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Influence of composition on the fluorescence spectra and fluorescence decay profiles of Nd^{3+} in the laser material ASN (Aluminate de Strontium-Néodyme)

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Abstract

This paper reports on the time-resolved emission spectra and the fluorescence decays of Nd^{3+} in ASN ($\text{Sr}_{1-x}\text{Nd}_y\text{La}_{x-y}\text{Mg}_x\text{Al}_{12-x}\text{O}_{19}$ with $0 < x < 1$ and $0 < y \leq x$). The Nd^{3+} decay profiles are fitted according to the Hopping model. The decay time decreases with increasing y due to strong interactions between Nd^{3+} ions. Fluorescence intensity is given as a function of x and y and is maximum for y values close to 0.1.

1. Introduction

ASN is a new laser material^{1,2} with composition $\text{Sr}_{1-x}\text{Nd}_y\text{Mg}_x\text{Al}_{12-x}\text{O}_{19}$. Laser experiments have been performed, demonstrating efficient diode-pumped laser output and tunability in the one-micron band.

In order to maintain a low rate of neodymium to avoid quenching of fluorescence while improving the crystal quality by preventing the formation of parasitic phases, one can dilute Nd^{3+} by La^{3+} ions. In $\text{Sr}_{1-x}\text{Nd}_y\text{La}_{x-y}\text{Mg}_x\text{Al}_{12-x}\text{O}_{19}$ (mentioned as $\text{Sr}_{1-x}\text{Nd}_y\text{La}_x$), the Sr^{2+} and Nd^{3+} concentrations can vary independently. The floating zone method is used to grow three series of single crystals : $\text{Sr}_{1-y}\text{Nd}_y$, $\text{Sr}_{0.8}\text{Nd}_y\text{La}_{0.2-y}$ and $\text{Sr}_{0.6}\text{Nd}_y\text{La}_{0.4-y}$. Fluorescence properties as a function of x and y values are studied in order to optimize optical properties with a view to perform comparative laser tests in a near future.

2. Fluorescence spectra

The $^4\text{F}_{3/2} \rightarrow ^4\text{I}_{9/2}$ transition at room and low temperature has been studied in details. The general features of the fluorescence spectra (band shifts and line widths) depend only on the Sr^{2+} rate and not on the Nd^{3+} one (Figure 1).

At room temperature we can detect at least seven lines for the $\text{Sr}_{0.98}\text{Nd}_{0.02}$ crystal (Figure 1.a). Each one can be assigned to one of the ten transitions of $^4\text{F}_{3/2} \rightarrow ^4\text{I}_{9/2}$. The

fluorescence spectrum of $\text{Sr}_{0.60}\text{Nd}_{0.10}\text{La}_{0.3}$ has been recorded at 13 K (Figure 2). At this temperature, we can clearly observe three lines and two shoulders which correspond to the five transitions issued from the lowest Stark level of $^4F_{3/2}$, the only one populated at 13 K.

Fig.1 : Nd^{3+} emission spectra ($^4F_{3/2} \rightarrow ^4I_{9/2}$) at room temperature ($\lambda_{\text{exc}} = 532 \text{ nm}$) in
a) $\text{Sr}_{0.98}\text{Nd}_{0.02}$ b) $\text{Sr}_{0.80}\text{Nd}_{0.02}\text{La}_{0.18}$ and
c) $\text{Sr}_{0.60}\text{Nd}_{0.02}\text{La}_{0.38}$.

Fig.2 : Nd^{3+} emission spectrum ($^4F_{3/2} \rightarrow ^4I_{9/2}$) at 13 K in $\text{Sr}_{0.6}\text{Nd}_{0.1}\text{La}_{0.3}$ ($\lambda_{\text{exc}} = 532 \text{ nm}$).

3. Fluorescence decays

Since the decay curves are non exponential, a mean decay time τ_m is calculated as the normalized area under the decay curve. The mean lifetime strongly decreases when the Nd^{3+} rate increases. For example, τ_m falls down from 380 to 80 μs when y increases from 0.01 to 0.20 in $\text{Sr}_{0.8}\text{Nd}_y\text{La}_{0.2-y}$. This behaviour is typical of strong interactions between Nd^{3+} ions. On the other hand, the strontium rate dependence of τ_m is very weak.

In a more quantitative investigation of the RT and 13 K decay curves, we consider the Hopping formula which specify the nature of the interactions between Nd^{3+} ions :

$I = I_0 \exp(-t/\tau_0 - K_D - \gamma\sqrt{t})$ where I_0 is the intensity at $t = 0$, τ_0 is the radiative decay time, K_D corresponds to the energy migration while γ is connected to the cross-relaxation. For long times after the excitation pulse, the fluorescence intensity decays exponentially at a rate τ_1 called lifetime at long times and defined as follow : $1/\tau_1 = 1/\tau_0 + K_D$.

The τ_1 versus y curves are given for the three series $\text{Sr}_{1-y}\text{Nd}_y$, $\text{Sr}_{0.8}\text{Nd}_y\text{La}_{0.2-y}$ and $\text{Sr}_{0.6}\text{Nd}_y\text{La}_{0.4-y}$ on Figure 3. As for τ_m , τ_1 decreases strongly when the Nd^{3+} concentration increases. But, in this case, this occurs from energy migration exclusively. For very small Nd^{3+} content, τ_1 is almost constant so energy migration is negligible. This gives a first estimation of τ_0 close to 420 μs . Knowing τ_1 , γ can be extracted from the decay curves at short time. The γ curves for the three series of compounds are given on Figure 4. Up to 20 %

of Nd^{3+} , the variation of γ is almost linear and follows the formula $\gamma = 4/3 \pi^{3/2} N_A \sqrt{C_{DA}}$ where N_A is the number of acceptor ions per unit volume and C_{DA} the cross-relaxation microparameter. For this low concentration domain, we can calculate the microparameter $C_{DA} = 2.21 \times 10^{-40} \text{ cm}^6 \text{ s}^{-1}$.

Fig. 3 : Lifetime at long times τ_l as a function of Nd^{3+} content (y %) in $\text{Sr}_{1-x}\text{Nd}_y\text{La}_{x-y}$ for various x values.

Fig. 4 : Cross-relaxation parameter γ as a function of Nd^{3+} content (y %) in $\text{Sr}_{1-x}\text{Nd}_y\text{La}_{x-y}$ for various x values.

4. Fluorescence intensity as a function of neodymium content

Fig.5: Integrated Nd^{3+} (${}^4F_{3/2} \rightarrow {}^4I_{9/2}$) emission intensity as a function of Nd^{3+} content (y %) in $\text{Sr}_{1-x}\text{Nd}_y\text{La}_{x-y}$ for various x values.

Fluorescence intensity is proportional to the total number of emitted photons. As lifetimes vary with Nd^{3+} concentration, one has measured the time-integrated intensity. Besides, as line widths of the spectra are x dependent, we have integrated the fluorescence intensity on the whole range of wavelength for the transition ${}^4F_{3/2} \rightarrow {}^4I_{9/2}$. The fluorescence intensity measured this way (Figure 5) depends strongly upon Nd^{3+} concentration. Whatever the series studied, the intensity curve shows off a maximum for a Nd^{3+} concentration between 5 and 15 %.

5. Conclusion

The aim of our work was to find the best composition of $\text{Sr}_{1-x}\text{Nd}_y\text{La}_{x-y}\text{Mg}_x\text{Al}_{12-x}\text{O}_{19}$ from the optical properties point of view. The fluorescence intensity is maximum for a Nd^{3+} rate between 5 and 15 ions % with respect to the number of large cation sites corresponding to neodymium concentrations between 1.7 and 5×10^{20} ions per cm^3 . These values are rather large compared to optimum Nd^{3+} concentration in YAG : Nd (1 % Nd^{3+} , e.g., 1.4×10^{20} ions per cm^3). It has to be noticed that in this concentration range, the lifetimes are still quite high ($\approx 300 \mu\text{s}$ for $y = 10 \%$) which looks promising for the laser properties to be studied.

From the optical properties, a selection of the Nd^{3+} concentration range is made. But, in view of laser applications, other considerations have to be taken into account. The main problem for ASN is that for high strontium content the melting of the compounds is no longer congruent. Besides, the higher the Sr^{2+} content is, the easier the $\vec{c}$ growth (the preferred direction for laser action) occurs.

On the basis of these results, several selected compositions ($\text{Sr}_{1-x}\text{Nd}_y\text{La}_{x-y}$ with $0.6 \leq 1-x \leq 0.8$ and $0.05 \leq y \leq 0.15$) are going to be grown by the Czochralski method in order to obtain large samples on which laser tests will be performed.

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