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Calcium–niobium–gallium and calcium-lithium-niobium-gallium garnet crystals as active media for diode-pumped lasers

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Abstract. The energy and spectral parameters of calcium $$ niobium - gallium and calcium - lithium - niobium - gallium garnet crystals pumped by a 2-W laser diode are studied. The stable parameters of laser radiation are demonstrated upon small variations in the temperature of the pump laser diode.

Keywords: semiconductor pump, output characteristics, garnet crystals.

The spectral and lasing properties of calcium–niobium– gallium garnet $Ca_3Nb_{1.68}Ga_3O_{12}$ (CNGG) crystals doped with Nd³⁺ ions were first studied in papers $[1-3]$ and those of calcium - lithium - niobium - gallium garnet (CLNGG) crystals ë in pap[er \[4\]. A](#page-2-0)t present, disordered media doped with rare-earth ions, in particular, CNGG crystals doped with Nd^{3+} ions [5, 6] again attract the attention of researchers.

The attractive properties of these crystals are their low melting temperature (1460 $^{\circ}$ C), which allows one to grow them from platinum crucibles, and their specific structure giving rise to a strong inhomogeneous broadening of the spectral lines of impurity rare-earth ions. The inhomogeneous broadening of spectral lines in CNGG and CLNGG crystals is comparable to that in glasses, however, these crystals have a higher heat conduction and feature stable optical properties and a high mechanical strength.

We optimised the ion composition of these crystals and the impurity Nd^{3+} ion concentration for using them as active elements in flashlamp-pumped Q -switched and freerunning lasers. The parameters of these lasers at the average output power up to 30 W were better than those for Nd : YAG lasers [\[4, 7\].](#page-2-0)

We studied earlier mode-locked lasing in these crystals. Due to the inhomogeneous broadening of the luminescence lines of Nd^{3+} in Nd^{3+} :CNGG and Nd^{3+} :CLNGG crystals, we obtained $10 - 15$ -ps output pulses both upon passive and active mode locking. The lasing efficiency of the crystals proved to be higher than that of industrial laser glasses [\[8, 9\].](#page-2-0)

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However, a lower heat conduction of these crystals compared to that of a YAG crystal limited their applications upon flashlamp pumping. The use of semiconductor lasers for pumping Nd^{3+} : CNGG and Nd^{3+} : CLNGG crystals removes this limitation.

We studied several Nd^{3+} : CNGG and Nd^{3+} : CLNGG crystals with the different atomic concentration n_a of Nd³⁻ ions. The crystals were grown by the Czochralski method in platinum crucibles. The activator concentration was controlled by the X-ray fluorescence method.

Fig. 1 shows the absorption spectra of Nd^{3+} : CNGG crystals at different concentrations n_a and of a standard Nd^{3+} : YAG crystal with $n_a = 1\%$. The maxima of absorption lines of the Nd³⁺:CNGG and Nd³⁺:CLNGG crystals are located at 807.5 nm. One can see from Fig. 1 that the peak intensity of the absorption line of these crystals is lower than that of the Nd³⁺: YAG crystal (at the same concentration of Nd^{3+}), but the line half-width is larger, being equal to 4 nm for Nd^{3+} :CNGG and Nd^{3+} :CLNGG crystals and to 1 nm for the Nd^{3+} : YAG crystal. To obtain the peak absorption intensity equal to that for the Nd^{3+} : YAG

Figure 1. Absorption spectra of Nd³⁺:YAG (a) and Nd³⁺:CNGG (b) crystals at different concentrations n_a .

crystal, the concentration of Nd^{3+} in CNGG and CLNGG crystals should be increased to 3%.

To find the optimal concentration of Nd^{3+} , one should study cross-relaxation of excitation from the ${}^{4}F_{3/2}$ level accompanied by nonradiative relaxation. For this purpose, we studied the decay of the ${}^4F_{3/2} - {}^4I_{9/2}$ luminescence of Nd^{3+} ions upon resonance excitation by 10-ns pulses at 880 nm.

Fig. 2 shows the kinetics of luminescence decay for three CNGG crystals doped with Nd^{3+} ions at different concentrations. The luminescence of the crystal with $n_a \sim$ 1% (10^{20} cm⁻³) decays exponentially in a broad dynamic range with the lifetime $237 \mu s$. At higher concentrations of Nd^{3+} , the luminescence decay curves exhibit the Förster decay.

Figure 2. Decay kinetics of the luminescence intensity I from the ${}^4F_{3/2}$ level of the Nd³⁺ ion in CNGG crystals at different concentrations n_a .

The cross-relaxation of garnet crystals doped with neodymium is well described by the Förster $-$ Dexter formula for the case of dipole-dipole interaction

$$
I = I_0 \exp\left[-\left(\frac{t}{\tau} + \gamma\sqrt{t} + w\right)\right],\tag{1}
$$

where I is the luminescence intensity; τ is the radiative lifetime; w is the probability of migration-limited relaxation; and γ is a parameter of the direct static interaction. The parameter γ , which can be determined from (1), gives the microparameter of the donor-acceptor interaction

$$
C_{\text{da}} = \frac{9\gamma^2}{16\pi^3 n_a^2}.\tag{2}
$$

However, because Nd^{3+} ions form centres of different types in Nd^{3+} : CNGG and Nd^{3+} : CLNGG crystals, strictly speaking, expression (2) cannot be applied to these crystals, although the decay curves at some concentrations are adequately described by expression (1). The microparameter C_{da} at the concentration $n_a = 3\%$ was calculated to be 4.5×10^{-41} cm⁶ s⁻¹, which strongly differs from its value 5.4×10^{-40} cm⁶ s⁻¹ for $n_a = 8\%$.

This can be explained by the fact that due to the disorder of the crystals under study, we deal with several interaction constants both inside groups of centres and between these groups, and the relation between concentrations of different groups of centres changes when the Nd^{3+} concentration is changed. Thus, the most objective parameter for estimating the cross-relaxation contribution is the decay kinetics integral $\int_s I dt$. The values of this integral show that the quantum yield of luminescence of the Nd^{3+} : CNGG crystal for $n_a = 3\%$ is approximately equal to 75% of that for the same crystal with $n_a = 1\%$.

We used the Nd^{3+} : CNGG and Nd^{3+} : CLNGG crystals of diameter 5 mm and lengths $L = 1, 1.5,$ and 2 mm in laser experiments. The crystals were pumped by a 2-W laser diode at 807-nm wavelength. The active element in the form of a plane-parallel disc, with an antireêection coating deposited on one of its sides and a selective mirror on its other side with the reflectivities $R_{1.06} = 99.9\%$ and $R_{0.8} < 10\%$, was placed in the focus of an optical system consisting of two lenses. The length of the optical cavity was 50 mm. The reflectivity of a plane output mirror was $R = 99.4\%$. The output parameters of cw lasing are presented in Fig. 3.

Figure 3. Dependence of the output lasing power P_{out} on the pump power P_p for active elements of different lengths.

The emission spectrum of the Nd^{3+} : CNGG and Nd^{3+} :CLNGG lasers operating in the cw mode using a nonselective cavity consists of two lines in the regions between $1055 - 1062$ and $1063 - 1066$ nm. The structure of these lines depends on the pump power. These data are consistent with our results obtained for the same lasers pumped by a flashlamp [\[7\]](#page-2-0) and diode-pumped Nd^{3+} : CNGG lasers [\[6\].](#page-2-0)

A large width of the absorption line of the Nd³⁺ ions in CNGG and CLNGG crystals results in stable output lasing parameters upon variation the laser diode temperature in the range $\pm 1.5^{\circ}$ C. This substantially alleviates the requirements imposed upon thermal stabilisation and the identity of the emission wavelengths of different diodes used in diode arrays. Our study has demonstrated that Nd^{3+} -doped CNGG and CLNGG crystals are promising as active elements for diode-pumped lasers.

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