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Shift and broadening of emission lines in Nd^{3+}:YAG laser crystal influenced by input energy

SEYED EBRAHIM POURMAND[1],* and GHASEM REZAEI[2]
[1]Department of Optics and Laser Engineering, Estahban Branch, Islamic Azad University, Estahban, Iran
[2]Department of Physics, College of Sciences, Yasouj University, Yasouj, 75914-353, Iran
*Corresponding author. E-mail: se.pourmand@gmail.com

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Abstract. Spectroscopic properties of the flashlamp-pumped Nd^{3+}:YAG laser as a function of input energy were studied over the range of 18–75 J. The spectral widths and shifts of quasi-three-level and four-level inter-Stark emissions within the respective intermanifold transitions of ${}^4F_{3/2} \rightarrow {}^4I_{9/2}$ and ${}^4F_{3/2} \rightarrow {}^4I_{11/2}$ were investigated. The emission lines of ${}^4F_{3/2} \rightarrow {}^4I_{9/2}$ shifted towards longer wavelength (red shift) and broadened, while the positions and linewidths of the ${}^4F_{3/2} \rightarrow {}^4I_{11/2}$ transition lines remained constant by increasing the pumping energy. This is attributed to the thermal population as well as one-phonon and multiphonon emission processes in the ground state. This phenomenon degrades the output performance of the lasers.

Keywords. Nd^{3+}:YAG crystal; heat generation; three-level emission lines; four-level emission lines; input energy.

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1. Introduction

Trivalent neodymium ion (Nd^{3+}), as a dopant in the yttrium aluminum garnet (YAG), is the most practical and popular activator laser host having optical, thermal and mechanical properties suitable for laser mediums. Although Nd^{3+}:YAG has good thermal conductivity and optical transparency over a wide spectral region, pumping energy and non-radiative processes cause heat generation in the gain medium. However, the heat generated leads to shift and thermal broadening of laser lines which seriously affect the slope efficiency, laser threshold power and frequency stability of the lasers and obstruct the lasing performance [1–4].

Modelling of high-power solid-state lasers requires precise knowledge of temperature and pumping energy dependency of spectral linewidth and line broadening to ensure a good control on the effects of the generated heat in the gain medium.

Following the classical paper by McCumber and Sturge [5], many research works have been carried out on thermally-induced line broadening in trivalent rare-earth ions doped in different crystal hosts [4,6–10]. Kushida [11] measured linewidth and thermal shifts of several lines in Nd^{3+}:YAG crystal. Similar work was also reported by Xing group [12]. They have made considerable efforts on the shifts of all the spectral lines of intermanifold $^4F_{3/2}$ to $^4I_{11/2}$. Furthermore, they also studied the shift changes between the Stark sublevels R_1 and R_2 of Nd^{3+}:YAG crystal as a function of temperature over 20–200°C range.

However, most of the previous researchers have studied the temperature dependency of the spectroscopic properties of the laser hosts. To the best of our knowledge, input energy dependency of the shift and broadening of emission lines of Nd^3:YAG laser host has not been reported up to now. Therefore, in this paper, we report the experimental evidence that linewidths and lineshifts of quasi-three-level transition lines of Nd^{3+}:YAG crystal are affected by the input energy.

2. Theory

A famed equation for the temperature dependence of the ith energy level width is given by [4,8,9]

$$\Delta E_i \left(\mathrm{cm}^{-1}\right) = \Delta E_i^{\mathrm{strain}} + \Delta E_i^{\mathrm{D}} + \Delta E_i^{\mathrm{M}} + \Delta E_i^{\mathrm{R}}, \tag{1}$$

where the terms on the right-hand side of the equation represent, respectively, the broadenings due to the crystal strain inhomogeneity $\left(\Delta E_i^{\mathrm{strain}}\right)$, direct one-phonon processes $\left(\Delta E_i^{\mathrm{D}}\right)$ between the ith energy level and other nearby levels (j), multiphonon emission processes $\left(\Delta E_i^{\mathrm{M}}\right)$ which occur between two levels whose energy difference is greater than the greatest energy of the available phonons, shown to be essentially temperature independent, and the Raman phonon scattering processes $\left(\Delta E_i^{\mathrm{R}}\right)$ which are temperature-dependent and related to phonon scattering by the impurity ions.

The direct one-phonon broadening processes comprise a temperature-dependent part, ΔE_i^{DT}, and a temperature-independent part, $\sum_{j<i} \beta_{ij}$. The temperature-independent part is due to spontaneous one-phonon emission, and as the energy separation between the Stark levels is in the order of 10–10^2 cm^{-1} for rare-earth ions, $\sum_{j<i} \beta_{ij}$ can cause a visible broadening.

Equation (1) can be written as

$$\Delta E_i(T) = \Delta E_i^{\mathrm{strain}} + \sum_{j<i} \beta_{ij} + \Delta E_i^{\mathrm{D}}(T) + \Delta E_i^{\mathrm{M}} + \Delta E_i^{\mathrm{R}}(T)\,. \tag{2}$$

Inhomogeneous broadening due to crystal strains gives rise to Gaussian transition lineshape, while one-phonon emission, multiphonon emission and the Raman scattering process give rise to homogeneous broadening with Lorentzian transition lineshape.

3. Experimental method

The schematic diagram of the experimental set-up is shown in figure 1. The system comprises two parts. The first part energizes a laser crystal and stabilizes the stimulating emission of fluorescence radiation. The second part is a measurement equipment which comprises a spectrometer.

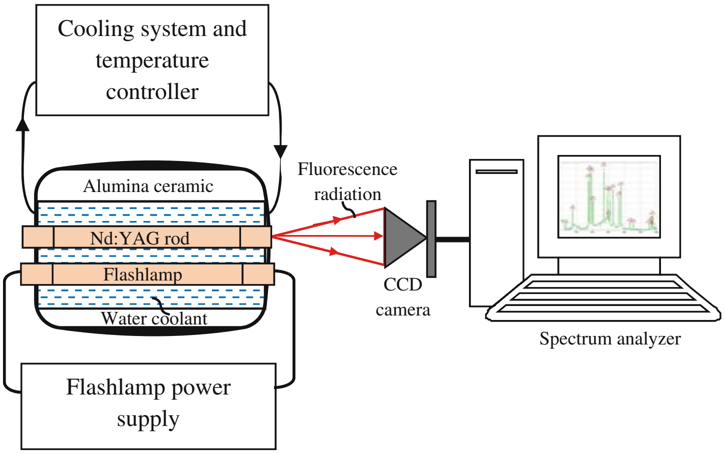

Figure 1. Schematic diagram of the experimental set-up.

A commercial laser rod Nd^{3+}:YAG laser crystal was utilized as the gain medium. The doping level of the laser rod was 1 at%. The rod was of 4 mm in diameter and 70 mm in length and was enclosed in a ceramic reflector.

The laser rod was placed parallel to a linear flashlamp-filled xenon gas at 450 Torr. The flashlamp was pumped by a homemade power supply [13]. The driver was triggered by simmer mode technique. A capacitor bank with 150 μF capacitance was charged by a maximum voltage of 1000 V and thus the input energy was varied between 5 and 75 J.

The Nd:YAG crystal together with the flashlamp was flooded with distilled water which acts as a coolant. The flashlamp was further enclosed with a samarium flow tube to absorb UV light radiation and to keep the flow rate in a steady-state condition. The fluorescence radiation, after pumping, was emitted at one end of the laser rod. The light was detected by a CCD camera placed 20 cm from the source. The emission spectrum was analysed using a Wavestar version 1.05 software. The resolution of this detection system was 0.5 nm and it could resolve most of the transition lines appeared from the pumping rod.

4. Results and discussion

The normalized fluorescence spectra of the main lines of the manifold transitions $^4F_{3/2}\rightarrow{}^4I_{9/2}$ (quasi-three-level system) and $^4F_{3/2}\rightarrow{}^4I_{11/2}$ (four-level system) in Nd^{3+}:YAG are shown in figure 2. The recorded spectrum was used to investigate the input energy dependency of the lineshifts and linewidths of the main quasi-three-level transition lines of $R_1\rightarrow Z_5$ (946.0 nm), $R_2\rightarrow Z_5$ (938.5 nm) and four-level transition lines of $R_2\rightarrow Y_1$ (1051.9 nm), $R_1\rightarrow Y_1$ (1061.3 nm), $R_2\rightarrow Y_3$ (1063.9 nm), $R_2\rightarrow Y_4$ (1067.6 nm), $R_1\rightarrow Y_3$ (1073.3 nm) and $R_1\rightarrow Y_4$ (1077.5 nm).

The input energy dependency of lineshift of transition lines at input energies from 18 to 75 J is shown in figure 3. The experimental results show that the positions of four-level system emission lines of Nd^{3+}:YAG crystal remained constant with input energy. Meanwhile, the quasi-three-level emission lines were apparently input energy-dependent. The

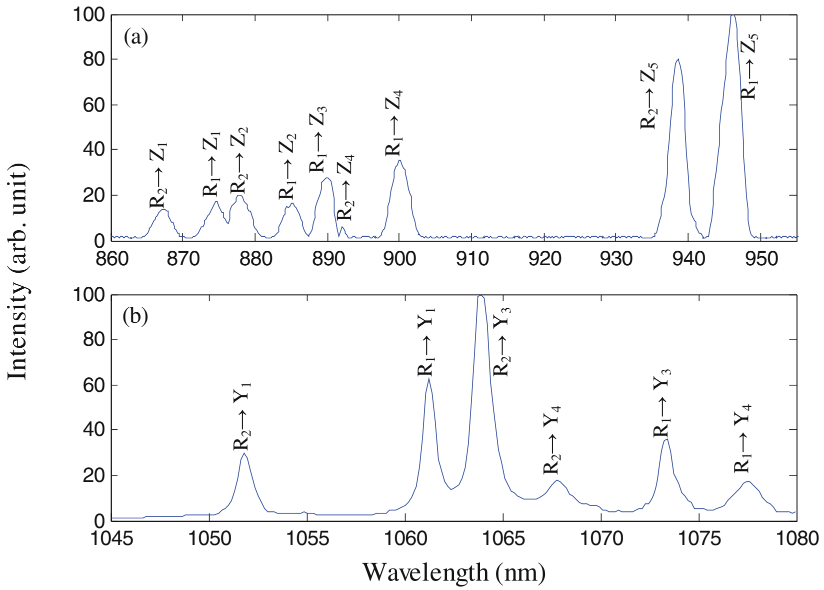

Figure 2. Normalized fluorescent emission spectra for (**a**) quasi-three-level and (**b**) four-level transitions of Nd^{+3}:YAG.

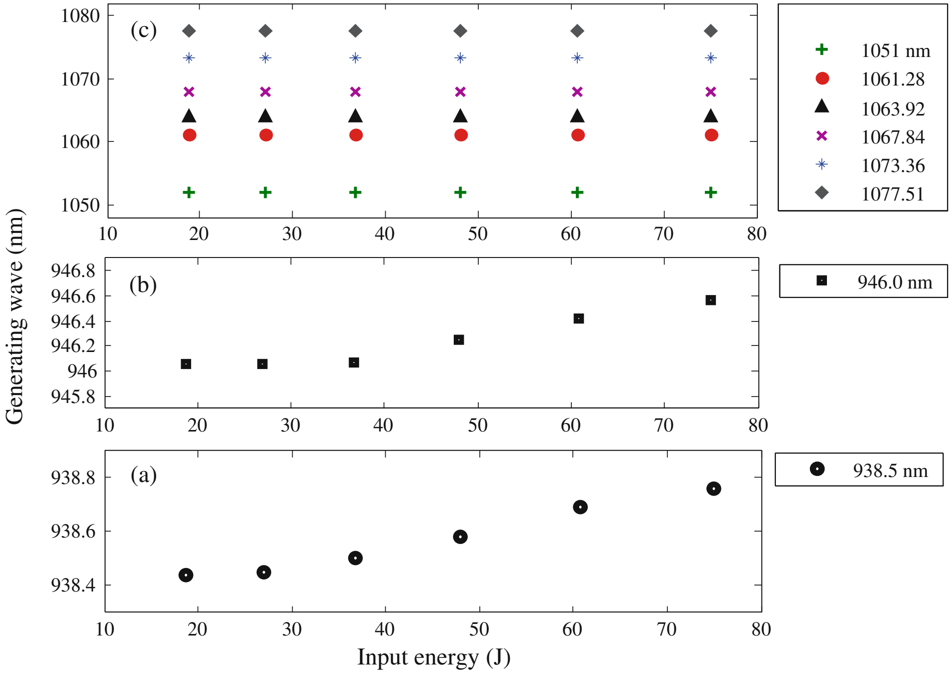

Figure 3. Input energy dependence of (**a**) line of 938.5 nm, (**b**) line of 946 nm and (**c**) several four-level emission lines.

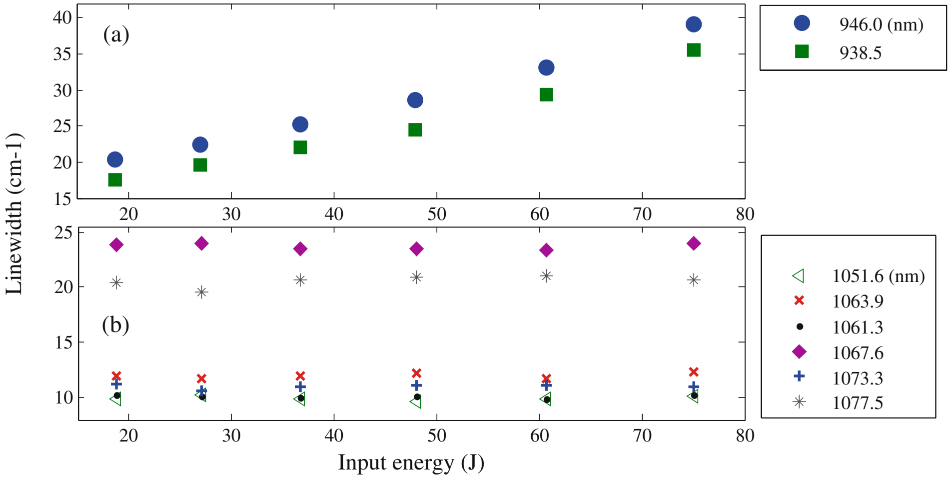

Figure 4. Linewidth of (**a**) 938.5 and 946 nm lines and (**b**) several four-level system lines vs. input energy.

938.5 and 946.0 nm lines in intermanifold $^4F_{3/2}\rightarrow{}^4I_{9/2}$ shifted to longer wavelength (red shift), and as a result, energy level of Stark sublevels of Z_5 and Z_4 moved upward. Figure 3 shows that the main emission lines of quasi-three-level system shifted from 938.44 and 946.06 nm at 18 J to 938.76 and 946.57 nm at 75 J, respectively.

Figure 4 indicates the linewidths dependency of 938.5 and 946.0 nm emission lines along with 1051.9, 1061.3, 1063.9, 1067.6, 1073.3 and 1077.5 nm emission lines with input energies from 18 to 75 J. The linewidths of transition lines of intermanifold $^4F_{3/2}\rightarrow{}^4I_{11/2}$ was found to remain constant with input energy. In the case of quasi-three-level system lines, as the input energy increased, the spectral linewidths increased by 17.97 and 18.81 cm^{-1} for the $R_1\rightarrow X_5$ and $R_2\rightarrow X_5$ transitions, respectively. As there were no changes of linewidth and position at four-level system transition lines, it could be assumed that there were no ion–phonon interactions, such as direct one-phonon processes $R_1\rightarrow R_2$ and $Y_1\rightarrow Y_2$, $Y_3 \ldots Y_6$ and multiphonon emission processes from Stark levels R_1 and R_2 to $^4F_{3/2}$ manifold by increasing the input energy.

As the shape of the spectral lines of the 938.5 and 946.0 nm could be fitted by Lorentzian at room temperature, the line broadening arising from the crystal strain inhomogeneity could therefore be neglected. In addition, the present work was done at a constant temperature of 20°C. Thus only input energy dependence of linewidth and position of transition lines were considered. Therefore, Raman scattering and one-phonon temperature-dependent part were neglected. Hence, the thermal broadening and linewidth of the emission lines were mainly related to the temperature-independent part of the direct one-phonon and multiphonon emission processes.

5. Conclusion

The influence of input energy on broadening and position of sharp spectral lines of the inter-Stark transitions of $^4F_{3/2}\rightarrow{}^4I_{9/2}$ and $^4F_{3/2}\rightarrow{}^4I_{11/2}$ in Nd^{3+}:YAG crystal were investigated. The wavelengths of the quasi-three-level system of 938.5 and 946.0 nm transition lines were shifted toward longer wavelength (red shift), while the wavelengths of the four-level system transition lines remained constant with input energy. In addition, the linewidths of $^4F_{3/2}\rightarrow{}^4I_{9/2}$ transition lines increased, while the linewidths of $^4F_{3/2}\rightarrow{}^4I_{11/2}$ transition lines were almost constant with increasing input energy. The input energy dependence of linewidths and positions of quasi-three-level transition lines are due to thermal population, one-phonon and multiphonon processes in the ground level. This new observation may contribute to the development of tunable lasing systems.

Acknowledgements

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