PROCEEDINGS

OF THE
INTERNATIONAL CONFERENCE
ON

LASERS 2001

TUCSON, ARIZONA **DECEMBER 3–7, 2001**

V.J. CORCORAN & T.A. CORCORAN
Editors

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THE SOCIETY FOR OPTICAL & QUANTUM ELECTRONICS

STS PRESS • McLEAN, VA • 2002

SPECTROSCOPIC AND LASER PROPERTIES OF Nd^{3+} IN $LaSc_3(BO_3)_4$

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Abstract

Spectroscopic laser properties have been characterized for Nd^{3+} in $LaSc_3(BO_3)_4$. The Judd-Ofelt analysis has been applied to the room temperature absorption spectrum to determine the radiative decay rates and branching ratios of Nd^{3+} transitions from the $^4F_{3/2}$ state to the ^4I_J lower-lying manifolds. The parameters Ω_2, Ω_4, and Ω_6 are larger than those reported for Nd^{3+} in other laser host crystals. The value of Ω_4/Ω_6 is approximately 3.0 times larger than that of Nd^{3+} in YAG and about 1.4 times larger than that of Nd^{3+} in the β-phase of $LaSc_3(BO_3)$ reported recently. The room temperature fluorescence lifetime of the $^4F_{3/2} \rightarrow {}^4I_{11/2}$ transition is 150 μs and the Judd-Ofelt analysis predicts a radiative lifetime for the $^4F_{3/2}$ state to be 249 μs. Therefore, the fluorescence quantum efficiency is determined 60%. The emission cross-sections of the $^4F_{3/2} \rightarrow {}^4I_{11/2}$ and $^4F_{3/2} \rightarrow {}^4I_{13/2}$ transitions have been also determined at room temperature. Finally, these results are compared with those of the standard laser material Nd^{3+}:YAG.

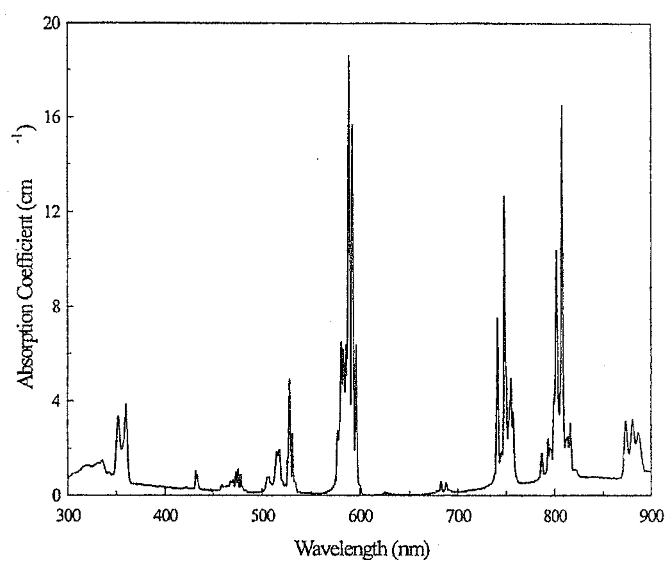
Introduction

Trivalent neodymium-doped lanthanum scandium borate, $LaSc_3(BO_3)_4$, also known as NLSB, demonstrates potential as an efficient material for diode-pumped microchip lasers. Some of the earliest work on NLSB was reported by Meyn, Jensen, and Huber,[1] and by Kutsvoi et al.[2] More recently, Gruber and coworkers have reported results obtained from modeling the detailed crystal-field splitting of the Nd^{3+} ion energy levels and the Q-switch performance of 1.06 μm and 1.34 μm stimulated emission from microchip NLSB used in different applications, including range finding.[3,4]

The LSB crystal host can accommodate relatively high concentrations of Nd^{3+} ions with relatively low concentration quenching as a consequence. We report in this article the Judd-Ofelt analysis of the spectroscopic parameters of Nd^{3+} in this crystal. To support our analysis of NLSB, we include experimental data, including lifetime, emission cross sections, and branching ratios obtained at room temperature for intermanifold transitions $^4F_{3/2} \rightarrow {}^4I_J$ and compare these results with those of NYAG.

Experiments

The room temperature absorption spectrum of NLSB was recorded with a Cary 14 spectrophotometer and is presented in Fig. 1. The room temperature luminescence spectra were measured on Nd^{3+}:LSB by exciting the sample with the 514.5 nm line from a Spectra Physics argon-ion laser model 2005. The emission from the sample was collected by a pair of positive lenses at 90° with respect to the exciting laser beam, and detected by a liquid nitrogen-cooled Ge detector attached to the exit slit of a SPEX model 340E monochromator. A reflection grating with 600 grooves/mm blazed at 1 μm was used for the measurements of all emission spectra. The signal was then taken from the detector to a Spex Datascan model DS 1000. A personal computer was used to control the monochromator and to acquire and analyze the data. The emission lifetimes were measured by exciting the sample with the 514.5 nm line from the argon-ion laser; the laser light was chopped at 20 Hz. The decay signals were then displayed on a 150 MHz Tektronix model 2445A



oscilloscope to determine the lifetimes. The room temperature decay of the $^4F_{3/2}$ state in Nd^{3+}:LSB was found to be fit a single exponential curve for the $^4F_{3/2} \rightarrow {}^4I_{11/2}$ transition.

Results and Discussions

Ten absorption bands identified in the room temperature absorption spectrum between 300 and 900 nm portrayed in Fig. 1, tabulated in Table I and were chosen to determine the phenomenological Judd-Ofelt (J-O) parameters for Nd^{3+}:LSB. Measured line strengths, S_{meas} $(J \rightarrow J')$, of the chosen bands are determined using the following expression:[5]

$$S_{meas}(J \rightarrow J') = \frac{3ch(2J+1)n}{8\pi^3 \bar{\lambda} e^2 N_0}\left[\frac{9}{(n^2+2)^2}\right]\Gamma, \quad (1)$$

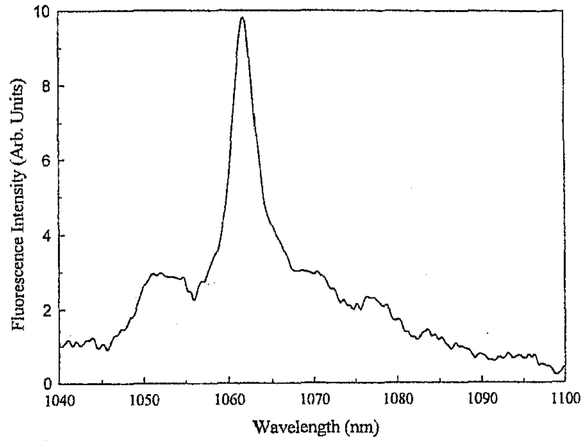
where J and J' are the total angular momentum quantum numbers of the initial and final states, respectively, n is the refractive index, N_0 is the Nd^{3+} ion concentration, $\bar{\lambda}$ is the mean wavelength of the specific absorption band, $\Gamma = \int \alpha(\lambda) d\lambda$ is the integrated absorption coefficient as a function of λ, and c and h have their usual meaning. The factor $[9/(n^2 + 2)^2]$ in Eq. (1) represents the local field correction for the ion in the dielectric host medium. The measured line strengths were then used to obtain the J-O parameters Ω_2, Ω_4, and Ω_6 by solving a set of ten equations for the corresponding transitions between J and J' manifolds in the following form[5]:

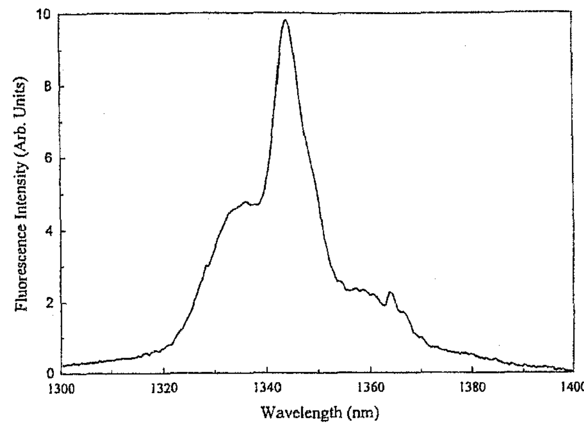
$$S_{calc}(J \rightarrow J') = \sum_{t=2,4,6} \Omega_t \left|\left\langle (S,L)J \left\| U^{(t)} \right\| (S',L')J' \right\rangle\right|^2, \quad (2)$$

where the matrix elements $\left\langle \left\| U^{(t)} \right\| \right\rangle$ are doubly reduced unit tensor operators of rank t calculated in the intermediate-coupling approximation and are independent of the crystal host. Values of the reduced matrix elements for the chosen Nd^{3+} bands are taken from Carnall et al.[6] For these calculations of absorption line strengths and other spectroscopic parameters, the index of refraction was taken to be 1.83 (independent of wavelength). The values of the measured absorption line strengths, S_{meas} are tabulated in Table I. A least-squares fitting of S_{meas} to S_{calc} provides the following values for the three J-O parameters for Nd^{3+} in LSB host material: $\Omega_2 = 5.349 \times 10^{-20}$ cm^2, $\Omega_4 = 4.124 \times 10^{-20}$ cm^2, and $\Omega_6 = 3.852 \times 10^{-20}$ cm^2.

Table I. Measured and calculated absorption line strengths of Nd^{3+} in LSB.

Transition (from $^4I_{9/2}$)	λ (nm)	S_{meas} (10^{-20} cm^2)	S_{calc} (10^{-20} cm^2)
$^4F_{3/2}$	883.5	0.948	1.157
$^4F_{5/2} + {}^2H_{9/2}$	807.4	3.300	3.039
$^4F_{7/2} + {}^4S_{3/2}$	749.6	2.548	2.732
$^4F_{9/2}$	687.7	0.126	0.203
$^4G_{5/2} + {}^2G_{7/2}$	586.9	7.916	7.917
$^2K_{13/2} + {}^4G_{7/2} + {}^4G_{9/2}$	521.2	1.728	1.744
$^2K_{15/2} + {}^2G_{9/2} + ({}^2D,{}^2P)_{3/2} + {}^4G_{11/2}$	472.3	0.434	0.327
$^2P_{1/2}$	433.8	0.147	0.151
$^4D_{3/2} + {}^4D_{5/2} + {}^2I_{11/2} + {}^2D_{1/2}$	356.5	2.357	2.379
$^2L_{15/2}$	332.2	0.314	0.140





The J-O parameters can now be applied to Eq. (2) to calculate the line strengths corresponding to the transitions from the $^4F_{3/2}$ state to the lower lying $^4I_{J'}$ (J' = 15/2, 13/2, 11/2, 9/2) manifolds of Nd^{3+} in NLSB. Using these line strengths, the radiative transition rates for electric dipole transitions between an excited state and the lower lying terminal levels, $A\ (J \rightarrow J')$, can be calculated using the following expression:

$$A(J \rightarrow J') = \frac{64\pi^4 e^2}{3h(2J+1)\bar{\lambda}^3} \frac{n(n^2+2)^2}{9} S_{calc}(J \rightarrow J'). \qquad (4)$$

The radiative lifetime, τ_r, for an excited state (J) is calculated by

$$\tau_r = \frac{1}{\sum A(J \rightarrow J')}, \qquad (5)$$

where the sum is over all final lower-lying states J'. The radiative rates for all four transitions are added to obtain the total radiative rate of 4021.78 s^{-1} of the $^4F_{3/2}$ metastable state. The radiative lifetime of this level is, therefore, determined 249 μs.

The room temperature luminescence branching ratios, $\beta(J \rightarrow J')$, can be determined from the radiative decay rates by

$$\beta(J \rightarrow J') = \frac{A(J \rightarrow J')}{\sum A(J \rightarrow J')} = A(J \rightarrow J')\tau_r, \qquad (6)$$

where the sum runs over all final states J'.

Figures 2 and 3 display the room temperature emission spectra for the Nd^{3+} $^4F_{3/2} \rightarrow {}^4I_{11/2}$ and $^4F_{3/2} \rightarrow {}^4I_{13/2}$ transitions, respectively. The emission cross sections of these transitions can be determined from the following expression[7]

$$\sigma(J, J'; \tilde{\nu}) = \frac{\lambda^2}{8\pi c n^2} \frac{\beta(J, J')}{\tau_r} g(\tilde{\nu}), \qquad (7)$$

where λ is the central peak wavelength, $\tilde{\nu} = \lambda^{-1}$, $\beta(J, J')$ is the luminescence branching ratio from the upper manifold J to the lower manifold J', τ_r is the radiative lifetime of the excited state manifold J, and $g(\tilde{\nu})$ is the line shape function. The values of radiative lifetime and intermanifold branching ratios, and the values of λ and n are applied to Eq. (7) to determine the emission cross sections for the $^4F_{3/2} \rightarrow {}^4I_{11/2}$ and $^4F_{3/2} \rightarrow {}^4I_{13/2}$ transitions, which are given in Table II.

Table II. Optical properties predicted by Judd-Ofelt theory and principal inter-Stark and intermanifold emission cross sections of Nd3+ in a-LSB.

Radiative lifetime	$\tau_r(^4F_{3/2}) = 249\ \mu s$
Branching ratios	$\beta(^4F_{3/2} \rightarrow {}^4I_{9/2}) = 0.44$
	$\beta(^4F_{3/2} \rightarrow {}^4I_{11/2}) = 0.47$
	$\beta(^4F_{3/2} \rightarrow {}^4I_{13/2}) = 0.087$
	$\beta(^4F_{3/2} \rightarrow {}^4I_{15/2}) = 0.005$
Emission cross sections	$\sigma(^4F_{3/2} \rightarrow {}^4I_{11/2}) = 1.45 \times 10^{-19}\ cm^2$
	$\sigma(^4F_{3/2} \rightarrow {}^4I_{13/2}) = 4.083 \times 10^{-20}\ cm^2$

Conclusions

An in-depth spectroscopic analysis of Nd^{3+} in LSB laser host has been performed following the Judd-Ofelt theory. The three phenomenological parameters were found to be $\Omega_2 = 5.349 \times 10^{-20}\ cm^2$, $\Omega_4 = 4.124 \times 10^{-20}\ cm^2$, and $\Omega_6 = 3.852 \times 10^{-20}\ cm^2$; these values are higher than those reported for other Nd^{3+}-based laser crystals. The spectroscopic quality factor, $X = \Omega_4/\Omega_6$, of this sample is approximately 1.07, which is approximately 3.0 times larger than that of Nd:YAG.[7] As a result, the major laser transitions for NLSB are likely to be more intense than in other laser crystals. The room temperature fluorescence lifetime (τ_f) of the $^4F_{3/2} \rightarrow {}^4I_{11/2}$ transition of NLSB is measured as 150 μs, and the radiative lifetime (τ_r) of the $^4F_{3/2}$ state determined by the J-O analysis was found to be 249 μs, resulting in the fluorescence quantum efficiency ($QE = \tau_f / \tau_r$) of 60%.

The emission cross sections of the $^4F_{3/2} \rightarrow {}^4I_{11/2}$ and $^4F_{3/2} \rightarrow {}^4I_{13/2}$ transitions have been determined from the room temperature emission spectra. The emission cross section of the well-known transition, $^4F_{3/2} \rightarrow {}^4I_{11/2}$, of Nd^{3+}:LSB is found to be $1.45 \times 10^{-19}\ cm^2$, while that of Nd^{3+}:YAG is $2.8 \times 10^{-19}\ cm^2$. These values are agreeable to within the experimental uncertainties.

Finally, this study shows that NLSB possesses some competitive spectroscopic and laser properties and should be considered a potential contender as an efficient laser system that can be pumped with powerful diode lasers whose pump wavelength overlaps the strong Nd^{3+} absorption band around 808 nm, which is about three times broader than that of Nd:YAG.

Acknowledgement

The authors would like to thank Dr. Thomas A. Reynolds for supplying them with the sample investigated. This research was supported by the National Science Foundation Grant No. DMR 0099479.

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