

The Variation Effect of Doping Percentage upon Radiative Fluorescence Transitions for Nd: glass

Adawiya J. Haider*, Shehab A. Al-Zubaidi, Duha S. Ahmed and Lina Y. Majeed
Applied Physics Department, University of Technology, Baghdad-Iraq.

* E-mail: Adawiya_Haider@yahoo.com.

Abstract

In this work we determine an optimum set of intensity parameters ($\Omega_{(i)}$) by using the standard least-square fitting. The values of radiative transition probability (A_{rad}), radiative lifetime (t_{rad}^c) and emission branching ratio coefficient (b_r) have been determined for the major excited state. The present work focuses on the absorption bands at 530.81 nm wavelength for the active medium, which coincidences with the emission spectrum of the diode laser which is commonly used to pump the solid state laser.

Keyword: host material, active space, solid state laser, Nd: YAG, Nd: Glass.

1- Introduction

The laser active medium should have a narrow fluorescence line, wide absorption band zone and high quantum efficiency which agree with the spectrum emission fluorescence for the specific wavelength, these characteristics are cleared in solid-state laser (crystal and glass) that is made impure with small amount of elements which have a spectral transitions in the inner unsaturated levels, like transition metal, rare earth materials and actinides [1]. The important characteristic that distinguishes the solid-state laser from other lasers (gas, liquid) is the high density active ions that contributes to laser action, its density is about ($10^{22} - 10^{25} \text{ ion}/\text{m}^3$) when compared with gas and liquid media which is about ($10^{18} - 10^{20} \text{ ion}/\text{m}^3$) that means the required volume to produce definite quantity power is small compared with gas laser media. In addition the lifetime of the electrons in the excited level for solid is about (10^{-3} sec) compared with (10^{-6} sec) in case of gases, that makes the active media of solid materials a big energy reservoir that gives a high pulse laser energy [2].

Solid-State lasers are preferable for most applications because of their ruggedness, relative simplicity and ease of operation but the problem is that such lasers are not currently available with specific operational parameters required for some applications. The requirements for solid-state lasers having a

wide variety of operational parameters for different types of applications have provided the stimulus for significant technological advancements in this field over the past several years [3].

Neodymium ions form the basis for a series of high power solid state lasers. In the two most common variants, the ^{+3}Nd ions are doped into either Yttrium Aluminum Garnet (YAG) crystals or into a glass host, these two lasers are known as either Nd:YAG or Nd:glass. The main laser transition is in the near infrared at 1064nm [4]. When doped in YAG, the Nd: YAG crystal produced laser output primarily at 1064 nm; when doped in glass, the Nd: glass medium lases at wavelengths ranging from 1054nm to 1062nm depending on the type of glass used [5]. The wavelength does not change much on varying the host. For the relative efficiency, it is known that ^{+3}Nd :Glass lasers are more efficient than ^{+3}Nd :YAG lasers with discrepancy up to (5%), which could be attributed to two causes [6]:

1. High doping ratio in glass which reaches (6%).
2. Absorption lines are wider in glass.

The critical components of solid-state lasers include the laser material, the mechanism of pumping and the cavity configuration. The most fundamental requirement for a laser material is that it can be easily and economically produced with high quality in a large size. The host material must

be transparent to both the pump light and the lasing light. The optical spectral properties of a laser material are determined by the electronic transitions of the host. The types of ions that are useful for laser emission in the near ultraviolet, visible and near-infrared spectral regions are transition-metal ions and rare-earth ions. Both of these types of ions have electrons configurations that include unfilled shells and thus have electron transitions between energy levels within a specific shell (d-d transitions for transition metal ions and f-f transitions for rare-earth ions). These give rise to absorption and emission transitions in the appropriate spectral range [3].

2-Experimental work

2-1 Measurements for Radiative Fluorescence Transitions

From the absorption spectrum and fluorescence transitions for the solid state active medium samples, it could be possible to calculate many parameters that demonstrate the sample's characteristics, where the active medium is a collection of atoms or molecules which can be stimulated to a population inversion and emit electromagnetic radiation in a stimulated emission [7]. Fluorescence is the emission of light of a particular wavelength as a result of absorption of light at shorter wavelength, it is a property of some materials and each material has a specific wavelength of absorption and emission [7]. The calculations of spectroscopic fluorescence transitions from the excited state ${}^4F_{3/2}$ that is the population inversion level for ${}^{+3}Nd$ active ion to the lowest sublevels (${}^4I_{9/2}$, ${}^4I_{11/2}$, ${}^4I_{13/2}$, ${}^4I_{15/2}$) gives a fluorescence lines at 880, 1064, 1350 and 1880nm wavelengths respectively [10, 11]. In addition to that, it could be possible to calculate the spectral parameters for fluorescence transitions from the excited level ${}^4F_{3/2}$, fluorescence parameters involve spontaneous emission probability $A_{(rad)}$, radiative lifetime t_{rad}^c . Then from the fluorescence transitions one can calculate the branching ratio coefficient b_r and laser transition lines intensity $S_{(f)}$.

The (J-O) parameters could be applied as in equation (1) to calculate the fluorescence line strength ($S_{(f)}$) corresponding to the transitions from the excited state to the lower sublevels [11].

$$S_{ED(ab)} = \sum_{t=2,4,6} \Omega_t \left| \langle (S, L)J | U^{(t)} | (\overline{S}, \overline{L})\overline{J} \rangle \right|^2 \quad \dots(1)$$

where ($\Omega_{(t)}$) is the intensity parameters for the fluorescence transitions calculated from the absorption spectrum measurements with the square numerical matrix element values ($|U^{(t)}|^2$) as in table 1.

$|(S, L)J\rangle$ is initial manifold

$|\overline{(S, L)}\overline{J}\rangle$ is terminal manifold

$S_{ED(ab)}$ is calculated line strength transition

The measured values of line strength $S_{ED(m)}$ could be determined by performing numerical integration on main basic absorption bands to evaluate area under the curve ($\int K_{(\lambda)} d\lambda$) by using (L.S.F) and Simpson's rule program using equation 2, and then the equation below gives $S_{ED(m)}$ [12]:

$$\left(\int K_{(\lambda)} d\lambda \right) = \frac{8\pi^3 e^2 \bar{\lambda} \rho}{3ch(2J+1)n} \left[\frac{(n^2+2)^2}{9} \right] S_{ED(m)} \quad \dots(2)$$

where:

$K_{(l)}$ is the absorption coefficient at wavelength λ . r is the ${}^{+3}Nd$ ion concentration.

$\bar{\lambda}$ is the average wavelength value of the absorption band. J the total angular momentum of the initial value.

$n = n_{(l)}$ The index of refraction at wavelength $\bar{\lambda}$.

The refractive index values are taken for each sample in the fluorescence Wavelengths range as in equation 3 to implement the calculation of spontaneous radiative transition probabilities A_{rad} from the line strength for excited ${}^4F_{3/2}$ to 4I_J manifold [11]:

$$A_{rad} [(S, L)J; (\bar{S}, \bar{L})\bar{J}] = \frac{64\pi^4 e^2}{3h(2J+1)\lambda^3} \left[\frac{n(n^2+2)^2}{9} \right] S_f \quad \dots(3)$$

where:

S_f is the fluorescence line strength.

A_{rad} is the spontaneous radiative transition.

n is the refractive index of material.

h is Planck's constant.

I is the wavelength value of the absorption band. Now from the summation of these transition probabilities one can find the total spontaneous radiative transition probability A_{total} for each sample.

The radiative lifetime (t_{rad}^c) is related to the total radiative transition probabilities A_{rad} of all transitions from the initial manifold (J) to the final manifold (\bar{J}) as in the equation as explained equation 4 [13, 14]:

$$\tau_{rad}^c = \left\{ \sum_{\bar{S}, \bar{L}, \bar{J}} A [(S, L)J; (\bar{S}, \bar{L})\bar{J}] \right\}^{-1} = A_{total}^{-1} \quad \dots(4)$$

where A_{total} is the total spontaneous radiative transition probability, t_{rad}^c is the radiative lifetime. It therefore involves the effective average over site to the variation of Nd^{3+} active ion environment in host materials, the emission branching ratio transition b_r originating from initial manifold can be obtained from the radiative transition probabilities A_{rad} using equation 5 [11]:

$$\beta [(S, L)J; (\bar{S}, \bar{L})\bar{J}] = \frac{A [(S, L)J; (\bar{S}, \bar{L})\bar{J}]}{\sum_{\bar{S}, \bar{L}, \bar{J}} A [(S, L)J; (\bar{S}, \bar{L})\bar{J}]} \quad \dots(5)$$

where \sum is the summation over all terminal manifold. $f(J, \bar{J})$ is the theoretical oscillator strength of the ($J \rightarrow \bar{J}$) transition at the mean frequency. The radiative quantum efficiency (h_c) could be calculated from equation 6 after getting the measured fluorescence lifetime (t_f^m) [15]:

$$\eta_c = \frac{\tau_f^m}{\tau_{rad}^c} \quad \dots(6)$$

where t_f^m is the measured fluorescence

lifetime due to all relaxation processes t_{rad}^c is radiative lifetime.

Now the peak stimulated emission cross-section (σ_{em}) could be calculated using equation 7 with existence of the effective fluorescence bandwidth (ΔI_{eff}) as in equation 8 [16]:

$$\sigma_{em} = \frac{\lambda_p^4}{8\pi c n^2(\lambda_p) \Delta \lambda_{eff}} A \left[\left({}^4F_J \right); \left({}^4I_J \right) \right] \quad \dots(7)$$

where I_p is the peak fluorescence wavelength of emission band and c is the speed of light in vacuum. $n_{(I_p)}^2$ is the refractive index at each emission peak wavelength and ΔI_{eff} is the effective fluorescence bandwidth determined by integration of the fluorescence line shape ($\int I_l dI$) divided by the intensity at peak fluorescence wavelength (I_p) as in the equation 8 [17]:

$$\Delta I_{eff} = \int (I_{(l)} dI / I_{max}) \quad \dots(8)$$

While the average non-radiative decay (W_{nr}) can be calculated as in equation 9 [13]:

$$W_{nr} = \frac{1}{t_f^m} - \sum_b A_{(ab)} \quad \dots(9)$$

where n_r is the refractive index, $A_{(ab)}$ is radiative transition probabilities, t_f^m is the measured fluorescence lifetime due to all relaxation processes. As a final remark table 2 illustrates the spectral parameter values for fluorescence transitions from the excited state ${}^4F_{3/2}$ for each sample.

3- Results and Discussions

3-1 The Effect of the Doping Percentage

Variation upon the Radiative Fluorescence Transition

The spectral parameter values for the four fluorescence transitions from the excited state (${}^4F_{3/2}$) to the lower states (4I_J) are influenced by the varying of active ion doping percentage for the same host; this could be indicated from the intensity parameter values

$(\Omega_{(i)})$ that are calculated using (J-O) theoretical model.

The fluorescence line strength values $(S_{(f)})$ increase obviously with the active ion doping percentage, which is clear from the following transitions:

$${}^4F_{3/2} \rightarrow {}^4I_{9/2} \text{ (880 nm)}$$

${}^4F_{3/2} \rightarrow {}^4I_{11/2}$ (1064 nm) strongest transition

As a result the spontaneous emission probability value A_{rad} increases according to the fluorescence line strength $(S_{(f)})$ which is illustrated in table 2, this increase is clear in the same two transitions above.

The laser intensity line transitions are limited to the emission branching ratio coefficient b_r , as illustrated in table 2, the transition for the two transitions is shown below:-

$${}^4F_{3/2} \rightarrow {}^4I_{13/2} \text{ (1350 nm)}$$

$${}^4F_{3/2} \rightarrow {}^4I_{15/2} \text{ (1800 nm)}$$

This is very small, so it is always neglected in comparison with the other transitions. The summation of the spontaneous emission probability values A_{total} is illustrated in table 2, it increases with the active ion doping percentage increasing. The radiative lifetime t_{rad}^c is related to the total radiative transition probabilities A_{total} for all transitions from the excited state ${}^4F_{3/2}$ to the lower states (4I_J). The radiative lifetime varied with the increasing in active ion doping percentage because of the reciprocal interaction between the active ions themselves. In low doping percentage this effect will be neglected while in the high doping percentage the effect will appear clearly because of the increase in reciprocal interaction between the active ions that causes a concentration quenching for fluorescence wavelengths, due to the dynamical processes resulting from the interchange reaction among the active ions (ion-ion) interaction.

3-2 The Effect of the Host Medium Variation upon the Radiative Fluorescence

Transitions

The spectral parameter values for the four fluorescence transitions from the excited state ${}^4F_{3/2}$ to the lower states (4I_J) are influenced by varying the host material Silicate (L_1) and Borate (L_2, L_3) samples for the same active ion doping percentage. that could be noticed from the intensity parameter $(\Omega_{(i)})$ values. The fluorescence line strength intensity values $(S_{(f)})$ increase obviously in the transitions as shown below:

$${}^4F_{3/2} \rightarrow {}^4I_{9/2} \text{ (880 nm)}$$

${}^4F_{3/2} \rightarrow {}^4I_{11/2}$ (1064 nm) strongest transition

The fluorescence line strength value $(S_{(f)})$ increases according to the host material, where it is for Silicate (L_1) more than for Borate (L_2) at the same active ion doping percentage, this happened because of the difference in the physical density of these two samples. The spontaneous emission probabilities A_{rad} for fluorescence transitions are related to the fluorescence line strength transitions $(S_{(f)})$ which depends on the intensity parameters $(\Omega_{(i)})$ as shown in table 2. The same table illustrates the branching ratio coefficient b_r , values that depends on all the emission transition probability ratio for every transition which are determined from the excited state ${}^4F_{3/2}$ to the lower states (4I_J), it is generally larger for the transition ${}^4F_{3/2} \rightarrow {}^4I_{11/2}$ at (1064nm) wavelength as compared to other transitions, which can be used to measure the relative intensity of different laser transitions. The probability of transition to the levels ${}^4I_{13/2}, {}^4I_{15/2}$ is small as compared to other transitions. This agrees with the reference [8].

Again from table 2 it could be noticed that there is no relative variation in branching ratio values with the host material variation. The spontaneous emission probabilities A_{rad} vary in accordance with the host variation, the radiative lifetime (t_{rad}^c) dependence on the spontaneous emission probabilities A_{rad} as

shown in table 2, this is attributed to the change in the intensity parameter ($\Omega_{(t)}$) values with host variation, which reflects the local field effect of the host on the sites of active ions. It is observed that the decreasing of intensity parameter values ($\Omega_{(t)}$) especially in Ω_4, Ω_6

causes decrease in spontaneous emission probability values A_{rad} and as a result increase in the radiative lifetime (τ_{rad}^c) [9], it is more in Borate (L_2) than in Silicate (L_1).

Table (1)
Numerical values of square matrix elements $\left(|U^{(i)}|^2 \right)_{i=2,4,6}$ calculated for neodymium ion transitions from ${}^4F_{3/2}$ excited state [10].

Band	$\bar{\lambda}$ (nm)	State (s) [S' L' J']	Energy cm^{-1}	$ U^2 ^2$	$ U^4 ^2$	$ U^6 ^2$
1	880	${}^4I_{9/2}$	11030	.	0.230	0.056
2	1060	${}^4I_{11/2}$	9020	.	0.142	0.407
3	1300	${}^4I_{13/2}$	7020	.	0	0.212
4	1880	${}^4I_{15/2}$	5450	.	.	0.028

Note: $\langle U^i \rangle = \left| \left\langle ({}^4F_{3/2}) \| U^i \| (S' L' J') \right\rangle \right|^2$

Table (2)
Illustrate values of fluoresces line strength $S_{(f)}$, spontaneous emission probability values A_{rad} for ${}^4F_{3/2} \rightarrow {}^4I_3$, radiative lifetime τ_{rad}^c (msec) and calculated emission branching ratio coefficient b_r , $S_{(f)} * 10^{-20} cm^2 units$.

Sample symbol		L1			L2			L3		
${}^4F_{3/2} (S', L') J'$	λ (nm)	$S_{(f)}$	A_{rad}	b_r	$S_{(f)}$	A_{rad}	b_r	$S_{(f)}$	A_{rad}	b_r
${}^4I_{9/2}$	880	4.18	2329.3	0.52	3.82	2123.9	0.55	2.90	1614.6	0.69
${}^4I_{11/2}$	1060	5.80	1826.4	0.41	2.79	880.1	0.39	3.63	1140.7	0.29
${}^4I_{13/2}$	1300	1.83	282.2	0.06	0.25	38.5	0.055	1.04	160.7	0.013
${}^4I_{15/2}$	1880	0.24	13.8	0.003	0.03	1.8	0.002	0.14	7.86	0.0006
$A_{(total)} (sec^{-1})$		4451.8			2923.9			3044.3		
τ_{rad}^c (μsec)		224.6			342			328.5		
(J-O) Intensity parameters	Ω_2	1.2			0.13			0.48		
	Ω_4	16.1			16.31			11.42		
	Ω_6	8.64			1.18			4.92		

4- Conclusions

- 1- For the same host and different active ion doping percentage ($L_2=2\%$, $L_3=4\%$), the spectroscopic parameters increase linearly with the increase in active ion doping percentage which is agree with references [4].
- 2- The differences in values of intensity parameters are due to difference in host environments associated with ^{+3}Nd ion.
- 3- The main absorption band for the laser media especially for 530 nm and 810 nm wavelengths bands coincides with the emission spectral of the common diode laser to produce laser at 1064 nm wavelength which consider an important wavelength for different applications. In other words these materials are an efficient laser system that is pumped with powerful diode lasers whose pump wavelength overlaps the strong and broad ^{+3}Nd ion absorption band whose peaks are around 530 nm and 818 nm.

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الخلاصة

تم في هذا البحث تحديد افضل مجموعة لمعاملات الشدة ($\Omega_{(t)}$) وذلك باستخدام البرنامج المعايير (lest- square fitting) لغرض الحصول على قيم احتمالية الانبعاث التلقائي (A_{rad}) و العمر الزمني للاشعاع (t_{rad}^c (msec) ومعامل نسبة التفرغ الاشعاعي (b_r) للمستوي المتهيج، مع التركيز على حزم الامتصاص ضمن المديات الطيفية (530, 810 nm) للاوساط الليزرية الفعالة المستخدمة في البحث و التي تتطابق مع طيف الانبعاث القياسي للمصادر الليزرية نوع الدابود الليزري الشائعة الاستخدام في ضخ الليزر في الحالة الصلبة.