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Control of the spectral parameters of vanadate lasers*

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Abstract. **It is experimentally established that the luminescence spectra of Nd^{3+} ions at the $^4F_{3/2}-{}^4I_{11/2}$ transition in $Nd^{3+}:YVO_4$, $Nd^{3+}:GdVO_4$, $Nd^{3+}:Gd_{1-x}Y_xVO_4$, and $Nd^{3+}:Sc_{1-x}Y_xVO_4$ vanadate crystals strongly depend on the recording and polarisation angles with respect to the crystallographic axes. Using these angular dependences of luminescence spectra, it is possible to control the gain coefficient for lasers based on vanadate crystals, create broad gain bands for pico- and femtoseconds lasers, and change radiation wavelengths. Efficient operation of a passively *Q*-switched laser based on a $Nd^{3+}:GdVO_4$ crystal cut at an angle to the *a* axis ($\theta = 25°$, $\varphi = 0$) is demonstrated for the first time.**

Keywords: *luminescence spectra, vanadate crystals, passive Q-switching, gain coefficient.*

1. Introduction

The $Nd^{3+}:YVO_4$ [1], $Nd^{3+}:GdVO_4$ [2], $Nd^{3+}:Gd_{1-x}Y_xVO_4$ [3] and $Nd^{3+}:Sc_{1-x}Y_xVO_4$ [4] vanadate crystals are excellent materials for diode-pumped lasers. These crystals have large absorption and stimulated emission cross sections, as well as broad absorption lines at the pump wavelength. The combination of the spectral and mechanical properties of these crystals ensures a high efficiency of cw and pulsed lasers. The lattice anisotropy of vanadate crystals makes it possible to obtain polarised radiation, while the high thermal conductivity allows efficient cooling of the active media.

The difference in the properties of vanadate crystals along different axes allows one, if needed, to choose directions with preferable gain cross section. Most of known studies on vanadate crystals are devoted to the use of *a*-cut vanadate crystals for π-polarised ($\boldsymbol{E}\|c$) laser radiation, because the gain in this case is maximal. In passively *Q*-switched lasers with $Cr^{4+}:YAG$ or $V^{3+}:YAG$ *Q*-switches, one uses *c*-cut ($\boldsymbol{E}\perp c$) [5, 6] or, in the case of σ-polarised radiation, *a*-cut ($\boldsymbol{E}\perp c$) [7, 8] vanadate crystals. In these cases the gain in the active media is minimal.

In [9, 10], it was proposed to obtain two-frequency lasing based on equalisation of the cavity Q-factors for different spectral regions with control of the active-medium gain by rotating the vanadate crystal. However, the angular dependences of gain coefficients in different spectral regions have not been studied in detail.

The aim of this work is to experimentally study the angular dependences of the luminescence of $Nd^{3+}:YVO_4$, $Nd^{3+}:GdVO_4$, $Nd^{3+}:Gd_{1-x}Y_xVO_4$ and $Nd^{3+}:Sc_{1-x}Y_xVO_4$ vanadate crystals at the $^4F_{3/2}-{}^4I_{11/2}$ transition, as well as to study the possibility of using these dependences when creating new laser media. As an example of controlling gain in active media, we demonstrate efficient operation of passively *Q*-switched lasers based on $Nd^{3+}:GdVO_4$ gadolinium vanadate crystals cut at an angle to the *a* axis ($\theta = 25°$, $\varphi = 0$) and $Cr^{4+}:YAG$ saturable absorbers.

2. Study of luminescent parameters of vanadate crystals as functions of the polarisation angle

In [8–10], it was shown that the shape of luminescence spectra of Nd^{3+} at the $^4F_{3/2}-{}^4I_{11/2}$ transition in *a*-cut $Nd^{3+}:YVO_4$, $Nd^{3+}:GdVO_4$, $Nd^{3+}:Gd_{0.7}Y_{0.3}VO_4$, and $Nd^{3+}:Sc_{0.01}Y_{0.99}VO_4$ vanadate crystals for the σ-polarised laser radiation coincides with the shape of luminescence spectra of the *c*-cut crystals ($\boldsymbol{E}\perp c$). The absolute maxima in the luminescence spectra for the σ-polarisation are shifted to longer wavelengths with respect to the maxima for the π-polarisation. Therefore, the wavelengths of lasing in these crystals are different for the σ- and π-polarisations. To better understand the mechanism of transformation of the luminescence spectra of vanadate crystals with rotating the crystals from π- to σ-polarisation or with varying the angle θ between the observation axis and the *c* axis ($\varphi = 0$), we experimentally studied the angular dependences of the luminescence spectra of $Nd^{3+}:YVO_4$, $Nd^{3+}:GdVO_4$, $Nd^{3+}:Gd_{1-x}Y_xVO_4$ and $Nd^{3+}:Sc_{1-x}Y_xVO_4$ vanadate crystals at room temperature and at 77 K (liquid nitrogen boiling temperature).

All the studied crystal were grown by the Czochralski method at the General Physics Institute, Russian Academy of Sciences. The spectroscopic characteristics of the laser crystals were studied using a spectrometer based on a UF-90 autocollimation chamber (reciprocal linear dispersion 0.1 nm mm^{-1}) with a TCD130JK (Toshiba) linear multichannel photodetector. The crystals were excited by a fibre-coupled LIMO HLU30F200 laser diode system (depolarised radiation, the maximum output power up to 30 W at the wavelength λ_p = 808 nm). The pump beam was focused into the crystal to a spot 200–600 μm in diameter.

The angular dependences of luminescence were measured for *a*-cut cylindrical vanadate crystals using two schemes (Fig. 1). In the first scheme (Fig 1a), the crystal was rotated

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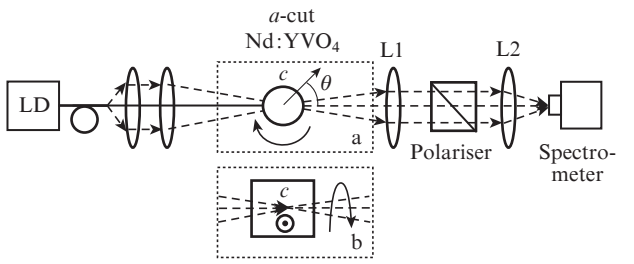

Figure 1. Scheme for measuring the dependences of luminescence of Nd^{3+} ions in cylindrical *a*-cut vanadate crystals on the angle θ (a) and the polarisation plane rotation angle φ (b).

around the *a* axis while the pump radiation propagated through the polished cylindrical surface. The luminescence spectra were recorded for different angles θ between the observation direction and the *c* axis ($\varphi = 0$). In the second case, pumping was performed through the face of the crystal, which was rotated around the observation axis so that the laser radiation smoothly changed from π- ($\boldsymbol{E} \parallel c$) to σ-polarised ($\boldsymbol{E} \perp c$) ($\theta = 0$).

Figure 2 shows the energy level diagram of neodymium and the luminescence spectra of $Nd^{3+}:YVO_4$ and $Nd^{3+}:GdVO_4$ crystals for the σ- and π-polarisations.

A specific feature of neodymium-doped vanadate crystals is a weak splitting of the upper $^4F_{3/2}$ and lower $^4I_{11/2}$ levels, because of which the spectra consist of overlapping transitions between Stark sublevels. The spectrum in the studied wavelength range 1060–1070 nm consists of four transitions, R_1-Y_1, R_2-Y_1, R_1-Y_2, and R_2-Y_2.

The splitting of the upper $^4F_{3/2}$ level for gadolinium vanadate is very small and is noticeable only at liquid helium temperature. Thus, the spectrum for the σ-polarisation in the wavelength range 1060–1070 nm has a typical double-humped shape (the wavelengths of the $R_1-Y_{1,2}$ and $R_2-Y_{1,2}$ transitions are close to each other). The spectrum of yttrium vanadate in this range has three peaks (wavelengths of the R_1-Y_1 and R_2-Y_2 transitions are similar). The splitting of the $^4F_{3/2}$ and $^4I_{11/2}$ levels of neodymium ions in mixed vanadates ($Nd^{3+}:Gd_{1-x}Y_x$ or $Nd^{3+}:Sc_{1-x}Y_x$) depends on the concentration ratio of Gd or Sc to Y. A change in the ratios $Gd_{1-x}Y_x$ or $Sc_{1-x}Y_x$ leads to a transformation of the gain profile in the wavelength range 1060–1070 nm, because of which the spectrum for the σ-polarisation in the general case has four peaks. This change in the luminescence wavelengths of mixed vanadates occurs for both polarisations.

The luminescence spectra of Nd^{3+} ions in vanadate crystals are characterised by pronounced dependences on the observation and polarisation angles with respect to the crystallographic axes. The vanadate crystals rotated as shown in the schemes in Fig. 1 demonstrate transformation of the shape of the luminescence spectra. The evolution of the lumines-

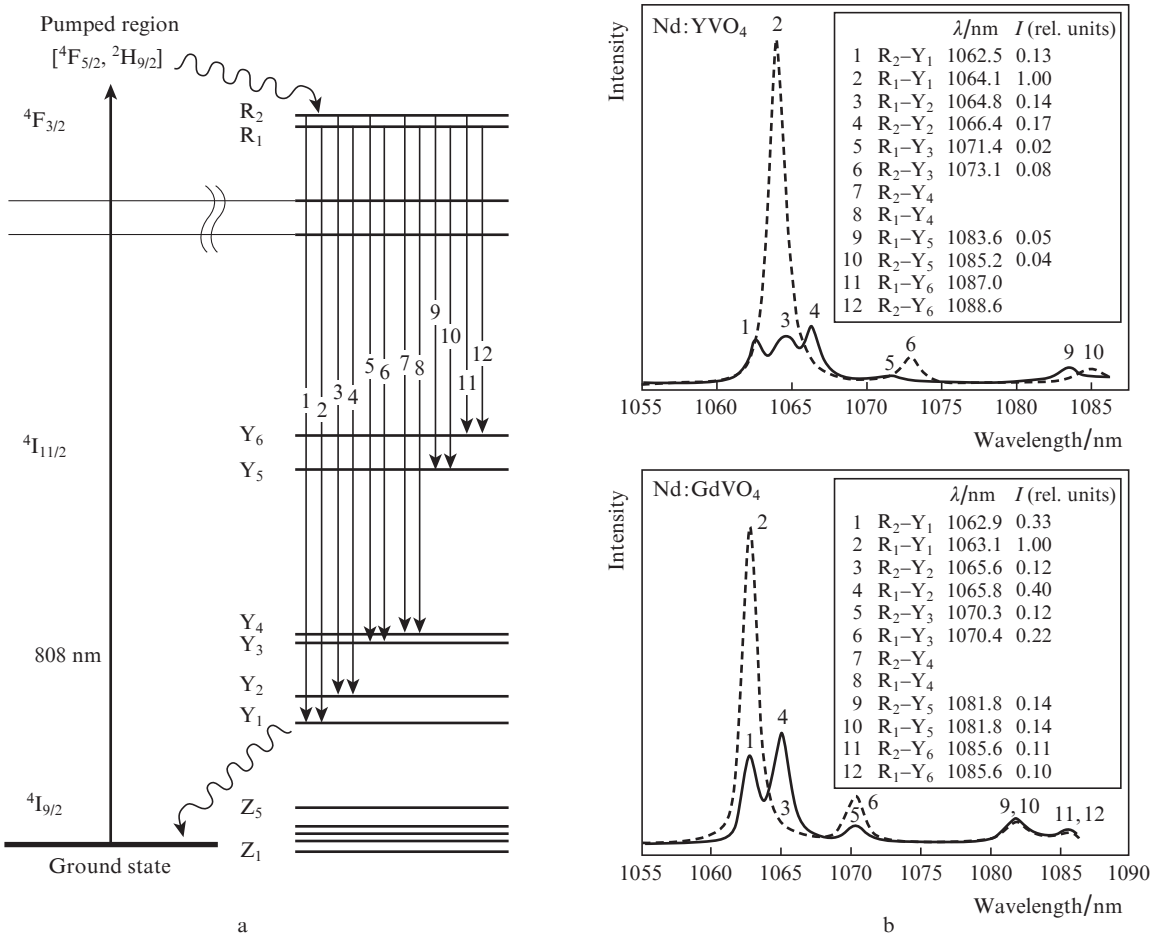

Figure 2. Energy level diagram of Nd^{3+} ions (a) and luminescence spectra of yttrium ($Nd^{3+}:YVO_4$) and gadolinium ($Nd^{3+}:GdVO_4$) vanadates at the $^4F_{3/2}-{}^4I_{11/2}$ transition for the π- and σ-polarisation (solid and dashed lines, respectively) (b).

cence spectra of $Nd^{3+}:YVO_4$ and $Nd^{3+}:GdVO_4$ crystals with rotation of the polarisation plane is shown in Figs 3a, 3b.

Since the vanadate crystal have four Stark transitions of Nd^{3+} ions in the spectral range 1060–1070 nm, then, for processing the experimental data, we presented the spectra in the form of four peaks with fixed positions and calculated their amplitudes for each rotation angle. The dependences of emission amplitudes of each Stark transition on the rotation angle φ of the polarisation plane were given by the function $A = A_0 \sin(\varphi + \phi)$, where A_0 and ϕ are the approximation parameters.

As a result, we obtained the dependences of the emission intensity of four Stark transitions on the rotation angle of the crystal (Figs 3c, 3d). Note that the radiation has identical polarisation for some transition (R_1-Y_1, R_1-Y_3, and R_1-Y_5) and orthogonal for the others. With rotation of crystals by 90°, the luminescence cross section changes almost by five times (Figs 3e, 3f).

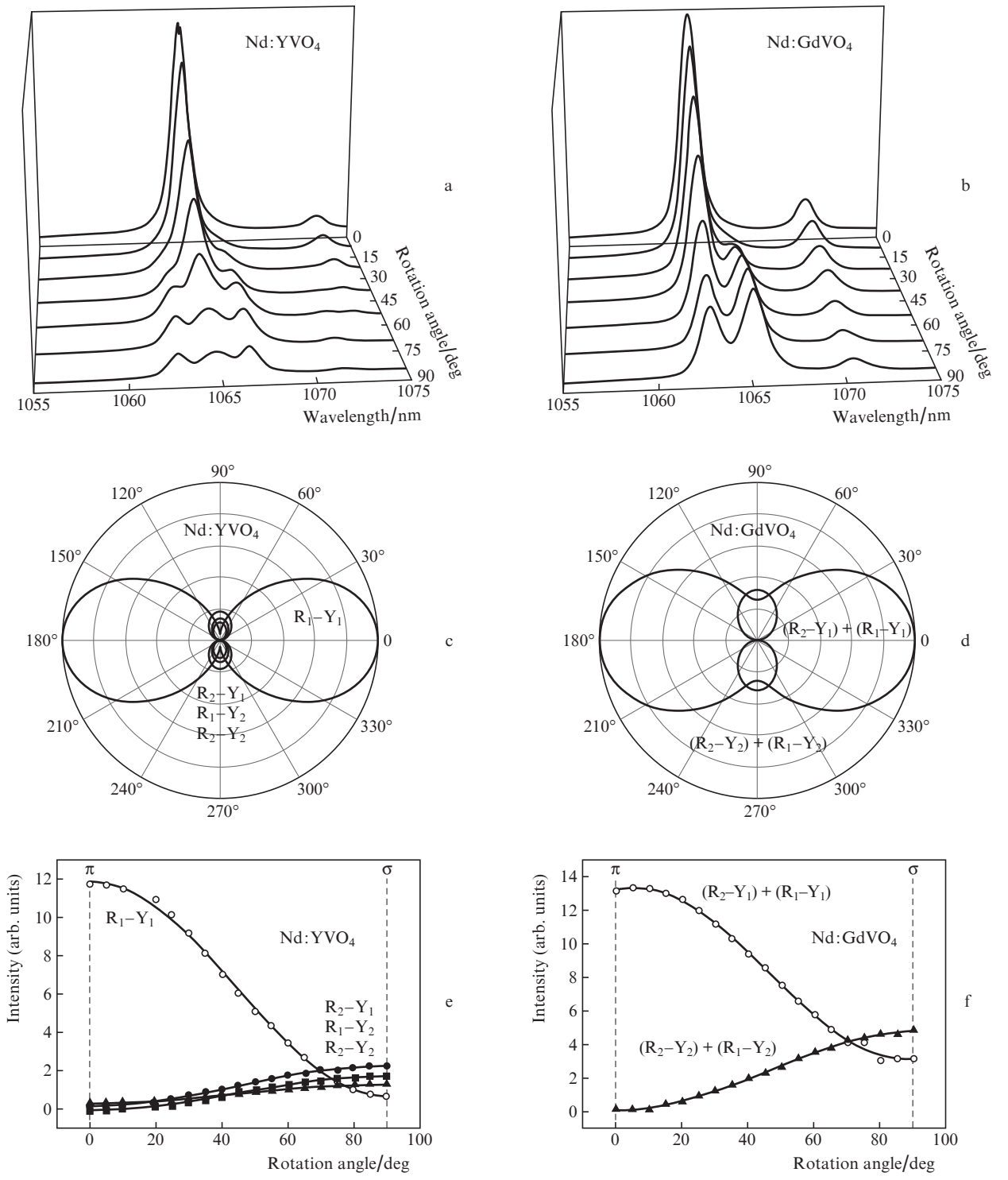

Figure 3. Luminescence parameters as functions of the polarisation plane rotation angle for $Nd^{3+}:YVO_4$ (a, c, e) and $Nd^{3+}:GdVO_4$ (b, d, f) crystals.

Of interest are the points of intersection of the curves for the R_1-Y_1 and R_2-Y_1 transitions (see Fig. 3e), in which the emission intensities of the Stark transitions are equal to each other (points A and B in Fig. 4). Point B corresponds to the scheme shown in Fig. 1b; in this case, the laser operates at the wavelength of the R_2-Y_1 transition to the right from the point B and at the R_1-Y_1 transition of the same polarisation to the left from this point. Point A corresponds to the scheme in Fig. 1b, and, like in the previous case, the laser operates at the R_2-Y_1 wavelength to the right from the point A and at the R_1-Y_1 wavelength to the left, but with the orthogonal polarisation in the latter case.

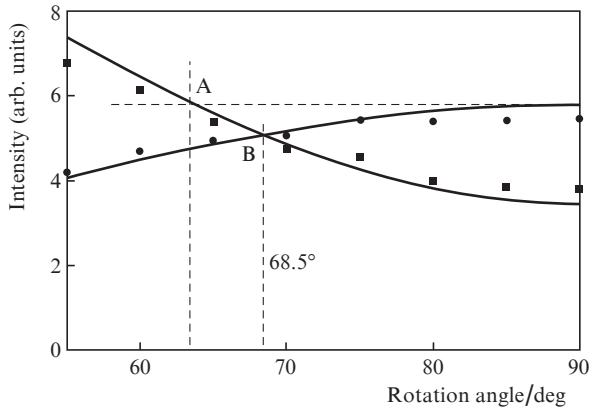

Figure 4. Special points on the angular dependence of the intensities of the Stark transitions of the $Nd^{3+}:GdVO_4$ crystal.

In addition, lasing in these points occurs at two frequencies with both parallel and orthogonal polarisations. The two-frequency lasing regime is based on the equality of the cavity Q-factor for different spectral regions, which is achieved by rotating the crystal at an angle at which the gain is identical for two frequencies.

Thus, cutting the crystal at a particular angle and using the angular dependences of the intensities of Stark transitions, it is possible to form laser crystals with required parameters, control gain coefficients in a wide range, and change the shape of luminescence spectra and laser wavelengths.

3. Formation of active media based on vanadate crystals

We propose two schemes of creating active media with wide uniform luminescence bands, namely, either by using composite active elements composed of $Nd^{3+}:YVO_4$, $Nd^{3+}:GdVO_4$ and mixed vanadate crystals $Nd^{3+}:Gd_{1-x}Y_xVO_4$ or $Nd^{3+}:Sc_{1-x}Y_xVO_4$, or by changing the concentrations of components in $Gd_{1-x}Y_x$ or $Sc_{1-x}Y_x$ in mixed vanadates $Nd^{3+}:Gd_{1-x}Y_xVO_4$ or $Nd^{3+}:Sc_{1-x}Y_xVO_4$.

Combination of vanadate crystals. The luminescence spectra of the $^4F_{3/2}-{}^4I_{11/2}$ transition of Nd^{3+} ions in the yttrium ($Nd^{3+}:YVO_4$) and gadolinium ($Nd^{3+}:GdVO_4$) vanadate crystals, as well as in vanadate crystals cut for σ-polarised radiation ($\boldsymbol{E}\perp c$) ($\theta = 0$, $\varphi = 0$) (see Fig. 3), allow us to conclude that the summed spectrum of these crystals can have five peaks (Fig. 5a). Unfortunately, the top of this spectrum is strongly carved. To obtain a smoother spectrum, we can use the angular dependences of its components. In particular, summing the luminescence spectrum of the yttrium vanadate crystal cut at the angle ($\theta = 30°$, $\varphi = 0$) and the spectrum of the $Nd^{3+}:Gd_{0.7}Y_{0.3}VO_4$ mixed vanadate crystal ($\theta = 35°$, $\varphi = 0$), we obtain almost ideal spectrum (Fig. 5b) with a halfwidth of 5.4 nm and a flat top, which can be used, for example, for generation of femtoseconds pulses.

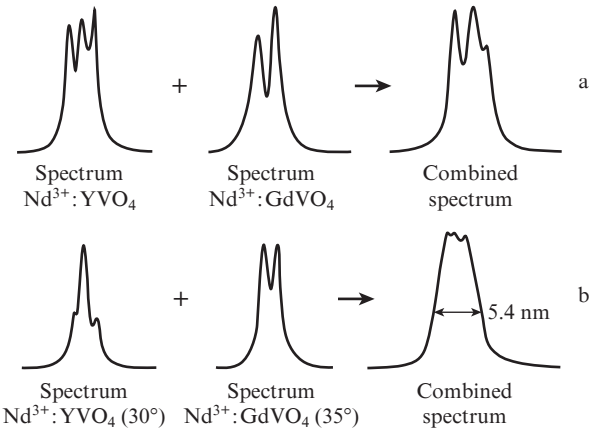

Figure 5. Combined luminescence spectra of Nd^{3+} ions in active elements composed of a-cut ($\theta = 0$, $\varphi = 0$) yttrium ($Nd^{3+}:YVO_4$) and gadolinium ($Nd^{3+}:GdVO_4$) vanadate crystals (a) and composed of $Nd^{3+}:YVO_4$ ($\theta = 30°$, $\varphi = 0$) and $Nd^{3+}:Gd_{0.7}Y_{0.3}VO_4$ ($\theta = 35°$, $\varphi = 0$) crystals (b).

Mixed vanadate crystals. The use of mixed vanadates is even more promising, because the wavelengths of all the Stark levels of the neodymium ion in $Nd^{3+}:Gd_{1-x}Y_xVO_4$ or $Nd^{3+}:Sc_{1-x}Y_xVO_4$ vanadates depend on the relative concentrations of Y, Gd, and Sc and each sublevel is broadened. This makes it possible to correct the amplitudes of Stark transitions to a greater or lesser degree by cutting the crystals at different angles.

Active media based on mixed vanadates can be formed according to the following scheme: the relative concentrations of Y and Gd are selected so that the distance between four Stark transitions (R_1-Y_1, R_2-Y_1 and R_1-Y_2, R_2-Y_2) would be identical and then a cut angle is selected to equalise the amplitudes of luminescence of the Stark levels.

In our experiments, the best crystal from the viewpoint of equality of the distances between the four Stark transition is the $Nd^{3+}:Gd_{0.45}Y_{0.55}VO_4$ mixed vanadate crystal cut at the angle $\theta = 20°$, $\varphi = 0$.

4. Continuous-wave laser based on a $Nd^{3+}:GdVO_4$ crystal with the cut angles $\theta = 18°$ and $90°$, $\varphi = 0$

We studied cw lasing in $Nd^{3+}:GdVO_4$ crystals cut at the angles ($\theta = 18°$, $\varphi = 0$) and ($\theta = 90°$, $\varphi = 0$). These angles allow lasing to the left and right from points A and B (Fig. 4). The experimental scheme is shown in Fig. 6.

We used $Nd^{3+}:GdVO_4$ crystals 4×4×8 mm in size with a neodymium concentration of 0.5 at.%. The laser crystal was wrapped with indium foil and mounted in a water-cooled copper block.

The laser cavity was formed by a plane highly reflecting mirror M1 (with a dielectric coating highly reflecting at the wavelength $\lambda_{gen} = 1064$ nm and with an antireflection coating

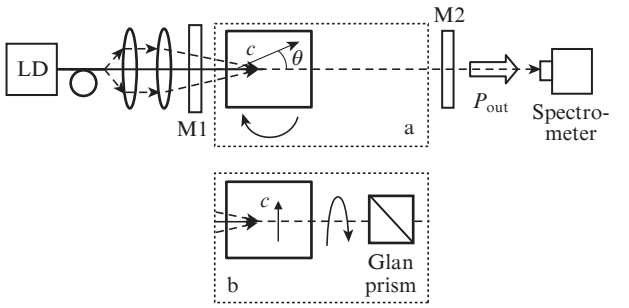

Figure 6. Scheme of a cw laser based on $Nd^{3+}:GdVO_4$ crystals cut at the angles $\theta = 18°$, $\varphi = 0$ (a) and $\theta = 90°$, $\varphi = 0$ (b).

for the wavelength $\lambda_p = 808$ nm) and a plane output mirror M2 (with the reflectivity $R = 10\%$ at the fundamental frequency). Both faces of the active element (AE) were antireflection coated for the wavelengths 808 and 1064 nm ($R \approx 0.02\%$).

When the $Nd^{3+}:GdVO_4$ crystal cut at the angle $\theta = 18°$, $\varphi = 0$ is rotated in the plane of the figure (see Fig. 6a), its luminescence spectra transform and the gain differently changes in different spectral regions. As a result, lasing occurs first at the wavelength $\lambda = 1063.2$ nm, then simultaneously at the wavelengths 1063.2 and 1065.5 nm, and finally at $\lambda = 1065.5$ nm. In this case, the radiation at first has one polarisation, then simultaneously two orthogonal polarisations, and finally the polarisation orthogonal to the first case.

This two-frequency lasing method is based on a balance of the cavity Q-factors in two spectral ranges, which is achieved by equalising the gain coefficient in the active medium. Although the cavity had no selective elements at all, the laser operation at two frequencies was very stable.

A similar angular dependence of the gain in different spectral regions was also observed when rotating an a-cut AE ($\theta = 90°$, $\varphi = 0$), during which the π-polarisation was changed to the σ-polarisation (Fig. 6b). The rotation was performed around the cavity axis. In this scheme, it is possible to fix the polarisation and rotate only the Glan prism. Similar to the previous scheme, we observed the transition from one wavelength to another and the occurrence of two-frequency lasing. However, the two-frequency operation of the latter scheme was less stable than in the first variant.

5. Passively Q-switched laser based on a $Nd^{3+}:GdVO_4$ crystal cut at the angle $\theta = 25°$, $\varphi = 0$

In [8], we studied the dependence of lasing characteristics of neodymium-doped vanadate crystals at the $^4F_{3/2}-{}^4I_{11/2}$ transition on the polarisation type (π or σ) in the passive Q-switching regime and showed that the best characteristics are achieved for these lasers with $Cr^{4+}:YAG$ Q-switches in the case of σ-polarised radiation or c-cut vanadate crystals. However, the wavelength in the latter cases does not coincide with the wavelength obtained in the case of the π-polarisation, at which the gain in the medium is maximal. To create an amplifier for the π-polarisation, one needs a medium which can operate, on the one hand, at a required wavelength and, on the other hand, in the passive Q-switching regime. These two conditions can be fulfilled by choosing a proper crystal cut angle.

The $Nd^{3+}:GdVO_4$ crystal cut at the angle $\theta = 25°$, $\varphi = 0$ has the same gain as the a-cut crystal ($\theta = 0$, $\varphi = 0$) for the σ-polarisation, and, at the same time, its lasing wavelength coincides with the wavelength for the π-polarised radiation.

The cavity of this laser is formed by a dielectric coating with a high reflectivity at $\lambda_{gen} = 1064$ nm and a low reflectivity at $\lambda_p = 808$ nm deposited on the entrance face of the AE and by a plane output mirror ($R = 75\%$ at λ_{gen}). The exit face of the AE was antireflection coated for the wavelengths 808 and 1064 nm ($R \approx 0.02\%$).

As passive Q-switches for the $Nd^{3+}:GdVO_4$ laser ($\theta = 25°$, $\varphi = 0$), we used $Cr^{4+}:YAG$ crystals. The optimal initial transmission T_0 of the Q-switch and the output mirror reflectivity at 1064 nm were 80% and 75%, respectively. The cavity length was about 12 mm. The time characteristics of pulses were measured by an LFD-2 germanium avalanche photodiode and a Tektronix TDS 3052 oscilloscope.

The dependences of the laser parameters (including pulse energy and power) on the absorbed pump power are presented in Fig. 7.

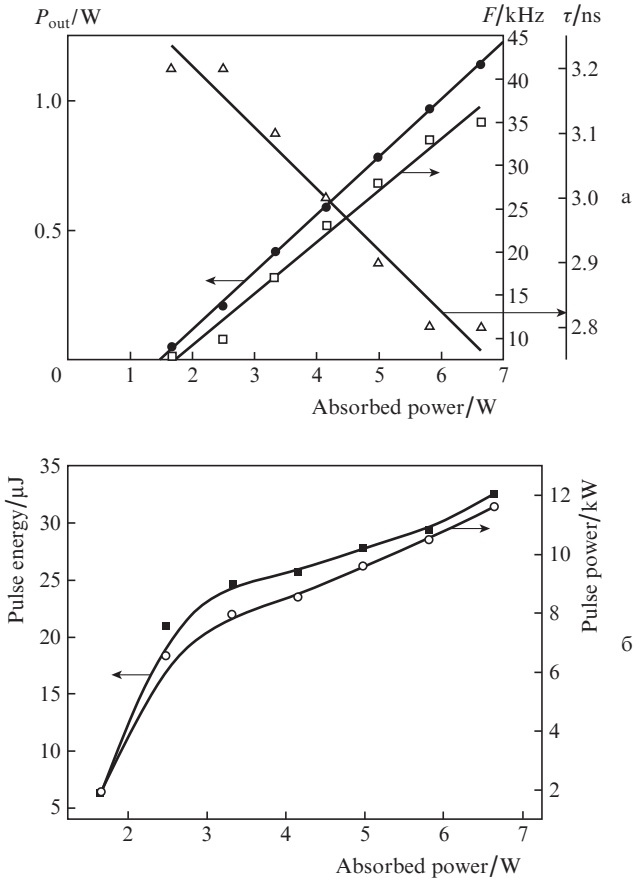

Figure 7. Dependences of the average power P_{out}, pulse duration τ, and pulse repetition rate F (a), as well as of the pulse energy and peak power (b) on the absorbed pump power for a $Nd^{3+}:GdVO_4$ laser ($\theta = 25°$, $\varphi = 0$).

The threshold pump power and the slope efficiency reached 1.45 W and 22% in the pulsed regime. The average power was 0.92 W at an absorbed pump power of 5.9 W. The minimum pulse duration of 2.8 ns was achieved at a pulse repetition rate of 35 kHz. The maximum pulse power exceeded 12 kW, while the pulse energy was higher then 32 μJ.

Thus, it is shown that the temporal and energy parameters of the passively Q-switched $Nd^{3+}:GdVO_4$ laser ($\theta = 25°$, $\varphi = 0$) are comparable or better than the best data reported in the

literature for lasers based on *c*-cut vanadate crystals ($\boldsymbol{E}\perp c$) [5, 6] or on *a*-cut vanadate crystals with σ-polarised radiation ($\boldsymbol{E}\perp c$) [7, 11]. In addition, the described laser operates at a wavelength of 1063.2 nm, which coincides with the wavelength of the $Nd^{3+}:GVO_4$ laser with π-polarised radiation (linear polarisation). This crystal is an ideal medium for an oscillator–amplifier system and for subsequent nonlinear frequency conversion.

6. Conclusions

The angular dependences of the luminescence parameters of Nd^{3+} at the ${}^4F_{3/2}-{}^4I_{11/2}$ transition in $Nd^{3+}:YVO_4$, $Nd^{3+}:GdVO_4$, $Nd^{3+}:Gd_{1-x}Y_xVO_4$, and $Nd^{3+}:Sc_{1-x}Y_xVO_4$ vanadate crystals are experimentally studied for the first time. It is shown that the use of these dependences for vanadate crystals makes it possible to control the gain coefficient, create maximally wide gain lines for pico- and femtosecond lasers, or control the laser wavelength. Efficient operation of a passively *Q*-switched $Nd^{3+}:GdVO_4$ laser ($\theta = 25°$, $\varphi = 0$) is demonstrated for the first time.

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