Continuous wave $Pr^{3+}:BaY_2F_8$ and $Pr^{3+}:LiYF_4$ lasers in the cyan-blue spectral region

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We report on the first continuous wave quasi 3-level lasers emitting in the cyan-blue spectral range in praseodymium doped crystalline materials. Applying $Pr^{3+}:BaY_2F_8$ as active medium, up to 201 mW of output power at 495 nm could be obtained with a slope efficiency of 27% under pumping with an optically pumped semiconductor laser (2ω-OPSL) at 480 nm. In the same pumping scheme, utilizing $Pr^{3+}:LiYF_4$ output powers up to 70 mW were realized at 491 nm and 500 nm, respectively. With $Pr^{3+}:BaY_2F_8$ also diode pumped laser operation with up to 11% slope efficiency and 44 mW output power was achieved. In the latter case, detailed investigations on the temperature dependency of the laser output were conducted. Moreover, comparative experiments were carried out for the first time with green emitting $Pr^{3+}:BaY_2F_8$ lasers at 524 nm and 553 nm both under diode and 2ω-OPSL excitation.

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During the last decades the trivalent praseodymium ion has been established as the most promising active ion for the direct generation of visible laser radiation in solids. Efficient continuous wave (cw) laser operation could be demonstrated especially in fluoride crystals like $LiYF_4$ [1], $LiLuF_4$ [2], KY_3F_{10} [3], and BaY_2F_8 [4] at wavelengths covering wide parts of the green, orange, red, and dark red spectral regions [5]. Oxide crystals like $YAlO_3$ [6] and $SrAl_{12}O_{19}$ [7], allowing for visible efficient laser oscillation, have been investigated as well. These lasers were all operated in four level schemes, which, due to the high emission cross sections of Pr^{3+} of up to 10^{-19} cm^2, allowed for very low laser thresholds. However, the four level laser scheme has an intrinsically lower Stokes efficiency and thus a lower maximum achievable overall efficiency compared to lasers operated in a three or quasi three level scheme. Furthermore, the energetic gap between the two uppermost and lowermost energy levels is typically bridged by phonons. This leads to a stronger heating of the active medium. Therefore, praseodymium doped three level lasers appear interesting. In fact, first investigations on visible emitting praseodymium three level lasers, conducted by Esterowitz *et al.* [8], date back to 1977. In these experiments $LiYF_4$ was applied as host material. At that time, pulsed dye lasers were typically applied as excitation sources. Their high fluence allowed for high inversion levels despite the short lifetime of the emitting ^3P_J manifold of only several tens of μs. Thus, the resulting Pr^{3+}-lasers were pulsed and emitted on the zero phonon line around 480 nm, which typically provides the highest emission cross section in the cyan-blue part of the spectrum. In this way, laser operation could be demonstrated in a variety of host materials. Beside the $Pr^{3+}:LiYF_4$ laser, which was operated at room temperature [8], Malinowski *et al.* enlarged the range of suitable host crystals with $Y_3Al_5O_{12}$, $YAlO_3$, and other materials at cryogenic temperature [9,10]. Cw three level laser oscillation of trivalent praseodymium at room temperature was so far limited to doped fluoride glass fibers [11,12].

In this letter we report on our first results on cw cyan-blue laser operation of praseodymium doped BaY_2F_8 (BYF) and $LiYF_4$ (YLF) crystals at room temperature. To operate such lasers on the zero phonon line, very high inversion levels of approximately 50% are necessary, requiring – together with the short effective lifetime of the emitting Pr^{3+}-level – highly intense excitation sources. In contrast, transitions terminating in energetically higher lying Stark levels of the $^3H_4$ ground state are less affected by reabsorption. This should allow for ground state laser operation at comparatively low inversion levels and thus reasonable threshold pump powers for cw laser operation.

As host material we chose on the one hand side YLF, which – also due to the high quality of the available crystals – allowed for the highest slope efficiencies of any praseodymium laser so far [13-15].

On the other hand side, the Pr:BYF crystal seems, due to the presence of a comparatively strong emission peak at 495 nm, to be a more suitable gain medium for three level laser operation with low laser threshold [16,17]. The effective lifetimes and ground state absorption cross sections of these crystals at comparable doping concentrations are very similar. Therefore it is sufficient to compare their gain spectra to predict their suitability for laser operation into the ground state manifold. Thus, we first determined emission cross sections σ_{em} applying the Füchtbauer-Ladenburg method [18] and absorption cross sections σ_{abs} from transmission spectra of a sample with a known dopant concentration. From these spectra, gain spectra of Pr^{3+} doped BYF and YLF were calculated according to

$$\sigma_{\text{gain}} = \beta\sigma_{\text{em}} - (1-\beta)\sigma_{\text{abs}} \qquad (1)$$

which are depicted in figure 1 for different orientations of the electric field vector E and realistic inversion levels β. Although gain occurs at comparable inversion levels in both systems, at higher inversions the gain cross sections of Pr:BYF, in particular for $\boldsymbol{E}$ || X orientation, clearly exceed those of Pr:YLF. For both materials the effective absorption and emission cross sections σ_{abs} and σ_{em} for suitable pump or laser transitions are listed in Tab. 1. In addition to the cross sections resulting from direct transitions between the ground state multiplet $^3H_4$ and the laser emitting thermally coupled ^3P_J, $^1I_6$ multiplets, emission cross sections for transitions terminating in the $^3H_5$ multiplet are listed for BYF as well. Green emitting lasers oscillating on these transitions are well suited to serve as comparison to the cyan-blue lasers, since they provide short emission wavelength as well, cross sections in the same scale, but are operated in a four level scheme. Appropriate lasers in YLF are documented in the literature [14,15].

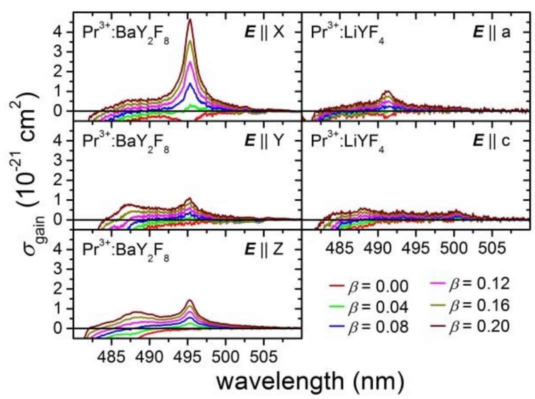

Fig. 1. Inversion dependent gain cross sections for transitions into the ground state manifold in Pr:BYF and Pr:YLF for different orientations of the electric field vector $\boldsymbol{E}$ and varying inversion levels β.

The setup for the laser experiments is depicted in Fig. 2. The nearly hemispheric cavity consisted of two mirrors M1 and M2. All mirrors applied as M1 were plane and always served as the input coupling mirror. Due to the small separation between pump and laser wavelength, the different plane input coupling mirrors M1 also exhibited a certain degree of transmission for the laser wavelength in most configurations (output 1 in Fig. 2). The end mirror M2 had a radius of curvature of 50 mm and was placed in a distance of 48 mm from M1. Unless otherwise noted, the output powers in this work refer to the sum of output 1 and 2 in Fig. 2. As pump sources either an InGaN laser diode (LD) with approximately 1 W of output power, emitting at a wavelength of 445 nm, or optically pumped frequency doubled semiconductor lasers (2ω-OPSL) with up to 5 W output power around 480 nm were applied. Table 2 summarizes the properties of the available pump sources in more detail. The pump light always passed a $\lambda/2$-wave plate followed by a polarizing beam splitter cube, which in case of the laser diodes was used for setting the launched pump power while keeping the beam shape and the emission wavelength constant.

Table 1. Transition cross sections of Pr^{3+} between the $^3H_4$ and the ^3P_J multiplets when doped into the BYF and YLF matrices. The transitions in BYF resulting in wavelengths longer than 500 nm terminate in the $^3H_5$ multiplet (4-level scheme).

host	wavelength (nm)	σ_{abs} (10^{-20} cm^2)	σ_{em} (10^{-20} cm^2)	polarization
BYF	445	5.9	-	$\boldsymbol{E}$ | | Y
	480	8.1	9.2	$\boldsymbol{E}$ | | Y
	495	0.1	2.6	$\boldsymbol{E}$ | | X
	524	-	1.0	$\boldsymbol{E}$ | | Y
	553	-	1.5	$\boldsymbol{E}$ | | Y
YLF	444	8.8	-	$\boldsymbol{E}$ | | c
	479	17.0	19.4	$\boldsymbol{E}$ | | c
	491	0.1	0.6	$\boldsymbol{E}$ | | a
	500	-	0.2	$\boldsymbol{E}$ | | c

Moreover, the beam splitter cube allowed for a separation of the laser output 1 from the pump for all lasers except the 500 nm laser in Pr:YLF, where the pump and laser beams exhibit the same polarization. In case of the four level lasers this was not necessary because all the output power was delivered by output 2.
To align the polarization of the excitation beam parallel to the laser crystal axis providing the highest absorption cross section, a second $\lambda/2$-wave plate was inserted into the pump beam. In all experiments the pump source was focused into the active medium by a lens with a focal length of 40 mm. Accounting for the beam shape before the focusing lens, the pump spot diameters inside the active medium were estimated to be approximately 50 µm in case of 2ω-OPSL pumping and about 90 µm when InGaN laser diodes were applied as pump sources.

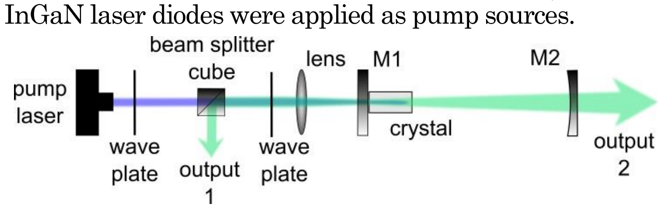

Fig. 2. Schematic of the laser setup. Depending on the combination of the mirrors M1 and M2 up to two output channels were present.

We prepared two uncoated samples according to the parameters listed in Tab. 3. Independent of the pump source, the small signal single pass absorption of the Pr:BYF crystal was approximately 70%. The Pr:YLF crystal absorbed about 90% of the pump light in a single pass. The samples were mounted in water cooled copper heat sinks, allowing for defined temperatures between 3 °C and ambient temperature. For the determination of the slope efficiency η_{sl} the absorbed fraction of the pump power at the laser threshold was determined.

Table 2. Emission wavelengths λ_{em}, beam quality factor M^2, maximum output power P_{max} and abbreviations of the applied pump sources.

λ_{em} (nm)	M^2	P_{max} (W)	abbreviation
Frequency doubled optically pumped semiconductor lasers:			
480	< 1.1	4.0	OPSL1
479	< 3	5.0	OPSL2
InGaN-laser diode:			
445	< 5	1.1	LD1

This fraction should remain constant above threshold enabling the estimation of the absorbed pump power during laser operation. Every absorbed pump power given in this letter furthermore accounts for a second pass of the pump beam which results from the back reflection of transmitted pump light at M2.

First the experiments under 2ω-OPSL-pumping were conducted. The crystal temperature was kept at 10 °C. Due to the low separation between the pump wavelength around 480 nm and the laser wavelengths between 491 nm and 500 nm, the requirements for the mirror coatings in particular for M1 are very strict. Therefore, the choice of available mirrors did not allow for an optimization of the output coupling. Laser operation was obtained with both crystals. The best results for each wavelength are depicted in Fig. 3.

Table 3. Parameters of the employed crystals.

host	doping level (at.% / 10^{19} cm^{-3})	polished plane	length (mm)	aperture (mm^2)
BYF	0.5 / 6.5	X-Y	2.7	5 × 5
YLF	0.5 / 7.0	a-c	3.0	1 × 1

For Pr:BYF we obtained laser operation at 495 nm under pumping with OPSL1 (see Tab. 2). In this case, an output power as high as 201 mW was obtained when the minimum total available output coupling transmission of T_{OC} = 0.4% was used. The laser had a slope efficiency of 27%. Increasing T_{OC} to 1.0% resulted in higher thresholds, decreased slope efficiencies, and an earlier onset of thermal problems. The maximum slope efficiency appears low compared to those of typical four level praseodymium lasers in fluoride hosts. Therefore, we recorded the slope efficiency at wavelengths of 524 nm and 553 nm. The resulting maximum values of 22% and 28%, respectively at output couplings of about 1% is low compared to the values of 72% and 60% we recently obtained for the corresponding transitions in Pr:YLF in the same pumping scheme [15], which might point towards a low crystal quality of the available Pr:BYF sample.

For Pr:YLF the laser threshold could be reached for two transitions at 491 nm and 500 nm under excitation with OPSL2 (see Tab. 2). The laser performance of the Pr:YLF lasers fell short of that of the Pr:BYF lasers by a factor of almost three. Both the maximum output powers at 491 nm and 500 nm were limited to about 70 mW at slope efficiencies of 7% and 6%, respectively. It should be noted that the laser at 500 nm could, due to the transmission characteristics of M1, be operated at the lowest output coupling of T_{OC} < 0.1% of all lasers presented in this letter. Moreover, it emitted in the same polarization as the pump light. Therefore, our setup did not allow for a determination of the power at output 1 which might have been in the same order as the power at output 2. For both wavelengths no improvement could be achieved by applying other output coupling transmissions.

Increasing the crystal temperature from 10 °C to room temperature led to decreased performance but laser operation could still be observed. Moreover, all lasers presented in Fig. 3 suffered a thermal roll over when the pump power was increased further.

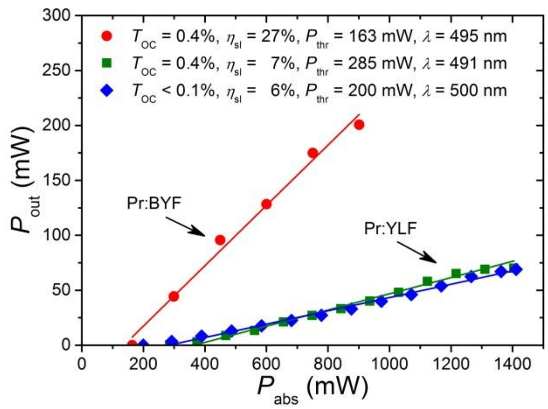

Fig. 3. Input-output characteristics of the investigated 2ω-OPSL pumped cyan-blue Pr^{3+} lasers. The cooling water had a temperature of 10 °C in all cases. The straight lines remark linear fits which were used to determine the slope efficiencies of the lasers.

Since the laser thresholds obtained under 2ω-OPSL excitation are within reach of available 1 W InGaN laser diodes, we also performed laser experiments under pumping with LD1 (see Tab 2). However, we only obtained diode pump laser operation with the Pr:BYF sample. Laser operation in Pr:YLF was probably impeded by the lower beam quality of the laser diode compared to the OPSL, hindering to reach the same inversion densities with the available pump power. Figure 4 depicts the results obtained from the diode pumped Pr:BYF laser at 495 nm. Also in this case the highest efficiency and output power were obtained at low output coupling transmission of 0.4%. At a crystal temperature of 10 °C a slope efficiency of 11% allowed to extract 44 mW from the resonator. The laser threshold was 200 mW and thus comparable to the experiments under OPSL pumping. At an increased output coupling of 1% the slope efficiency dropped to about 6% while the threshold pump power P_{thr} increased to 236 mW.

To characterize the temperature dependence of the laser performance, we conducted laser experiments at different crystal temperatures between 6 °C and 25 °C for T_{OC} = 1%. The resulting curves are also depicted in Fig. 4. The impact of the temperature on the threshold and the slope efficiency can be seen in the inset in Fig. 4. The increasing laser threshold results from an increased reabsorption due to a changing Boltzmann occupation of the Stark levels of the $^3H_4$ ground state. In contrast, from simple rate equations a decreased slope efficiency with increasing temperature is not expected. Within a temperature range of only 19 °C it is lowered by a factor of almost 2. Further investigations will be needed to explain this behavior. Under the same pumping conditions the lasers at 524 nm and 553 nm delivered maximum output powers of 45 mW and 95 mW, at slope efficiencies of 14% and 26%, respectively at an output coupling transmission of approximately 1%.

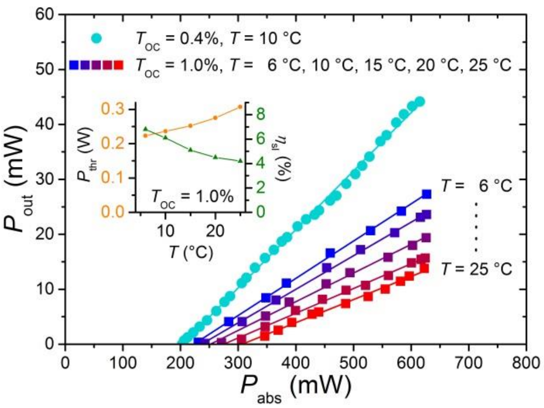

Fig. 4. Input-output curves for diode pumped Pr:BYF quasi three level lasers at 495 nm for different temperatures T and output coupler transmissions T_{OC}. The inset shows the threshold pump power P_{thr} and slope efficiency η_{sl} vs. temperature T. The straight lines remark linear fits which were used to determine the slope efficiencies of the lasers.

In a last experiment, wavelength tuning of the Pr:BYF ground state laser was carried out under diode excitation. For that purpose the laser cavity was slightly changed to a folded configuration, such that it provided an almost collimated beam section in which a 1.5 mm thick quartz crystal was introduced under Brewster's angle to serve as wavelength filter. In this configuration the cw output could be tuned within a total of 1.6 nm around 495 nm with a maximum output power of 13 mW at the center wavelength. The emission bandwidth of the laser was < 0.3 nm in these experiments.

In summary we presented the first cw quasi three level lasers based on Pr^{3+} doped crystalline materials. In case of Pr^{3+}:BYF laser operation was obtained both under laser diode and 2ω-OPSL excitation. A maximum output power of 201 mW was measured at a slope efficiency of 27%. Laser operation into the ground state manifold of Pr:YLF was so far limited to 2ω-OPSL excitation but the inversion level at threshold should in principle be achievable under diode excitation as well. The slope efficiencies of the lasers presented in this letter are lower than those of typical four level praseodymium lasers in fluoride hosts. While the crystal quality of the Pr:YLF sample used in the experiments is excellent, the quality of the Pr:BYF sample seems to be insufficient for obtaining highest possible efficiencies. However, in contrast to Pr:YLF, the cyan-blue lasers in Pr:BYF under 2ω-OPSL excitation showed comparable efficiencies as the green emitting four level laser at 553 nm. Since these efficiencies are only about a third of what is possible with high quality Pr:YLF crystals, a high potential for further improvement of the efficiencies of the Pr:BYF lasers can be expected in optimizing the crystal quality. From this point of view and comprising the spectroscopic differences of both systems, Pr:BYF seems more promising to reach high efficiencies in laser operation in the cyan-blue spectral range in future.

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