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# THE EFFECTS OF TEMPERATURE ON DIFFERENT LASER TRANSITIONS OF NEODYMIUM ORTHOVANADATE CRYSTAL

Ganesan Krishnan<sup>a,b\*</sup>, Noriah Bidin<sup>a,b</sup>

<sup>a</sup>Laser Center, Ibnu Sina Institute for Scientific and Industrial Research (ISI-SIR), Universiti Teknologi Malaysia, 81310 UTM Johor Bahru, Johor, Malaysia

<sup>b</sup>Department of Physics, Faculty of Science, Universiti Teknologi Malaysia, 81310 UTM Johor Bahru, Johor, Malaysia

# Graphical abstract



### Abstract

The temperature dependence of Nd:YVO<sub>4</sub> laser crystal pumped by laser diode emitting at 808 nm is studied within the range of 5 °C to 60 °C. The spectroscopy properties of quasi three level at 914 nm ( $4F_{3/2}$  - 4  $_{9/2}$ ) and four level at 1064 nm ( $4F_{3/2}$  - 4  $_{11/2}$ ) are characterized. The lineshape function of the transition lines were broadened as the temperature increases. The phenomenon is attributed to change in linewidth, lineshift and intensity. The linewidths for both laser transition of 914 nm and 1064 nm increases with temperature with the rate of 0.105 cm<sup>-1</sup>/°C and 0.074 cm<sup>-1</sup>/°C respectively. The peak of 914nm and 1064 nm lineshapes shifted to a longer wavelength with the rate of 3.0 pm/°C and 4.2 pm/°C respectively which correspond to same amount of lineshift. The lineshape broadening with respect to the temperature is due to one-phonon emission and Raman phonon scattering processes. The intensities of 914 nm and 1064 nm transition lines are found to be decreased at the rate of 0.15 %/°C and 0.45 %/°C respectively due to non-radiative effects. Quasi three level laser transition is more temperature dependent because it terminal level is close to the ground state which suffers from higher phonon-ion interaction rather than four level laser system.

Keywords: Temperature effects, spectroscopic, laser crystals

# Abstrak

Pergantungan suhu laser kristal Nd: YVO4 dipam oleh diod laser pada 808 nm dikaji dalam julat suhu 5 °C hingga 60 °C. Sifat-sifat spektroskopi laser aras separa tiga pada 914 nm (4F3  $_{/2}$  - 4  $_{3/2}$ ) dan aras empat pada 1064 nm (4F<sub>3/2</sub> - 411/2) telah dicirikan. Fungsi bentuk garis kedua-dua transaksi tersebut semakin meluaskan apabila suhu bertambah. Fenomena ini disebabkan oleh perubahan dalam lebar garis, peralihan garis dan keamatan. Lebar garis untuk kedua-dua transaksi laser pada 914 nm dan 1064 nm meningkat dengan suhu dengan kadar 0.105 cm<sup>-1</sup>/°C dan 0.074 cm<sup>-1</sup>/°C masing-masing. Puncak fungsi bentuk garis untuk transaksi 914 nm and 1064 nm beralih kepada panjang gelombang yang lebih panjang dengan kadar 3.0 pm/°C and 4.2 pm/°C masing-masing, ini juga menunjukkan nilai peralihan garis yang sama. Penambahan keluasan fungsi bentuk garis berkenaan dengan suhu adalah disebabkan oleh proses pelepasan satu fonon dan proses serakan fonon Raman. Keamatan setiap garis transaksi itu didapati menurun pada kadar 0.15 %/°C and 0.45 %/°C masing-masing disebabkan oleh kesan bukan radiasi. Transaksi laser aras separa tiga lebih bergantung kepada suhu kerana aras terminal terletak berhampiran dengan aras dasar yang mengalami interaksi fonon-ion yang lebih tinggi daripada laser aras empat.

Kata kunci: kesan suhu, spektroskopi, Kristal laser

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**Full Paper** 

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\*Corresponding author k.ganesan@utm.my

### **1.0 INTRODUCTION**

Neodymium ion doped laser crystals are extensively used in diode pumped solid state laser systems to produce outputs in near infrared and visible region of electromagnetic spectrum [1, 2]. Neodymium orthovanadate (Nd:YVO<sub>4</sub>) is an attractive laser crystal due to its high absorption cross-section and high emission cross-section at 1064 nm of  ${}^{4}F_{3/2}$  -  ${}^{4}I_{11/2}$ intermanifold transition. As the result, efficient lasers emission at 1064 nm and its second harmonic generation at 532 nm had been demonstrated [3-5]. In recent years, 914 nm emission of  ${}^{4}F_{3/2}$  -  ${}^{4}I_{9/2}$ intermanifold transition of Nd:YVO4 also gaining popularity due to its second harmonic generation yields blue laser which attracts attention for underwater communications, high-density optical storage and medical diagnostics [6]. The main disadvantages of Nd:YVO<sub>4</sub> crystal are short fluorescence lifetime and low thermal conductivity which is approximately half of Nd:YAG thermal conductivity [7]. As the result of low thermal conductivity, Nd:YVO4 suffers from thermal damage with high pumping power. It has been shown in previous works that the crystal temperature can increase beyond 100 °C during laser operation even with efficient diode pumping [8, 9]. Therefore, temperature effects of Nd:YVO<sub>4</sub> crystal on its physical and spectroscopic properties are crucial and desire greater attention in the architecture of diode pumped solid state laser system [10].

Previously, the significant effects of temperature on spectroscopic properties and laser performance of Nd:YVO4 have been reported [11, 12]. Amongst them are Mingxin et al. who claimed that a reduction in output power and a 4.7 pm/°C lineshift of 1064 nm emission by tuning the temperature of a microchip laser from 0 °C to 100 °C [13]. Sardar and Yow observed that thermal broadening and redshift of 1064 nm emission line when the temperature increased from -263 °C to 27 °C which due to phonon phenomenon [14]. Thermal broadening and lineshift affect the laser performance including laser gain, thermal tunability, output frequency stability and Q-switch modulation. Turri et al. investigated the emission cross section and peak emission wavelength of 1064 nm over temperature range of -263 °C to 77 °C and showed that a lineshift of 2 pm/°C for 1064 nm emission [15]. Recently, Delen et al. carried out similar work on 1064 nm emission cross section and peak emission wavelength by two complementary methods over typical laser operation temperature range [16].

Although intensive work have been performed regarding the temperature effects on linewidth and lineshift of fundamental beam of 1064 nm emission of Nd:YVO<sub>4</sub> crystal, but very little efforts have been focused on the spectroscopic properties for simultaneous dual-wavelength beam laser generation. Since the demonstration of such simultaneous dual-wavelength emission laser at 914 nm and 1064 nm by Pavel, it is desire to consider the effects of temperature on these radiations simultaneously [17].

In this present paper, the effects of temperature are demonstrated upon the major laser transition line of  ${}^{4}F_{3/2}{}^{-4}I_{11/2}$  and quasi three level  ${}^{4}F_{3/2}{}^{-4}I_{9/2}$  transitions. The spectroscopy properties including the intensities, linewidths and lineshifts for both emission lines are discussed in detail.

## 2.0 THEORY

The broadening and lineshift of sharp emission lines of Nd based laser crystals can be explained by phonon theory. The linewidth of the fluorescence emission is affected by several mechanisms including crystal strain inhomogeneity, direct one-phonon processes, multiphonon processes and Raman phonon scattering [18-20]. The width of *i*th energy level during ion-phonon interaction processes is expressed as:

$$\Delta v_i(cm^{-1}) = \Delta v_i^s + \Delta v_i^D + \Delta v_i^M + \Delta v_i^R \tag{1}$$

Where  $\Delta v_i^s$  is the crystal strain inhomogeneity,  $\Delta v_i^D$  is direct one-phonon processes between *i*th energy level and *j*th levels whereby the energy difference is smaller than available phonon energy,  $\Delta v_i^M$  is multiphonon processes and  $\Delta v_i^R$  Raman phonon scattering processes respectively.  $\Delta v_i^s$  is the linewidth of a inter-Stark transition at the lowest temperature and it is inhomogeneous broadening mechanism due to its random nature. Furthermore, it is independent of temperature. On the other hand,  $\Delta v_i^D$  consists of spontaneous one-phonon emission,  $\sum_{i < i} \beta_{ii}$ , which is temperature independent and a temperature-dependent part,  $\Delta v_i^D(T)$ .  $\Delta v_i^M$ is independent of temperature and can be ignored in the range of temperature used in this study [21]. Finally,  $\Delta v_i^R$  is temperature dependent linewidth broadening mechanism which caused by phonon scattering by impurity ions.

Therefore, in phonon phenomenon, the temperature-dependent linewidth of an emission line of neodymium ions can be written as [18, 19]:  $\Delta v(T)$ 

$$= \Delta v_{o} + \sum_{j < i} \overline{\beta}_{ij} \frac{1}{e^{\Delta E_{ij}/kT} - 1} + \sum_{j > i} \overline{\beta}_{ij} \frac{1}{e^{\Delta E_{ji}/kT} - 1}$$
$$+ \sum_{j < f} \overline{\beta}_{fj} \frac{1}{e^{\Delta E_{fj}/kT} - 1} + \sum_{j > f} \overline{\beta}_{fj} \frac{1}{e^{\Delta E_{jf}/kT} - 1} + \overline{\alpha} \left(\frac{T}{\theta_{D}}\right)^{7}$$
$$\times \int_{0}^{\theta_{D}/T} \frac{x^{6}e^{x}}{(e^{x} - 1)^{2}} dx \tag{2}$$

where  $\Delta v_o$  is the temperature independent residual linewidth of the transition between initial energy level *i* and terminal energy level *j*;  $\overline{\beta}_{ij}$  and  $\overline{\beta}_{fj}$  are the coupling coefficients for ion-phonon interaction;  $\Delta E_{ij}$  and  $\Delta E_{ji}$  are the difference in energy between *i* and *j* energy levels;  $\Delta E_{fj}$  and  $\Delta E_{jf}$  are the difference in energy between energy level *j* and intermediate energy level *f*;  $\overline{\alpha}$  is the coupling coefficient for the phonon-ion interaction;  $\theta_D$  is the effective Debye temperature; and  $x = \hbar \omega/kT$ .

For major line of  ${}^{4}F_{3/2}{}^{4}I_{11/2}$  transition at 1064 nm (R<sub>1</sub>-Y<sub>1</sub>), only phonon absorption processes are possible on both initial level (R<sub>1</sub>) and terminal level (Y<sub>1</sub>). Therefore, Eq. (2) can be simplified for 1064 nm emission line as shown below,

 $\Delta v_{1064}(T)$ 

$$= \Delta v_o + \overline{\beta}_{R_1 - R_2} \frac{1}{e^{\Delta E_{R_1 - R_2}/kT} - 1} + \sum_{j>1}^{\circ} \overline{\beta}_{Y_1 - Y_j} \frac{1}{e^{\Delta E_{Y_1 - Y_j}/kT} - 1} + \overline{\alpha} \left(\frac{T}{\theta_D}\right)^7 \times \int_0^{\theta_D/T} \frac{x^6 e^x}{(e^x - 1)^2} dx$$
(3)

As for 914 nm ( $R_2$ - $X_5$ ) emission line of  ${}^4F_{3/2}$ - ${}^4I_{9/2}$  transitions, only phonon emission processes are possible on both initial level ( $R_2$ ) and terminal level ( $X_5$ ). The temperature-dependent linewidth of the emission line can be simplified as

$$\begin{split} \Delta v_{914}(T) \\ &= \Delta v_{o} + \overline{\beta}_{R_{2}-R_{1}} \frac{1}{e^{\Delta E_{R_{2}-R_{1}}/kT} - 1} + \sum_{j<5}^{1} \overline{\beta}_{X_{5}-X_{j}} \frac{1}{e^{\Delta E_{X5-Xj}/kT} - 1} \\ &+ \overline{\alpha} \left(\frac{T}{\theta_{D}}\right)^{7} \times \int_{0}^{\theta_{D}/T} \frac{x^{6} e^{x}}{(e^{x} - 1)^{2}} dx \end{split}$$
(4)

Thermal lineshift of an interstark transition can be theoretically shown by assuming the lineshift is the algebraic sum shift of the initial and terminal levels. The thermal lineshift (in cm<sup>-1</sup>) of an emission line can be shown as [22];

$$\delta v(T) = \delta v_o + \alpha \left(\frac{T}{\theta_D}\right)^4 \int_0^{\theta_D/T} \frac{x^3}{e^x - 1} dx$$
(5)

where  $\delta v_o = v(0K) - v(10K)$ . v(0K) and v(10K) are the line positions at 0 K and 10 K respectively. The integrals in Eq. (2), Eq. (3), Eq. (4) and Eq. (5) can be found in tabulated form from references [23, 24].

The variation of intensity of an emission line with temperature can be determined by the linewidth of the emission at various temperatures. The peak emission intensity (photon per second) is given by [20],

$$I_p(T) = N_o I_a \sigma_a \tau_f c \phi \sigma_p \tag{6}$$

According to Füchtbauer–Ladenburg Equation, peak stimulated emission cross-section as shown below,

$$\sigma_p(T) = \frac{1}{8\pi} \frac{\lambda^4 \eta \beta}{n^2 c \tau_f} \frac{2}{\pi \Delta v(T)}$$
(7)

where  $N_o$  is the total number of Neodymium ions;  $I_a$ and  $\sigma_a$  are the pump beam intensity and absorption cross-section respectively;  $\tau_f$  is the fluorescence lifetime;  $\phi$  is the photon density;  $\eta$  is the quantum efficiency;  $\beta$  is the branching ratio; n is the refractive index;  $\Delta v$  is the emission linewidth.

# 3.0 METHODOLOGY

Figure 1 shows the schematic diagram of the experimental setup. An a-cut of 1 at % Nd:YVO4 with dimension of 3 mm x 3 mm x 2 mm is used as a gain medium. The Nd:YVO4 crystal was pumped to 4F5/2 level energy by a CW diode laser at 808 nm with output power of 0.45 W. The pump beam was focused using 808 nm AR coated lens with focal length of 25 mm into one end of the laser crystal. The diameter of beam at focal point is around 300 µm. The fluorescence output was measured perpendicular to the pump beam direction with an Ophir spectrum analyzer. This is to ensure that acquired fluorescence originated from uniformly pumped part of the crystal. The measurements were repeated five times. The Nd:YVO4 crystal was mounted onto a copper block. To enhance a smooth heat transfers a thermal interface material indium was employed. The assembly of the crystal holder was placed on a thermoelectric cooler (TEC). The temperature of the copper holder is controlled in the range of 0 °C to 60 °C. It is assumed that the Nd:YVO4 crystal and the copper holder are at thermal equilibrium during measurements.



Figure 1 Schematic of the experimental setup

### 4.0 RESULTS AND DISCUSSION

Fluorescence spectrums of 914 nm and 1064 nm emission in the range of temperature of 10 °C - 60 °C are shown in Figure 2 and Figure 3. Both emission lines are affected by temperature, consequently its lineshape functions broadened and peak output wavelengths redshifted. In contrast, the fluorescence intensities for both lines are significantly decreasing with temperature. The lineshape of 914 nm and 1064 nm emission lines are fitted by Lorentzian function. contribution of microscopic Hence strain, spontaneous one-phonon and multi-phonon emission processes to the observed linewidth broadening in Figure 2 and Figure 3 are negligible. Therefore, Raman scattering and one-phonon emission processes reveal Lorentzian lineshape. Both

mechanisms are responsible for the linewidth broadening. The wavelength shifts toward longer wavelength (redshift) in both inter-stark transitions are due to stationary effects of the phonon-ion interactions.



Figure 2 Fluorescence spectrum of quasi three level laser (914nm) at various temperatures



Figure 3 Fluorescence spectrum of four level emission (1064 nm) at various temperatures

The emission wavelength variation with temperature for both 914 nm and 1064 nm fluorescence are shown in Figure 4 and Figure 5 respectively. It was found that average shift in wavelength of 914 nm emission line is 3.0 pm/°C and 4.2 pm/°C for 1064 nm emission. Both laser transitions are observed to be shifted leading toward a longer wavelength or also known as red shifted. The red shift of 1064 nm emission is in good agreement with the previous researchers [7-9, 12]. However, due to the lack of references in laser transition of 914 nm, is difficult to make comparison. Even though wavelength shift of both transitions differ but lineshift of both transitions can be fitted by one theoretical line which is shown in Figure 6. In term of lineshift, these transitions shifted approximately 3.64 x 10<sup>-2</sup> cm<sup>-</sup> <sup>1</sup>/°C. Hereby, one can conclude that the algebraic sum of the shifts of the initial and terminal level of both emission lines are approximately the same. Through the theoretical fitting done, it was found that

the Debye temperature of these emissions is 400 K which will used in theoretical fitting of thermal broadening of linewidth.



Figure 4 Emission wavelength of quasi three level laser transition (914 nm)



Figure 5 Emission wavelength of four level laser transition (1064 nm)



Figure 6 Lineshifts of quasi three level and four level laser transitions

The broadening effect due to the change of temperature is quantified based on linewidth measurement. The results for both transition lines are presented in Figure 7. The experimental data and theoretical fitting trend of 914 nm and 1064 nm emissions is linear throughout investigated temperature range. The fitting parameters used are given in Table 1. The linewidths gradually increases with respect to the temperature. Apparently the linewidth of three level laser is observed to be greater than four level laser. The reason is due to the fact that the terminal level of guasi three level laser is positioned at the highest stark level of 419/2 manifold which is very close to the ground level. The thermal population in the ground level cause higher spontaneous one-phonon emission phenomenon in the terminal level which attributes to additional temperature independent linewidth broadened effects. In comparison to the four level laser at 1064 nm, the terminal level is the lowest stark level of  $4I_{11/2}$ which relatively far away from the ground level. Hence less potential for interaction of spontaneous one-phonon emission process to take place. In average, the rates of broadening of transition line of 914 nm and 1064 nm are 0.105 cm<sup>-1</sup>/°C and 0.074 cm-1/°C respectively. The linewidth broadening of quasi three level emission is inherently 2 times greater than four level transition. This indicates that quasi three level laser is strongly dependent on temperature parameter. The broadening effect is revealed from one-phonon emission as well as Raman phonon scattering processes.



Figure 7 Linewidth variation with temperature for 914 nm and 1064 nm emissions

 $\label{eq:table1} \begin{array}{l} \textbf{Table 1} \text{ fitting parameters for 914 nm and 1064 nm emission} \\ \textbf{linewidths} \end{array}$ 

	Fitting parameters			
Transitions	Line shift		Line width	
	θ <sub>D</sub> (K)	α (cm <sup>-1</sup> )	θ <sub>0</sub> (К)	α (cm <sup>-1</sup> )
914 nm (R <sub>2</sub> -X <sub>5</sub> )	400	48	400	138
1064 nm(R <sub>1</sub> -Y <sub>1</sub> )	400	48	400	98

The peak intensities of emission lines at 914 nm and 1064 nm with respect to temperature are shown in Figure 8 and Figure 9 respectively. The theoretical lines in Figure 8 and Figure 9 were drawn by assuming only linewidth variation occurs in the temperature range of this study while other parameters in Eq. (6) and Eq. (7) are assumed invariables. In general, both transition lines experience decreasing in intensity with temperature. The decreasing rate is greater for emission line of 1064 nm at the rate of 0.45 %/°C in comparison to line 914 nm which only 0.15 %/°C. The higher reduction of intensity in 1064 nm emission is due to depletion of  $R_1$  level of  ${}^4\!F_{3/2}$  manifold which is upper laser level of the emission according to the Boltzmann population distribution. In contrast, the initial level of 914 nm emission (R<sub>2</sub>) population increases at higher temperature. The decreasing trend of peak intensities for both emission lines upon temperature is also indicate higher rate of nonradiative transitions occur at higher temperature.



Figure 8 Intensity of 914 nm line as a function of temperature



Figure 9 Intensity of 1064 nm line as a function of temperature

As summarization, the change of temperature affected the emission line of laser transition in both quasi three level laser and four level laser transition. A stronger effect of temperature is found in quasi three level laser rather than four level laser. As mention earlier, the position of the terminal level in the interstark of manifold attribute to effect of the spectroscopic properties. Due to the existing of thermal population in quasi three level transition reveals to the effect of one-phonon emission and Raman phonon scattering effects. Thus, ion-phonon interaction and the Raman phonon scattering effect attributable to broadening of linewidth, lineshift and decreasing intensity effects on the quasi three level laser transition spectrum. All these mention mechanisms are linearly increase with the temperature range of this study.

# 5.0 CONCLUSION

Laser transition of Nd:YVO4 at guasi three level laser and four level laser are investigated with regard to At higher operation temperature, temperature. detrimental effects on spectroscopy properties for both transitions are realized. This is quantified based on linewidth, lineshift and intensity of the gain spectrum. In general, the transition lines undergo linewidth broadening, red lineshifted and intensity reduction with increase in temperature. The mechanisms responsible to such effects are due to the one-phonon emission, Raman phonon scattering, depletion process at the upper level and non-radiative phenomenon. Overall, the effect of temperature on quasi three level laser is greater than the four level laser transition. This is due to its terminal position which closer to the ground state.

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