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Laser performance of new crystal Cr^{3+}:(CeGd)$Sc_3(BO_3)_4$

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Abstract

Laser generation with Cr^{3+}:(CeGd)$Sc_3(BO_3)_4$ (Cr^{3+}:CSB) crystal is demonstrated for the first time to our knowledge. The spectroscopic properties of this crystal are similar to other tunable Cr^{3+}-doped crystals. A dye laser "Continuum ND6000" generating 10 ns pulses at 650 nm was used for pumping the CSB crystal in a hemifocal resonator. We measured threshold pump pulse energy of 2 mJ and a slope efficiency of 13%. In free-running operation we obtained an output pulse energy of 0.2 mJ centered at 877 ± 2 nm. We compare these results with Cr^{3+}:LiSAF laser performances and discuss the possible ways to obtain the continuous wave laser generation.

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Keywords: Laser generation; Solid state laser

Diode-pumped lasers based on a Cr-doped materials are very interesting alternative to the expensive and operation-heavy Ti:sapphire laser. Indeed, chromium-doped laser media on the one hand can be directly pumped by compact and powerful laser-diode sources in the wide spectral range 630–690 nm and, on the hand, their broad emission spectrum permits the wavelength tunability and ultrashort pulse generation.

1. Sample preparation and spectroscopic properties

A Cr^{3+}:(CeGd)$Sc_3(BO_3)_4$, (Cr^{3+}:CSB), single crystal was grown by the Czochralski method from the iridium crucible of 40 mm diameter in argon atmosphere at the temperature 1500 °C. Small slices of spectral graphite were placed in the growth chamber to make a slightly reduced atmosphere for obtaining of cerium III from CeO_2. The concentration of Cr^{3+} in the melt was 6.8×10^{19} cm^{-3}. Taking into account coefficient of segregation 1.1 we calculated the doping level of chromium in the crystal to be about 7.5×10^{19} cm^{-3}. There were no visible defects in the crystal but light scattering from a helium–neon laser beam

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was observed. A careful inspection of polished samples under the microscope revealed the presence of micro-cavities in the crystals. These micro-cavities are the main reason of the low optical quality of Cr^{3+}:CSB samples. We believe that this problem can hopefully be solved by the enhancement of the reducing atmosphere in the growth chamber.

In [1,2] we described the spectroscopic characteristics of chromium-doped CSB. The absorption and luminescence spectra are plotted in Fig. 1 for both polarizations. We have also characterized the upper-level lifetime thermal quenching and compared Cr^{3+}:CSB properties to fluoride crystal LiSAF [3]. It was shown that the decrease of the luminescence lifetime in CSB and LiSAF with good accuracy obeys to the equation $1/\tau = 1/\tau_r + W_{nr}$, where τ is the luminescence lifetime, $\tau_r = \tau_{r0}\exp(-\alpha T)$ is the radiative lifetime, τ_{r0} is the radiative lifetime at 0 K, $W_{nr} = s_0\exp(-E_{nr}/kT)$ is the probability of the non-radiative transition, s_0 is the frequency factor, E_{nr} is the activation energy for the non-radiative transition, T is the absolute temperature. The decrease of the luminescence lifetime in CSB is much slower in comparison with fluoride crystals in spite of the lower energy barrier E_{nr} for non-radiative transitions ($E_{nr} = 2050$ cm^{-1} for CSB and $E_{nr} = 5125$ cm^{-1} for LiSAF) (Fig. 2). The main reason for this phenomenon is related to a weak promotion interaction in Cr^{3+}:CSB crystals resulting in extremely low value of the frequency

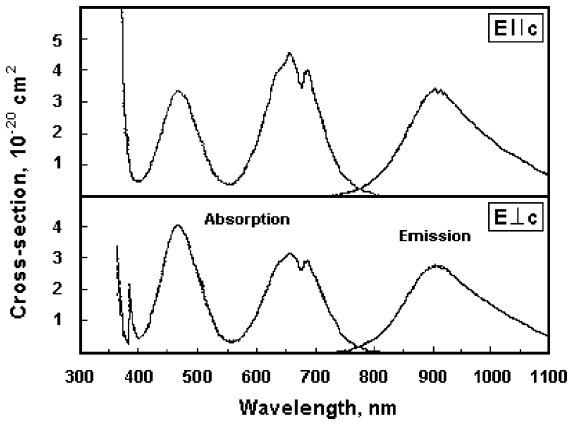

Fig. 1. Absorption and emission spectra of Cr:CSB for both polarizations.

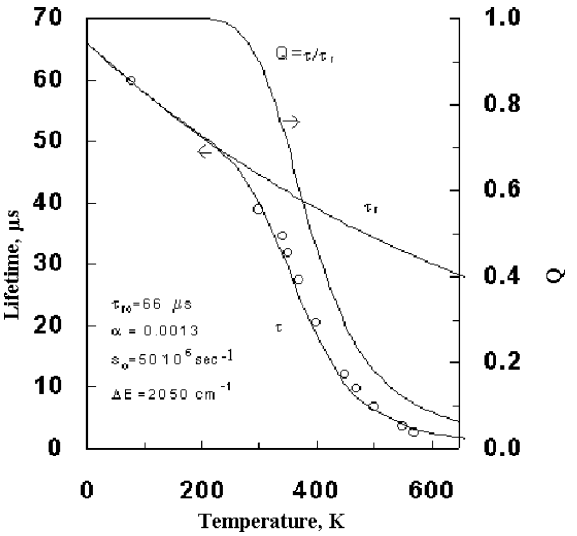

Fig. 2. Temperature dependence of Cr^{3+} lifetime and luminescence quantum output in Cr:CSB. Circles, experiment data; lines, theoretical dependences according to the equations: $1/\tau = 1/\tau_r + W_{nr}$, $\tau_r = \tau_{r0}\exp(-\alpha T)$, $W_{nr} = s_0\exp(-E_{nr}/kT)$, $Q = \tau/\tau_r$.

factor ($s = 5\times10^7$ s^{-1} for CSB and $s = 6\times10^{12}$ s^{-1} for LiSAF). As a result, at 300 K CSB luminescence lifetime is 40 μs (compared to less than 64 μs for the LiSAF) but it is still of 20 μs at 400 K, whereas LiSAF lifetime is no more than 4 μs. Thus the luminescence quantum efficiency of Cr^{3+}:CSB crystals is 45% higher at 400 K in comparison to LiSAF. This makes the Cr^{3+}:CSB crystal a particularly attractive laser medium for continuous wave laser.

2. Experimental

We used a 4.1 mm long Cr^{3+}:CSB coated for high reflectivity in the 850–900 nm range and high transmission at 670 nm on the pump side and antireflection coated in the 850–900 nm range on the opposite side. A hemi focal resonator of 17 mm length was completed with a spherical output coupler of 50 mm curvature radius, with $T = 1\pm0.25\%$ in the above spectral range. Two pump regimes were used to obtain the laser generation – continuous wave and pulse pumping.

As we expected all our attempts to obtain a continuous wave laser generation with a fiber coupled high brightness laser-diode pump (2.3 W at 665 nm, 116 μm fiber core, N.A. = 0.22) failed. We attributed this to the high passive losses due to the above-discussed crystal defects implying a much higher threshold power. Another possible origin of the very high threshold for the continuous wave generation may be an excited state absorption. At present, we have no reliable data about the latter but this work is in progress. Since we used a pulsed pumping by the dye laser "Continuum ND6000".

The crystal was longitudinally pumped with 10 ns pulses at 650 nm generated by the dye laser. The pump beam was focused with a spherical lens ($f = 1000$ mm) to the beam waist of 230 μm and we measured its absorption to be equal to 82%. The experiment is schematically represented in Fig. 3. The pulse energy of the pump laser at 650 nm was 32 mJ. A system of standard neutral filters without antireflection coatings was used to control the pump energy. By changing these filters before the laser cavity, we could increase step by step the pumping energy and measure the corresponding output laser energy. The energy was averaged over 10 pulses by the NOVA laser energy meter equipped with PE10 pyroelectric head from OPHIR Optronics, Inc. In Fig. 4, the dependence of the output laser pulse energy as a function of the absorbed pump energy is plotted. We calculated the threshold pulse energy of 2 mJ and a slope efficiency of 13%.

The output laser pulse shape was observed using a digital oscilloscope Tektronix TDS3052 (500 MHz bandwidth, 5 Gs/s) and a high-speed photo

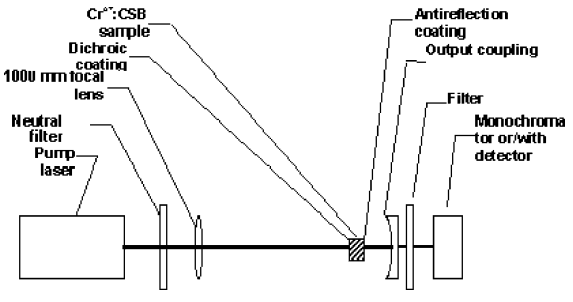

Fig. 3. Scheme of laser experiment.

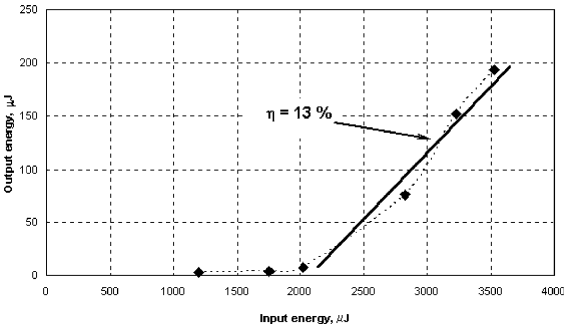

Fig. 4. Output pulse energy versus absorbed energy for Cr^{3+}:CSB pumped by 10 ns, 650 nm pulses in the hemifocal cavity.

detector THORLABS DET-210 (bandwidth > 100 MHz). The forms of laser pulses measured at different pumping pulse energies are shown in Fig. 5. FWHM was about 150–200 ns. The emission wavelength was measured with a monochromator M25-HUET and it was found to be 877 ± 2 nm. As it follows from Fig. 5, the laser pulse arises with a delay about 200–600 ns with respect to a pumping pulse.

The fact that the laser pulse is delayed with respect to a pumping pulse is in a good agreement with observed buildup of luminescence. Namely, after a 100 ns pump pulse at 650 nm one could observe a buildup of luminescence with characteristic time about 0.5–1.5 μs. The phenomenon of the luminescence buildup is a temperature dependent. In the framework of barrier model of nonradiative energy transfer interaction between levels ^2E and $^4T_2$ described by means of system of balance equations we have estimated the energy

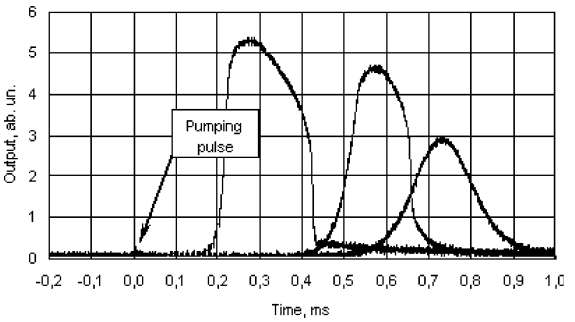

Fig. 5. Pulse shape of Cr^{3+}:CSB laser after 650 nm laser pumping pulse, 10 ns duration.

barrier $^2E \rightarrow {}^4T_2$ as $E_{ET} = 337$ cm^{-1} and the frequency factor $\omega_0 = 20 \times 10^6$ s^{-1} [2].

According to our numerical simulations, after the excitation pulse at 650 nm the significant part of excitations appears at the level ^2E due to the overlapping between ^2E and $^4T_2$ absorption bands. The relaxation of excitations from ^2E level to $^4T_2$ level at room temperature occurs rather slowly, mainly due to the small frequency factor. This slow process controls the buildup process and the delay of the pulse of laser oscillation. Thus the properties of Cr:CSB crystals are unusual – the thermal equilibrium between ^2E and $^4T_2$ levels just as the thermal bleaching of level $^4T_2$ occur slowly with anomalous low frequency factor which is lower than in traditional crystals of oxides and fluorides roughly in 10^5 times. The physical sense of the frequency factor can be seen from the formula $W_{nr} = s_0 \exp(-E_{nr}/kT)$. It is clear that the frequency factor is a probability of transition at high temperatures when energy barrier does not play a role already. The lowering of transitions probabilities in 10^3–10^6 times can be considered as a forbidding of the transitions. Thus the non-radiative transitions in Cr^{3+} impurity centers in borate crystals are forbidden. We suppose that there is a deficient of promotion type of lattice vibration in Cr^{3+}:CSB crystals. The origin of this phenomenon lies into the structural features of CSB, which belongs to the pronounced heterodesmic type of crystals [2].

Unfortunately, we could not obtain the laser generation from Cr^{3+}:CSB crystals in the pulse regime with another output coupling efficiency ($T = 2\%$ and 3% were available in our laboratory) because of the damage of the entrance face dichroic coating. The laser damaging LiSAF was the same obstacle to increase the pump power above 4 mJ. One can suppose that the above-mentioned micro-cavities deteriorate the quality of the optical coating. To verify this idea, we scanned the pump beam across the input surface to get the optimum of the output power and/or the threshold, but we could not obtain conclusive results. We may reasonably suppose that, once the problem of microcavities in the Cr^{3+}:CSB crystals is solved, it will be possible to obtain a higher slope efficiency.

3. Conclusion

In the light of the data presented above, it is possible to conclude that the Cr:CSB crystals is a promising laser medium but it needs a further investigations on the growth parameters. We are now heading to work on the improvement of optical quality of the matrix, and on the optimization of the activator concentration to obtain good quality crystals. At the moment the investigations of the excited state absorption are in progress.

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