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Non-radiative transitions between the sublevels of the working lower multiplet of the YAG:Nd laser: Effects on the generation spectrum ☆

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Abstract

The article presents the stationary solution of the Tang–Statz–DeMars system of equations describing the generation spectrum of a YAG:Nd laser at a wavelength of 1064.2 nm taking into account the seven ${}^4F_{3/2} \rightarrow {}^4I_{11/2}$ transition gain lines. The effect that line broadening has on the generation spectrum due to a temperature increase has been calculated. The effect of population distribution over the ${}^4I_{11/2}$ multiplet sublevels on the generation spectrum was also calculated. The generation spectrum shifting to the region of 1066 nm with a temperature increase was found to result from variations in the population of the ${}^4I_{11/2}$ multiplet sublevels.

Keywords: YAG:Nd laser; Generation spectrum; Nonradiative transition; Population distribution; Tang–Statz–Demars system of equations.

1. Introduction

The generation spectrum of a neodymium-doped yttrium aluminium garnet (YAG:Nd) laser at a wavelength of 1064.2 nm is known to widen and shift to longer wavelengths with a temperature increase by the following values: [1]:

$$d(\nu)/dT = 5 \times 10^{-2}\ \mathrm{cm}^{-1}\ \mathrm{K}^{-1};$$
$$d(\Delta\nu)/dT = 1.8 \times 10^{-2}\ \mathrm{cm}^{-1}\ \mathrm{K}^{-1}.$$

The broadening of the generation spectrum is explained by thermal vibrations of the crystal lattice and neodymium ions. However, a shift in the maximum of the generation spectrum to the wavelength of 1066 nm with an increase in the temperature of a YAG crystal cannot be explained solely through a model of YAG:Nd laser generation that takes into account three gain lines of 1064.40, 1064.15 and 1061.50 nm.

In literature, the influence of the YAG:Nd crystal temperature on the gain of the laser medium has been analyzed based on the classical balance equations [2] describing the thermal population of the ${}^4I_{11/2}$ multiplet from the lower level. With these equations, however, it is difficult to account for the impact of several gain lines and several upper and lower sublevels on the generation spectrum.

The laser generation spectrum is calculated using the system of Tang–Statz–DeMars equations which does account for several gain lines [3], the degree of broadening and the number of generated axial frequencies [4]. It is impossible to account for the effects of the Boltzmann population and the structure of

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Table 1
The partial structure of the energy levels and the $^4F_{3/2} \rightarrow {}^4I_{11/2}$ transitions of a YAG:Nd laser at 300 K.

Transition number	λ (nm)	Lower sublevel energy (cm^{-1})	Upper sublevel energy (cm^{-1})	$\sigma \cdot 10^{19}$ (cm^2)
1	1052.10	2028	11 507 (*A*)	2.7
2	1061.50	2002	11 423 (*B*)	4.7
3	1064.15	2110	11 507 (*A*)	7.1
4	1064.40	2028	11 423 (*B*)	1.9
5	1068.20	2146	11 507 (*A*)	1.8
6	1073.70	2110	11 423 (*B*)	2.6
7	1077.9	2146	11 423 (*B*)	1.2

Notations: λ is the transition wavelength (the center of a gain line) of a neodymium ion a YAG crystal; σ is the cross section of the electron transitions between two sublevels of the $^4F_{3/2}$ and $^4I_{11/2}$multiplets; (*A*) and (*B*) are the energies of two sublevels of the $^4F_{3/2}$ multiplet.

sublevels of the lower $^4I_{11/2}$ multiplet in the system in question, as the equations use only the population inversion.

It follows from the above that there is currently no theoretical explanation of the influence of the temperature on the generation spectrum of a YAG:Nd laser.

The purpose of the present article is to demonstrate the effects that the deviation of the population of $^4I_{11/2}$ sublevels from the thermal equilibrium has on the laser generation spectrum. The solution of the Tang–Statz–DeMars equations for all possible gain lines of the $^4F_{3/2} \rightarrow {}^4I_{11/2}$ laser transition is given.

2. The theoretical basis

It is known that a neodymium ion in a YAG crystal lattice on the $^4F_{3/2} \rightarrow {}^4I_{11/2}$ transition has seven gain lines; laser generation is possible for two of them (1064.15–1061.50 nm). The additional lines that do not participate in the generation can affect the laser's emission spectrum [5].

To analyze the generation spectrum of a YAG:Nd laser with the seven gain lines taken into account we used the $^4F_{3/2} \rightarrow {}^4I_{11/2}$ transitions [5] listed in Table 1.

Similar to the description in [6], let us put down the Tang–Statz–DeMars system of equations for seven gain lines of the laser based on the data of Table 1:

$$\frac{dm_k}{d\tau} = Gm_k\left[L_k^{(A)}(n_0+n_k)+L_k^{(B)}\left(n'_0+n'_k\right)-1-\beta_k\right];$$

$$\frac{dn_0}{d\tau} = A - n_0\left(1+\sum_{m=1}^{M} L_m^{(A)} m_m\right) - \sum_{m=1}^{M} L_m^{(A)} m_m n_m;$$

$$\frac{dn_k}{d\tau} = -n_\kappa\left(1+\sum_{m=1}^{M} L_m^{(A)} m_m\right) - \frac{1}{2} L_k^{(A)} m_k n_0;$$

$$\frac{dn'_0}{d\tau} = A' - n'_0\left(1+\sum_{m=1}^{M} L_m^{(B)} m_m\right) - \sum_{m=1}^{M} L_m^{(B)} m_m n'_m;$$

$$\frac{dn'_k}{d\tau} = -n'_\kappa\left(1+\sum_{m=1}^{M} L_m^{(B)} m_m\right) - \frac{1}{2} L_k^{(B)} m_k n'_0;$$

$$w = \frac{n_0}{n'_0} = \exp\left(\frac{E_B - E_A}{k_B T}\right);$$

$$L_k^{(A)} = L_k^{(1)} + L_k^{(3)} + L_k^{(5)},$$

$$L_k^{(B)} = L_k^{(2)} + L_k^{(4)} + L_k^{(6)} + L_k^{(7)} \quad (1)$$

where m_k is the intensity of the kth generated axial mode, n_0 and n_k are the spatially homogeneous inversion and its lattices at the transition from the upper working sublevel (11,507 cm^{-1}), n'_0 and n'_k are the spatially homogeneous inversion and its lattices at the transition from the upper working sublevel (11,423 cm^{-1}), M is the number of axial modes, $G = 2k/\gamma_{||}$, $\tau = t/\gamma_{||}$ ($\gamma_{||}$ and k are the relaxation velocities of the population inversion and the field in the resonator) β_k is the loss of the kth axial mode, A and A' are the pumping parameters at the upper sublevels, $L_k^{(i)}$ are the Lorenzian gain lines of the ith component normalized to the cross-section of a strong transition defined by the expression:

$$L_k^{(i)} = f_k(n)\frac{\sigma_i}{\sigma}\left[1+((p-k)\Delta_0+\Delta_i)^2\right]^{-1},$$

$$\Delta_i = \frac{\omega_0^{(i)} - \omega_0}{\gamma_\perp},$$

$f_k(n)$ is the function that takes into account the influence of the uniform gain on the spectrum generation [4], equal to

$$f_k(n) = \prod_{m=1}^{n}\left|\cos\left(\frac{\pi}{2m}k\right)\right|, \quad (2)$$

(n is the number of adjacent axial modes not involved in laser generation); ω_0 is the center of a strong gain line at the wavelength of 1064.15 nm, $\omega_0^{(i)}$ is the ith gain line center, σ_i is section of the ith gain line transition, Δ_0 is the intermode interval set by the length of the resonator, $\gamma_\perp$ is the polarization relaxation rate (equal to half of the width of a homogeneous gain line); E_A and E_B are the energies of the Stark sublevels, k_B is the Boltzmann constant.

The stationary solution of the system of equations (1) has the form:

$$n'_0 = \frac{1}{1+w}\left(A + A' - (1+\beta_k)\sum_{m=1}^{M} m_m\right),$$

$$n_0 = \frac{w}{1+w}\left(A + A' - (1+\beta_k)\sum_{m=1}^{M} m_m\right),$$

$$n_k = \left[\frac{(1+\beta_k) - L_k^{(B)} n'_0}{L_k^{(A)}} - n_0 \right] \left[F_2 + F_1 \frac{L_k^{(B)}}{L_k^{(A)}} \right]^{-1},$$

$$n'_k = \frac{(1+\beta_k) - L_k^{(B)} (n'_0 + n'_k)}{L_k^{(A)}} - n_0,$$

$$m_k = \frac{n_k \left(1 + \sum_{m=1}^{M} L_m^{(A)} m_m \right)}{-0.5 L_k^{(A)} n_0}, \qquad (3)$$

where

$$F_1 = n'_0 L_k^{(B)} \left(1 + \sum_{m=1}^{M} L_m^{(A)} m_m \right),$$

$$F_2 = n_0 L_k^{(A)} \left(1 + \sum_{m=1}^{M} L_k^{(B)} m_m \right).$$

Since the intermode interval $\Delta_0 = 0.05$, for calculation convenience, similarly to the supposition made in [6], let us assume that the axial mode of the resonator corresponding to the maximum gain at the 1064.15 nm wavelength is number 70. Then the modes corresponding to the maximum gains at the 1052.10, 1061.50, 1064.40, 1068.20, 1073.70, and 1077.90 nm wavelengths are number 746, 222, 56, –156, –464 and –699.

To account for the influence of the homogeneous broadening for a 30 mm-long resonator, all subsequent calculations will assume n equals 2 in Eq. (2) when generation occurs at four axial modes.

To account for the inhomogeneous broadening of the laser generation spectrum with an increase in the YAG crystal temperature, the Lorentzian profiles (2) need to be widened using the $s(T)$-function of inhomogeneous gain line broadening as follows:

$$L_k^{(i)} = \frac{s(T)\sigma_i}{\sigma} \left[s(T) + ((p-k)\Delta_0 + \Delta_i)^2 \right]^{-1}. \qquad (4)$$

The calculations showed that the following changes occur with an increase in inhomogeneous gain line broadening associated with a temperature increase:

the generation spectrum is broadening, which is consistent with the known data [1];

the maximum of the generation spectrum is shifting towards the short-wavelength region of the spectrum, which contradicts the known data predicting there must be a shift to longer wavelengths.

The shift to shorter wavelengths that occurs in the generation spectrum with a temperature increase is due to the proximity of the 1061.5 nm gain line. As the Tang–Statz–DeMars system of equations (1) implies that the lower level is not populated, the calculations were performed under the condition that the lower level (the $^4I_{11/2}$ multiplet) is cleared instantly.

It follows from calculations in the system of equations (3) factoring in Eq. (4) that the shift to longer wavelengths with gain line broadening is possible only when the transition cross-sections at the 1061.5 and 1064.4 nm wavelengths tend to zero. As seen in Table 1, the laser transitions with the 1061.50 and 1064.40 nm wavelengths occur between the $^4I_{11/2}$ multiplet sublevels with the energies of 2002 and 2028 cm^{-1}, respectively. The laser transitions with the 1064.15 and 1068.20 nm wavelengths are to higher sublevels of the $^4I_{11/2}$ multiplet with the energies of 2110 and 2146 cm^{-1}, respectively.

Accepting that the population of the $^4I_{11/2}$ multiplet sublevels follows the Boltzmann distribution, the possible difference between the populations of the $^4I_{11/2}$ multiplet sublevels does not exceed 25%. Therefore, the calculations do not confirm the Boltzmann distribution of the $^4I_{11/2}$ multiplet populations.

Balance equations were composed to determine the population distribution of the $^4I_{11/2}$ multiplet sublevels. The following assumptions were made to simplify the calculations:

– the sublevels of the $^4I_{11/2}$ multiplet with the energies of 2002 and 2028 cm^{-1} have been combined into one lower sublevel as they are very close;
– since the energy difference between the $^4I_{11/2}$ multiplet sublevels is much less than that between the $^4I_{11/2}$ and the $^4I_{9/2}$ multiplets, the rate of relaxation transitions from the sublevels of the $^4I_{11/2}$ multiplet to the $^4I_{9/2}$ multiplet are equal (w_{20}), and the thermal occupancy of the $^4I_{11/2}$ multiplet sublevels starting from the lower level $^4I_{9/2}$ is also equal (w_{02}) for all sublevels;
– the thermal relaxation transitions between the sublevels of the $^4I_{11/2}$ multiplet are equal (w), since the energy difference between the sublevel populations of the multiplet does not exceed 25%.

$$\frac{dn_1}{dt} = -(2w + w_{20} + w_f(\Delta E_{12}, T) + w_f(\Delta E_{13}, T))n_1 + w(n_2 + n_3) + w_{02};$$

$$\frac{dn_2}{dt} = A - (2w + w_{20} + w_f(\Delta E_{23}, T))n_2 + w_{f1} n_1 + w(n_1 + n_3) + w_{02};$$

$$\frac{dn_3}{dt} = -(2w + w_{20})n_3 + (w + w_{f1} + w_{f2})n_1 + (w + w_{f1})n_2 + w_{02},$$

where n_1, n_2 and n_3 are the populations of the sublevels of the $^4I_{11/2}$ multiplet with the energies of 2146, 2110 and 2028 cm^{-1}, respectively; $w_f(\Delta E_{ij}, T)$ are the rates of relaxation transitions between the i and j sublevels caused by the phonon–electron influence of the crystal lattice on neodymium ions.

The probability of a multi-phonon transition is described [7] by the following dependence on the crystal temperature T and the energy gap ΔE:

$$w(\Delta E, T) = [1 - \exp(-\hbar\omega/kT)]^{-p} [C^{(p)} \exp(-\alpha \Delta E)], \quad (5)$$

where p is the number of phonons resulting from the phonon–electron interaction; $C^{(p)}$ and α are constants characterizing the basis, in this case, the YAG crystal.

It is apparent from (5) that the probability of a relaxation transition is exponentially dependent on the energy gap. Therefore, the probability of a phonon–electron transition between sublevels 1 (2146 cm^{-1}) and 2 (2110 cm^{-1}) is more than the probability of a transition between sublevels 1 and 3 (2028 cm^{-1}), and also 2 and 3. We assumed for the calculations that

$$w_f(\Delta E_{12}, T) > w_f(\Delta E_{13}, T) \approx w_f(\Delta E_{23}, T).$$

Since the lifetime of the $^4I_{11/2}$ multiplet is approximately 10^{-8}s and the time of the phonon–electron transition $\approx 10^{-9}$–10^{-10} s, the dependence we used was

$$w_f = 10 w_{20}.$$

The calculation results showed that the relative population inversion of the multiplet sublevels has the following values:

$$n_1 = 0.062, n_2 = 0.336, n_3 = 0.601.$$

It was found that this distribution is strongly dependent on the w_f/w and w_f/w_{20} ratios.

The population of the $^4I_{11/2}$ multiplet sublevels with the energy of 2110 cm^{-1} depends on the rate of thermal relaxation transitions providing the Boltzmann distribution (Fig. 1a). The calculations showed that in order to adequately model the changes of the emission spectrum from the broadening of gain lines, it is necessary that the relative population of sublevel 2 was not less than 0.3 under heating. We may thus conclude that the phonon–electron relaxation rate in an activated crystal must be higher by at least an order of magnitude than the thermal equilibrium rate in accordance with the Boltzmann distribution.

Fig. 1b shows that the population of the upper sublevel of the $^4I_{11/2}$ multiplet with the energy of 2110 cm^{-1} does not depend on the rate of relaxation transitions relaxation to the main $^4I_{9/2}$ level.

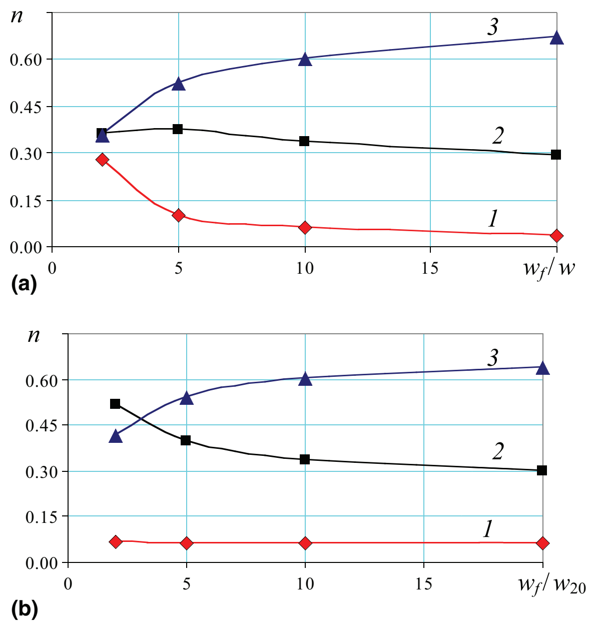

Fig. 1. The dependences of the relative populations of sublevels 1–3 of the $^4I_{11/2}$ multiplet on the w_f/w (a) and w_f/w_{20} (b) ratios.

The calculations proved that in order to adequately model the changes of the emission spectrum from the broadening of gain lines, under heating the relative population of sublevel 2 must be lower than that of sublevel 3. This leads us to conclude that the phonon–electron relaxation rate must be an order of magnitude higher than the rate of relaxation transitions to the main $^4I_{9/2}$ level.

The Boltzmann thermal population of the $^4I_{11/2}$ multiplet to the main $^4I_{9/2}$ level has virtually no effect on the population distribution of the multiplet sublevels, and, with a temperature increase, leads to a proportional population growth and subsequent population saturation of the multiplet.

3. Conclusion

The theoretical analysis demonstrated that the deviation of the population of $^4I_{11/2}$ sublevels from the thermal equilibrium affects the laser generation spectrum. It is evident from the solution of balance equations describing the population of the $^4I_{11/2}$ multiplet sublevels that the population of the lower sublevels grows with an increase in temperature, with the 2146 cm^{-1} sublevel remaining unpopulated. This ultimately leads to the generation spectrum shifting to the long-wave frequency region with a temperature increase, due to the influence that the gain line with a 1068.2-nm wavelength has on the generation spectrum.

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