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Spectroscopic studies of the population of high-energy levels of Nd^{3+}-doped laser crystals upon intense pumping

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***Abstract*. The luminescence of Nd : YAG and Nd : YAP crystals excited by intense 808-nm radiation from laser diodes and the second (532 nm), third (354.7 nm), and fourth (266 nm) harmonics of a pulsed Nd : YAG laser is studied in the range from 380 to 650 nm. A number of intense luminescence lines are assigned to the transitions from the high $^2F(2)_{5/2}$ level. A channel of the efficient population of this level upon simultaneous pumping a Nd : YAG crystal by a diode laser and the second harmonic of the Nd : YAG laser is found.**

Keywords: *intense pumping, absorption from an excited state, luminescence spectra of Nd^{3+}.*

1. Introduction

Laser crystals doped with Nd^{3+} ions, such as neodymium-doped yttrium-aluminium garnet Nd^{3+} : Y$_3$Al$_5$O$_{12}$ (Nd : YAG) and orthoaluminate Nd^{3+} : YAlO$_3$ (Nd : YAP) are one of the most popular active laser media. The luminescent properties of these crystals determined by transitions from the metastable $^4F_{3/2}$ level and some other levels are well studied [1, 2].

Recently, because of an extensive use of diode pumping, which provides a substantial population inversion due to excitation of many activator ions, absorption from the excited $^4F_{3/2}$ state and up-conversion attract great attention of researchers. These effects, which are manifested when the population of a metastable operating level is high, restrict an increase in the population inversion with pump intensity and reduce the gain at the lasing transition [3, 4].

On the other hand, such multistage excitation results in the population of high energy levels, which can make contributions to the dispersion and nonlinear optical properties of crystals [2, 5]. Recent studies of flashlamp-pumped Nd : YAG crystals showed, in particular, that the population of the high quasi-metastable $^2F(2)_{5/2}$ level with the lifetime of ~ 3 μs resulted in noticeable changes in the refractive index (Fig. 1) [5].

These changes in the refractive index can be used to produce dynamic nonlinear-optical gratings, which are promising for an adaptive correction of intracavity aberrations in high-power laser systems [5 – 7]. It was also shown that the populating of the $^2F(2)_{5/2}$ level was also accompanied by a strong increase in the luminescence intensity in the visible spectral region [8]. This effect is of interest for obtaining short-wavelength lasing in Nd^{3+}-doped crystals.

The aim of this work is to study the mechanisms of population of high energy levels in laser crystals Nd : YAG and Nd : YAP pumped by intense radiation from diode lasers and by the second, third, and fourth harmonics of a pulsed Nd : YAG laser, and to analyse the luminescence spectra of this crystals in the visible region.

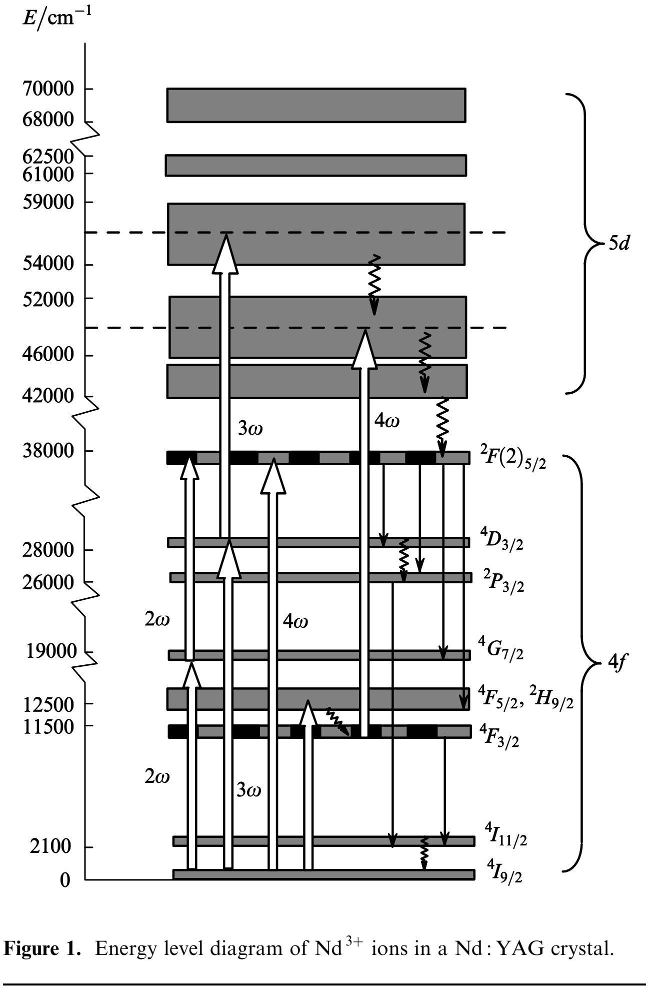

Figure 1. Energy level diagram of Nd^{3+} ions in a Nd : YAG crystal.

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2. Experimental

We studied the luminescence spectra of YAG and YAP crystals doped with Nd^{3+} ions at a concentration of $\sim 1\,\%$. The crystals were pumped by laser diodes and (or) by the Nd : YAG laser and its harmonics (Fig. 2).

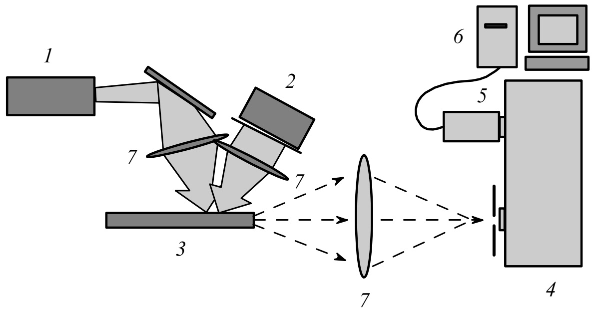


Figure 2. Scheme of the experimental setup: (*1*) Nd : YAG laser and generator of harmonics based on this laser; (*2*) laser diode array; (*3*) Nd : YAG or Nd : YAP crystal; (*4*) DFS-12; (*5*) FEU-79 photomultiplier; (*6*) ADC; (*7*) focusing lenses.

Pumping in the 807 – 808-nm region was performed by a Thalas-CSF cw laser diode array with an average power up to 25 W (at a 30-A current). To reduce crystal heating, cw radiation was modulated with a chopper (the 'light window' duration was ~ 6 ms at a pulse repetition rate of $\sim$ 12.5 Hz). In other experiments, we used a JENOPTIC Laserdiode GmbH pulsed diode array emitting 200 – 300-μs pulses at 808 nm with a peak power of up to 300 W. The emission wavelength of laser diodes was tuned within some spectral range by varying the temperature of cooling water.

Pumping was also performed by the second (532 nm), third (354.7 nm), or fourth (266 nm) harmonics of a pulsed 1064-nm Nd : YAG laser, which provided both direct and multistage population of the $^2F(2)_{5/2}$ level (Fig. 1). The duration of the 1064-nm laser pulse was 10 nm and the pulse repetition rate was 12.5 Hz. Radiation of the Nd : YAG laser was doubled in a CDA crystal, while the third and fourth harmonics were obtained with the help of a DKDP crystal. The energy of the second-, third-, and fourth-harmonic pulses was $\sim$ 10, 0.5, and $\sim$ 2 mJ, respectively.

Laser beams were focused on crystals under study. The diode arrays had cylindrical lenses at the exit, which reduced the beam divergence down to $0.1 - 1.0°$. The sizes of spots formed by focused beams of harmonics of the Nd : YAG laser and laser diodes were 10×0.1 mm and $7 \div 10 \times 0.2 \div 0.5$ mm, respectively.

The luminescence spectrum in the range from 380 to 650 nm was recorded with a DFS-12 spectrometer. The output signal from a FEU-79 photomultiplier was amplified and processed with a PC connected with the photomultiplier via a 12-bit analogue-to-digital converter. The signal-to-noise ratio was improved after averaging over 10 – 20 detected pulses.

3. Luminescence of crystals excited by harmonics of a Nd : YAG laser

We studied the luminescence spectra of Nd : YAG and Nd : YAP crystals excited by the second, third, and fourth harmonics of the Nd : YAG laser.

3.1 Luminescence of a Nd : YAG crystal

The luminescence spectra of the Nd : YAG crystal excited by the second or fourth harmonic of the Nd : YAG laser proved to be identical in the range from 400 to 500 nm (Fig. 3). The frequencies of most intense spectral lines in this range correspond, with good accuracy, to the transitions from two lower Stark sublevels of the $^2F(2)_{5/2}$ state with energies 37768 and 37789 cm^{-1} [9]. The final levels of these transitions were identified as levels $^2K_{13/2}$, $^2G_{9/2}$, $^2G_{7/2}$, $^4G_{5/2}$, $^4F_{9/2}$, $^2H_{9/2}$ and $^2F_{5/2}$. Several lines of lower intensity were assigned to the $^4D_{3/2} \to {}^4I_{15/2}$ transitions.

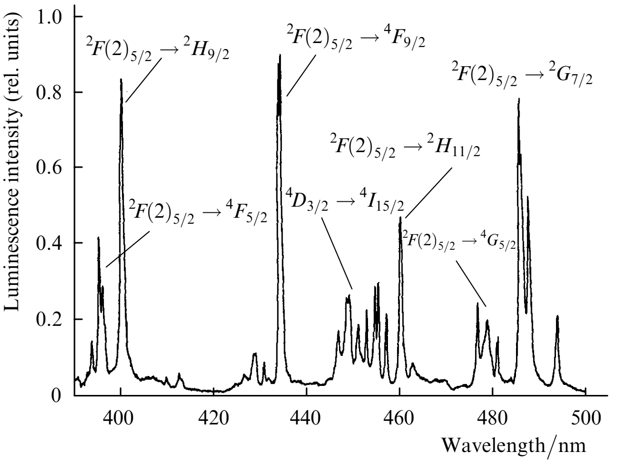


Figure 3. Luminescence spectrum of the Nd : YAG crystal excited at 532 or 266 nm.

One of the main goals of our work was to study the population of the high-energy quasi-metastable $^2F(2)_{5/2}$ level. We analysed the population of this level by monitoring luminescence at the 400.5-nm line corresponding to the $^2F(2)_{5/2} \to {}^2H_{9/2}$ transition [10]. The study of the intensity of this line showed that excitation by the second or fourth harmonics results in the population of the $^2F(2)_{5/2}$ level of Nd^{3+} ions due to transitions from the ground $^4I_{9/2}$ state. The intensity of the 400.5-nm line quadratically depended on the second harmonic intensity and linearly on the fourth harmonic intensity (Figs 4 and 5). These dependences are

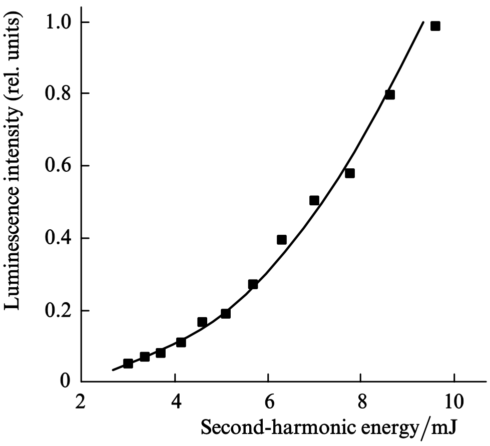


Figure 4. Dependence of the luminescence intensity of the Nd : YAG crystal at 400.5 nm on the second-harmonic pump energy.

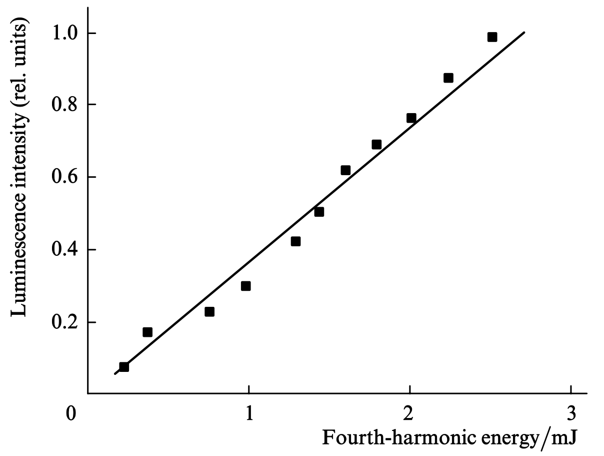


Figure 5. Dependence of the luminescence intensity of the Nd : YAG crystal at 400.5 nm on the fourth-harmonic pump energy.

explained by two- or single-stage excitation of the $^2F(2)_{5/2}$ level from the ground $^4I_{9/2}$ state, respectively.

Upon excitation of the crystal by the second harmonic, the $^2F(2)_{5/2}$ level was excited via the $^4G_{7/2}$ level having a short lifetime of ~ 0.37 ns [11]. For this reason, the excitation efficiency was low even when the second-harmonic pulse energy was almost four times greater than that of the fourth-harmonic pulse.

The luminescence spectrum of the Nd : YAG crystal excited by the third harmonic depended strongly on the laser beam energy. The luminescence lines excited by laser pulses with the energy lower than 0.2 mJ were assigned to the transitions $^4D_{3/2} \rightarrow {}^4I_{13/2}$, $^2P_{3/2} \rightarrow {}^4I_{11/2}$ and $^4D_{3/2} \rightarrow {}^4I_{15/2}$, $^2P_{3/2} \rightarrow {}^4I_{13/2}$ (Fig. 6). These transition occur from the long-lived $^4D_{3/2}$ and $^2P_{3/2}$ levels with the lifetimes equal to 2.2 and 300 ns, respectively [12]. These levels are populated due to transitions from the ground state upon absorption of third-harmonic photons.

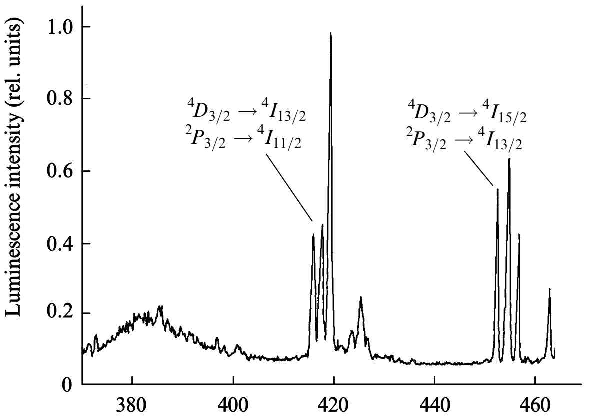


Figure 6. Luminescence spectrum of the Nd : YAG crystal excited by the third harmonic pulses with energy of no more than 0.2 mJ.

As the energy of third-harmonic pulses was increased, new luminescence lines appeared, which corresponded to the transitions from the $^2F(2)_{5/2}$ level to the levels $^4F_{5/2}$, $^2H_{9/2}$ and $^4F_{9/2}$. The intensity of these transitions depended quadratically on the pump energy. The quadratic dependence can be explained by two-step absorption, when Nd^{3+} ions undergo a transition from the excited Nd^{3+} level to the levels of the 5*d* configuration after absorption of the second photon. A total energy of such two-step excitation equal to 56 400 cm^{-1} corresponds to the third absorption band of the 5*d* shell in the Nd : YAG crystal [12]. Thermalisation of excitation inside the 5*d* shell and the fast nonradiative 5*d* → 4*f* transition (for ~ 2 ns [2]) lead to the population of the $^2F(2)_{5/2}$ level, from which we observed luminescence (Fig. 7).

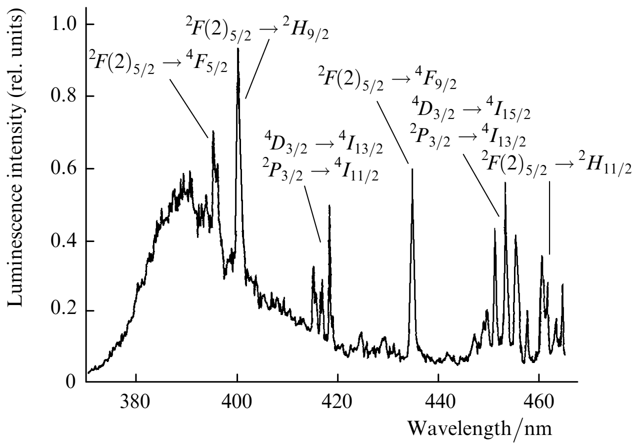


Figure 7. Luminescence spectrum of the Nd : YAG crystal excited by the ~ 0.5-mJ third harmonic pulses.

Note that the results of our study of the luminescence of the Nd : YAG crystal excited by a laser well agree with the results of earlier investigations [9 – 14]. At the same time, we observed several new spectral lines and, in particular, a broad band in the range from 380 to 400 nm (Figs 6, 7).

3.2 Luminescence of a Nd : YAP crystal excited by harmonics

We studied the luminescence spectra of the Nd : YAP crystal excited by the second, third, and fourth harmonics of a pulsed Nd : YAG laser (Figs 8 and 9). Upon excitation by the third harmonic, we observed luminescence lines corresponding to the transitions $^4D_{3/2} \rightarrow {}^4I_{13/2}$, $^2P_{3/2} \rightarrow {}^4I_{11/2}$, $^4D_{3/2} \rightarrow {}^4I_{15/2}$ and $^2P_{3/2} \rightarrow {}^4I_{13/2}$, whose frequencies

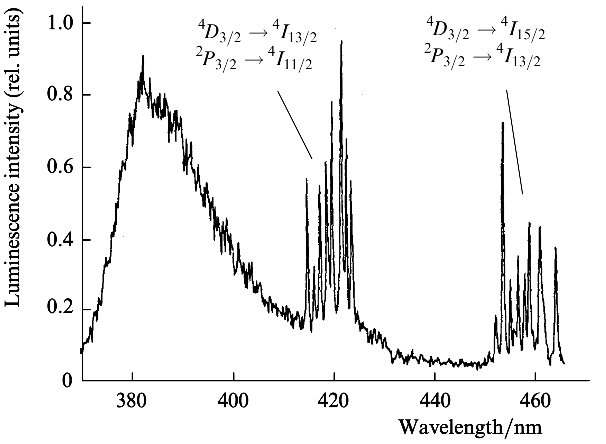
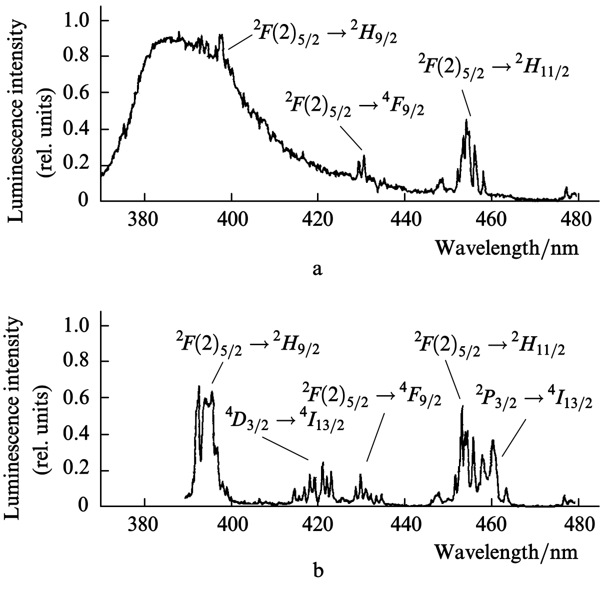


Figure 8. Luminescence spectrum of the Nd : YAP crystal excited by the third harmonic pulses.

were close to those of the lines of the Nd:YAG crystal (Fig. 8).

At the same time, the luminescence spectra of the Nd : YAP crystal excited by the second and fourth harmonics (Fig. 9) differ from the spectra of the Nd : YAP crystal observed upon the same excitation. Thus, the Nd : YAP crystal excited by the second harmonic exhibits, along with transitions from the levels $^2F(2)_{5/2}$ and $^4D_{3/2}$, the 461-nm luminescence line corresponding to the transition from the $^2P_{3/2}$ level, which was not observed in the luminescence spectrum of the Nd : YAG crystal. Upon excitation by the fourth harmonic, we observed only the lines corresponding to transitions from the $^2F(2)_{5/2}$ level. However, these lines in the luminescence spectrum of the Nd : YAP crystal were slightly shifted to the blue compared to similar lines in the Nd : YAG crystal. This can be explained by the fact that two lower Stark sublevels of the $^2F(2)_{5/2}$ states of the Nd^{3+} ions in the YAP matrix are located slightly higher than in the YAG matrix [11, 14].



a

b

Figure 9. Luminescence spectra of the Nd : YAP crystal excited by the second (a) and fourth (b) harmonic pulses.

Note that a broad luminescence band of the Nd : YAP crystal located in the region between 375 and 400 nm was more intense than such a band in the Nd : YAG crystal (Figs 8 and 9). This luminescence band can be assigned to the overlapped transitions from the levels of the 5*d* shell.

4. Short-wavelength luminescence observed upon combined pumping

We studied the short-wavelength luminescence of the Nd : YAG and Nd : YAP crystals in the spectral range from 380 to 650 nm excited by laser diodes at 808 nm and by harmonics of the Nd : YAG laser.

When the Nd : YAG and Nd : YAP crystals were excited by laser diodes only (both continuous and pulsed), no luminescence was observed in the 380 – 650 range up to the laser power densities 10 – 15 kW cm^{-2}. However, when we simultaneously excited the Nd : YAG crystal by a diode array at 808 nm and the fourth harmonic of a pulsed Nd : YAG laser, we observed a strong increase in the intensity of luminescence from the $^2F(2)_{5/2}$ level with increasing power of the diode pump (Fig. 10). The luminescence intensity depended on the delay of the fourth-harmonic pulse with respect to the diode pump pulse. When the delay was increased from zero to 200 μm, the short-wavelength luminescence intensity increased almost linearly. The maximum luminescence intensity was observed for the delay ~ 250 μm, which is equal to the lifetime of the metastable $^4F_{3/2}$ level. As the time delay was further increased, no increase in the luminescence intensity was observed.

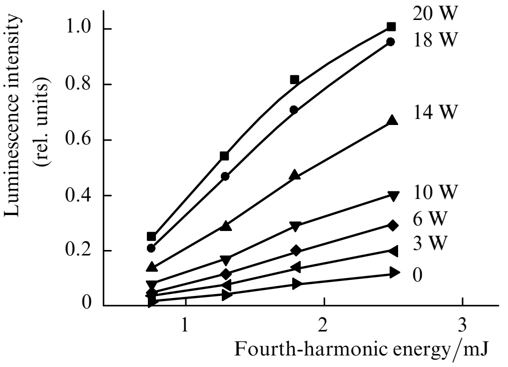


Figure 10. Luminescence intensity of the Nd : YAG crystal at 400.5 nm excited simultaneously by the laser diode array and the fourth harmonic of a pulsed Nd : YAG laser as a function of the fourth-harmonic pulse energy at different diode-array powers.

We studied the luminescence intensity at 400.5 nm excited simultaneously by the fourth harmonic of the Nd : YAG laser and the diode array as a function of the average power of the diode array at different wavelengths of the laser diode emission. The diode pump wavelength was tuned by varying the temperature of cooling water. We found the optimal operating temperature of the laser diode array (Fig. 11). The luminescence intensity in the optimal regime was an order of magnitude greater than that upon excitation by the fourth harmonic only. In this case, the ratio of the peak power of the fourth harmonic to that of the

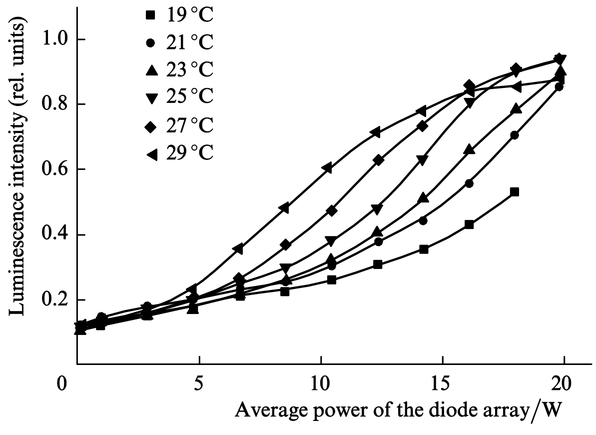


Figure 11. Luminescence intensity of the Nd : YAG crystal at 400.5 nm excited simultaneously by a cw laser diode array and the fourth harmonic of a pulsed Nd : YAG laser as a function of the average power of the laser diode array for different temperatures of cooling water.

diode array was $\sim 10^5$. This result demonstrates an important role of the diode pump (providing the population of the $^4F_{3/2}$ level) in excitation of luminescence from the $^2F(2)_{5/2}$ level.

In experiments with a pulsed diode array, we observed even more intense luminescence from the $^2F(2)_{5/2}$ level. The luminescence signal increased approximately by a factor of fifty when the power of the diode-array pulse was increased from zero to 200 W (Fig. 12). The delay of a fourth-harmonic pulse with respect to a diode-array pulse was selected experimentally by the luminescence intensity maximum, which was achieved at the end of the diode-array pulse (for delay $\sim$ 250 μs).

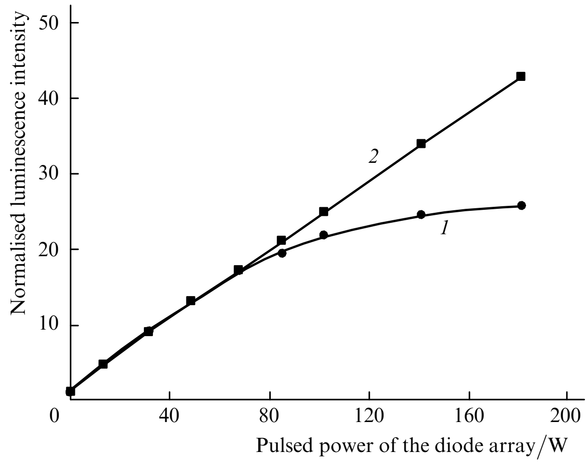


Figure 12. Luminescence intensity of the Nd : YAG crystal at 400.5 nm excited simultaneously by a pulsed laser diode array and the fourth harmonic of a pulsed Nd : YAG laser normalised to the luminescence intensity observed upon excitation by the fourth harmonic only as a function of the pulsed power of the laser diode array at a constant temperature of cooling water (*1*) and the cooling-water temperature reduced with increasing diode-array power (*2*).

A strong increase in the short-wavelength luminescence intensity of the Nd : YAG crystal with increasing the diode pump power can be explained by a two-state transition to the $^2F(2)_{5/2}$ level from the ground $^4I_{9/2}$ state via the intermediate metastable $^4F_{3/2}$ level (Fig. 1). Indeed, the diode pump at 808 nm provides the efficient population of the $^4F_{3/2}$ level, which has the lifetime of $\sim$ 250 μs and is the upper level of the lasing transition at 1064 nm. The population of the $^4F_{3/2}$ level under our experimental conditions was rather high. Its estimate from a small-signal gain showed that up to 10 % of all Nd^{3+} ions were excited (the atomic concentration of Nd^{3+} ions in a Nd : YAG crystal equal to 1% corresponds to their volume concentration equal to 1.38×10^{20} cm^{-3} [2]). For this reason, in the presence of the fourth-harmonic beam, along with transitions from the ground $^4I_{9/2}$ state, upward transitions occur from the metastable $^4F_{3/2}$ level. Absorption of the fourth-harmonic radiation from the excited $^4F_{3/2}$ state can transfer Nd^{3+} ions to the second band of the 5*d* shell (to levels with the energy $\sim$ 49000 cm^{-1}). This absorption from the excited state is rather strong because the cross section for the interconfiguration $4f \rightarrow 5d$ transition from the $^4F_{3/2}$ level to the second band of the 5*d* shell is two orders of magnitude higher than the cross section for the $4f \rightarrow 4f$ transitions, while the fourth-harmonic frequency lies near the resonance at a wavelength of 263 nm [2, 14].

After such a two-step transition to the 5*d* shell, nonradiative relaxation occurs to the lower levels of this shell, which is followed by nonradiative relaxation to the upper levels of the 4*f* shell (for the time $\sim$ 2 ns [2]), resulting, in particular, in the population of the quasi-metastable $^2F(2)_{5/2}$ level. Note that, despite the energy loss due to nonradiative relaxation, the efficiency of such two-step excitation of the $^2F(2)_{5/2}$ state proved to be much higher than that upon a direct absorption of UV radiation from the ground state (the transitions $^4I_{9/2} \rightarrow {}^2F(2)_{5/2}$ and $^4I_{11/2} \rightarrow {}^2F(2)_{5/2}$).

We did not observe any increase in the short-wavelength luminescence intensity upon excitation of the Nd : YAG crystal simultaneously by the second harmonic of the Nd : YAG laser and the diode array, as well as by the third harmonic of this laser and the same diode array. This can be explained by a low energy of photons of the second and third harmonics (compared to the energy of the $4f \rightarrow 5d$ transition) and by the absence of intermediate absorption resonances from the metastable $^4F_{3/2}$ level for this radiation (Fig. 1).

Note also that no increase in the short-wavelength luminescence intensity was observed in the Nd : YAP crystal simultaneously excited by the diode array and the second, third, or fourth harmonics of the pulsed Nd : YAG laser. This can be explained by the fact that the energy level diagram of Nd^{3+} ions in the Nd : YAP crystal is somewhat different. Two absorption bands corresponding to the 5*d* shell of the Nd : YAP crystal are located at 53400 and 55200 cm^{-1} [15]. We can easily estimate that the total energy of photons involved in simultaneous pumping (49100 cm^{-1}) is insufficient for the $4f \rightarrow 5d$ transition to occur.

5. Conclusions

We have studied the luminescence of the Nd : YAG and Nd : YAP crystals in the 390 – 650-nm spectral range excited by diode arrays and the second, third, or fourth harmonics of a pulsed Nd : YAG laser. We have established the mechanisms of the population of the high quasi-metastable $^2F(2)_{5/2}$ level. This level is populated most efficiently upon simultaneous pumping the Nd : YAG crystal by the diode array and the fourth harmonic of the Nd : YAG laser. In this case, we observed a strong (by an order of magnitude and more) increase in the intensity of luminescence emitted from the high $^2F(2)_{5/2}$ level (bright blue emission of the crystal was observed visually). The level was populated due to two-step excitation: the diode pump results in the population of the upper metastable operating level, while fourth-harmonic photons provide absorption from the excited state due to the allowed interconfiguration $4f \rightarrow 5d$ transition followed by nonradiative relaxation to the $^2F(2)_{5/2}$ level.

This effect is important for the understanding of the mechanism of a change in the refractive index of laser crystals caused by the difference between the polarisabilities of high-energy levels and of the ground state [5, 16]. The results that we obtained show, in fact, that we can control the optical nonlinearity of laser crystals by combining the diode and Nd : YAG laser pumps because the population of upper energy levels makes a substantial contribution to nonlinear variations of the refractive index [2, 5, 16]. Another important result of our study is the possibility of obtaining intense luminescence in the violet – blue spectral region.

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