex, France*

Abstract - After selective excitation in either the $^2P_{3/2}$ or the $^4D_{3/2}$ multiplet of Nd³⁺ in YAG or in YAP, the fluorescence properties of these levels are analysed. The YAG:Nd³⁺ sample exhibits a very strong visible fluorescence attributed to transitions from the $^2F(2)_{5/2}$ multiplet as a consequence of a very efficient excited state absorption mechanism from the $^2P_{3/2}$ metastable state.

1.- Introduction

Upconversion pumping of the blue and violet emission in neodymium doped materials due to two-step, excited state absorption processes has been reported in LaF₃ /1,2/, YLiF₄ /3-5/ and YAG /6/. The different origin and behavior of the laser emission in these materials is a strong motivation to understand the mechanisms of fluorescence decay from the upper state of Nd³⁺ in general. A recent paper reported a comparison of the lifetimes of the high lying metastable states $^2F(2)_{5/2}$, $^4D_{3/2}$ and $^2P_{3/2}$ in YLiF₄, LaF₃ and YAG /7/ using direct ultraviolet excitation. In this work, these fluorescence lifetimes in YAG:Nd³⁺ and in YAP:Nd³⁺ have been measured and analysed. In YAG:Nd³⁺ only, resonant excitation in the $^2P_{3/2}$ or $^4D_{3/2}$ multiplet induced antistokes fluorescence from the $^2F(2)_{5/2}$ metastable state.

2.- Experimental results

Single crystals containing neodymium with concentration of 0.11 at % in YAG and 2.5 at % in YAP were used for the fluorescence measurements.

In addition to the experimental equipment described earlier /7/, an analogic to digital conversion technique and a sampling oscilloscope LECROY 1410 were used to analyse the fluorescence properties at short time.

Resonant ultraviolet excitations into the $^2P_{3/2}$, $^4D_{3/2}$ and $^2F(2)_{7/2}$ multiplets of both Nd^{3+} activated crystals were used to measure their fluorescence decay times which are given in table 1.

In $YAP:Nd^{3+}$, the $^2P_{3/2}$ fluorescence exhibits an exponential decay with a time constant of 960 ns at 6K and 800 ns at 300 K. Figure 1 shows the emission spectrum at 6 K relative to the $^4D_{3/2} \rightarrow ^4F_{3/2}$ transition of Nd^{3+} in YAP. The 10 ns pulse duration of the laser used does not enable us to evaluate precisely the decay time of this fluorescence but our estimation agrees with the 15.5 ns reported in a recent paper /8/. After selective excitation in the $^2F(2)_{7/2}$ level, the emission spectrum is quite complex. The lines related to the $^2F(2)_{5/2}$ fluorescence are weak and their time evolution is not purely exponential. The value given in table 1 correspond to the short time exponential part. A long time component is observed until about 1 ms due to a background broad band emission. Moreover, many impurity lines are present in the whole visible range.

In $YAG:Nd^{3+}$, the $^2P_{3/2}$ fluorescence exhibits an exponential decay with a time constant of 300 ns at 6K and 150 ns at 300K. Figure 2 shows the emission spectrum at 6K around 6200 Å measured after selective excitation into one of the $^4D_{3/2}$ Stark component of Nd^{3+} in YAG. The lines related to the $^4D_{3/2} \rightarrow ^4F_{3/2}$ transition are weak compared to those related to the antistokes $^2F(2)_{5/2} \rightarrow ^2K_{15/2}$ fluorescence. The decay of the former on the oscilloscope display screen images the pulse laser shape showing a time constant below 10 ns in

Table 1: Summary of fluorescence decay times from the metastable states of Nd^{3+} in $YAP:Nd$ and in $YAG:Nd$.
from ref /10/

		$YAlO_3:Nd^{3+}$	$Y_3Al_5O_{12}:Nd^{3+}$
$^4F_{3/2}$	6K	160 μs	270 μs
	300K	144 μs	267 μs
$^2P_{3/2}$	6K	960 ns	300 ns
	300K	800 ns	150 ns
$^4D_{3/2}$	6K	~ 12 ns	< 10 ns
	300K		
$^2F(2)_{5/2}$	6K	3 μs	5 μs
	300K		3 μs #

agreement with the 2.2ns reported recently /8/, while the latter exhibit pure exponential decays with a time constant of 5 μ s. Figure 3 presents the fluorescence spectra related to the $^2F(2)_{5/2} \rightarrow ^4F_{5/2} + ^2H_{9/2}$ transition of Nd^{3+} in YAG using different excitation wavelengths. After excitation into $^4D_{3/2}$ or $^2P_{3/2}$, the spectrum shows only the lines related to the emission from the $^2F(2)_{5/2}$ metastable state. While after direct excitation in the $^2F(2)_{7/2}$ level, a background is observed in the whole near UV and visible range, showing a long time behavior until a few milliseconds.

Fig 1. Fluorescence spectrum relative to the $^4D_{3/2} \rightarrow ^4F_{3/2}$ transition of Nd^{3+} in $YAlO_3$, following excitation into $^4D_{3/2}$.

Fig. 2. Fluorescence spectrum of Nd^{3+} in YAG, following excitation into $^4D_{3/2}$. The lines (at 6190Å and 6223Å) related to the $^4D_{3/2} \rightarrow ^4F_{3/2}$ transition are weaker than those related to the $^2F(2)_{5/2} \rightarrow ^2K_{15/2}$ transition.

3.- Discussion

From an earlier study of the multiphonon emission rate versus energy gap between energy levels of trivalent rare-earth ions in various hosts /9/, it seems that this non radiative process is the dominant $^2P_{3/2}$ relaxation mechanism in $YAG:Nd^{3+}$ and in $YAP:Nd^{3+}$; while the

Fig. 3. Fluorescence spectra relate to the ${}^2F(2)_{5/2} \rightarrow {}^4F_{5/2} + {}^2H_{9/2}$ transition of Nd^{3+} in YAG a) following excitation into ${}^4D_{3/2}$ b) following excitation into ${}^2F(2)_{7/2}$

short ${}^4D_{3/2}$ fluorescence time constant (a few nanoseconds) may suggest a non negligible contribution of cross relaxation transfer process, specially in the highly Nd^{3+} doped YAP sample used. A concentration evolution of this lifetime would be necessary to check this hypothesis.

The antistokes ${}^2F(2)_{5/2}$ fluorescence was observed in $YAG:Nd^{3+}$ after excitation in ${}^2P_{3/2}$ or ${}^4D_{3/2}$. So, we think that it is a consequence of an efficient excited state absorption process from the ${}^2P_{3/2}$ metastable state and not from the ${}^4D_{3/2}$ level as it was suggested before /10/. The excited state absorption process terminates to a $4f^2-5d$ level. Rapid relaxation occurs to the lower edge of this band located at 42000 cm^{-1} /11/ and multiphonon relaxation to the ${}^2F(2)_{7/2}$ multiplet of the $4f^3$ configuration leads to the ${}^2F(2)_{5/2}$ fluorescence. In $YAP:Nd^{3+}$, such an ${}^2F(2)_{5/2}$ antistokes emission was not observed. In this material, the Nd^{3+} $4f^2-5d$ configuration is located between 51000 and 59000 cm^{-1} /12/. So, if an electron is excited to a $4f^2-5d$ level, the probability of multiphonon relaxation to the ${}^2F(2)_{7/2}$ multiplet (39550 cm^{-1}) is expected to be very low due to the large energy gap.

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