Visible laser operation of Pr^{3+}-doped fluoride crystals pumped by a 469 nm blue laser

Bin Xu,[1,2] Patrice Camy,[1,*] Jean-Louis Doualan,[1] Zhiping Cai,[2] and Richard Moncorgé[1]

[1]*Centre de Recherche sur les Ions, les Matériaux et la Photonique (CIMAP) UMR CEA-CNRS-ENSICaen, Université de Caen, 14050 Caen, France*

[2]*Department of Electronic Engineering, Xiamen University, Xiamen 361005, People's Republic of China*

**patrice.camy@ensicaen.fr*

Abstract: We report continuous-wave (CW) laser operation of Pr^{3+}-doped $LiLuY_4$, $LiYF_4$ and KY_3F_{10} single crystals in the Red, Orange and Green spectral regions by using a new pumping scheme. The pump source is an especially developed compact, slightly tunable and intracavity frequency-doubled diode-pumped Nd:YAG laser delivering a CW output power of 0.9W at 469.12 nm. At this pump wavelength, efficient room temperature laser emissions corresponding to the $^3P_0 \rightarrow {}^3F_2$, $^3P_0 \rightarrow {}^3H_6$ and $^3P_1 \rightarrow {}^3H_5$ Pr^{3+} transitions are observed. While a maximum slope efficiency of 45% is obtained in the red with Pr:$LiYF_4$, the demonstration is made for the first time of the orange laser operation of Pr:KY_3F_{10} at about 610 nm.

OCIS codes: (140.5560) Pumping; (140.5680) Rare earth and transition metal solid-state lasers; (140.7300) Visible lasers.

References and links

1. R. Moncorgé, L. D. Merkle, and B. Zandi, "UV-visible lasers based on rare-earth ions," MRS Bulletin **24**(9), 21–26 (1999).
2. L. Esterowitz, R. Allen, M. Kruer, F. Bartoli, L. S. Goldberg, H. P. Jenssen, A. Linz, and V. O. Nicolai, "Blue light emission by a Pr:$LiYF_4$-laser operated at room temperature," J. Appl. Phys. **48**(2), 650–652 (1977).
3. A. A. Kaminskii, A. I. Lyashenko, N. P. Isaev, V. N. Karlov, V. L. Pavlovich, S. N. Bagaev, A. V. Butashin, and L. E. Li, "Quasi-cw Pr^{3+}:$LiYF_4$ laser with $\lambda = 0.395$ μm and an average output power of 2.3 W," Quantum Electron. **28**(3), 187–188 (1998).
4. T. Sandrock, T. Danger, E. Heumann, G. Huber, and B. T. H. Chai, "Efficient continuous-wave laser emission of Pr^{3+}-doped fluorides at room temperature," Appl. Phys. B **58**(2), 149–151 (1994).
5. J. M. Sutherland, P. M. W. French, J. R. Taylor, and B. H. T. Chai, "Visible continuous-wave laser transitions in Pr(3+):YLF and femtosecond pulse generation," Opt. Lett. **21**(11), 797–799 (1996).
6. A. Richter, E. Heumann, E. Osiac, G. Huber, W. Seelert, and A. Diening, "Diode pumping of a continuous-wave Pr3+-doped LiYF4 laser," Opt. Lett. **29**(22), 2638–2640 (2004).
7. P. Camy, J. L. Doualan, R. Moncorgé, J. Bengoechea, and U. Weichmann, "Diode-pumped Pr($^{3+)}$:KY$_{(3)}$F$_{(10)}$ red laser," Opt. Lett. **32**(11), 1462–1464 (2007).
8. F. Cornacchia, A. Di Lieto, M. Tonelli, A. Richter, E. Heumann, and G. Huber, "Efficient visible laser emission of GaN laser diode pumped Pr-doped fluoride scheelite crystals," Opt. Express **16**(20), 15932–15941 (2008).
9. J. Nakanishi, T. Yamada, Y. Fujimoto, O. Ishii, and M. Yamazaki, "High power red laser oscillation of 311 mW in Pr^{3+} doped waterproof fluoro-aluminate glass fibre excited by GaN laser diode," Electron. Lett. **46**(18), 1285–1286 (2010).
10. A. Richter, E. Heumann, G. Huber, V. Ostroumov, and W. Seelert, "Power scaling of semiconductor laser pumped Praseodymium-lasers," Opt. Express **15**(8), 5172–5178 (2007).
11. E. Heumann, C. Czeranowski, T. Kellner, and G. Huber, "An efficient all-solid-state Pr3+:LiYF4 laser in the visible spectral range," Technical Digest CLEO'99, paper CTuG1, p86, (Baltimore, 1999).
12. C. Czeranowsky, E. Heumann, and G. Huber, "All-solid-state continuous-wave frequency-doubled Nd:YAG-BiBO laser with 2.8-W output power at 473 nm," Opt. Lett. **28**(6), 432–434 (2003).
13. F. Jia, Q. Xue, Q. Zheng, Y. Bu, and L. Qian, "5.3W deep-blue light generation by intra-cavity frequency doubling of Nd:$GdVO_4$," Appl. Phys. B **83**(2), 245–247 (2006).
14. A. A. Kaminskii, *Crystalline lasers: Physical Processes and Operating Schemes*, (CRC Press, Florida, 1996).
15. F. Mougel, G. Aka, A. Kahn-Harari, H. Hubert, J. M. Benitez, and D. Vivien, "Infrared laser performance and self-frequency doubling of Nd:GdCOB," Opt. Mater. **8**(3), 161–173 (1997).
16. A. Lupei, E. Antic-Fidancev, G. Aka, D. Vivien, P. Aschehoug, Ph. Goldner, F. Pelle, and L. Gheorghe, "Spectroscopic and crystal field studies of Nd^{3+} in GdCOB and YCOB," Phys. Rev. B **65**(22), 224518 (2002).

17. C. Q. Wang, Y. T. Chow, D. R. Yuan, D. Xu, G. H. Zhang, M. G. Liu, J. R. Lu, Z. S. Shao, and M. H. Jiang, "CW dual wavelength Nd:YAG laser at 946 and 938.5nm and intracavity nonlinear frequency conversion with a CMTC crystal," Opt. Commun. **165**(4-6), 231–235 (1999).
18. S. Bjurshagen, D. Evekull, and R. Koch, "Generation of blue light at 469 nm by efficient frequency doubling of diode pumped Nd:YAG laser," Electron. Lett. **38**(7), 324 (2002).
19. B. Xu, P. Camy, J. L. Doualan, Z. P. Cai, F. Balembois and R. Moncorgé, "Efficient three wavelengths blue laser generation by simultaneous frequency-doubling and sum-frequency mixing in a diode-pumped Nd:YAG laser cavity," (submitted)
20. D. Findlay, and R. A. Clay, "The measurement of internal losses in 4-level lasers," Phys. Lett. **20**(3), 277–278 (1966).

1. Introduction

It has been known since a long time that Pr^{3+} doped crystals and glasses and, among them, Pr^{3+} doped fluorides [1], thanks to more efficient radiative transitions, can be used to produce multicolor laser emissions for RGB (Red, Green, Blue) applications.

Still few years ago, however, the use of such laser active materials was somewhat limited by the lack of efficient and/or practical pump sources in the blue region. For example, the first Pr-laser, based on a Pr:YLF crystal, was pumped by a pulsed dye laser at 444 nm [2]. Then, demonstration was made of pulsed laser operation of the same Pr:YLF laser material under flash-lamp pumping [3], as well as of CW and/or femtosecond laser operation of Pr:YLF by pumping with an Argon ion laser [4,5].

In fact, the situation has improved a lot these last years thanks to significant progresses in the development of pump sources such as GaN laser diodes and optically-pumped and frequency-doubled semiconductor lasers (OPSLs) operating in the same spectral range as specific Pr^{3+} absorption peaks (see in the Fig. 1), and thanks to a series of very encouraging results with these pump sources. Laser operation of several Pr-doped crystals and glasses, including those already mentioned, has been demonstrated, for example, by pumping around 444 nm, with a GaN laser diode, into the $^3H_4 \rightarrow {}^3P_2$ absorption band of the materials [6–9]. Several interesting results were also reported by pumping the same materials directly into the $^3P_0$ emitting level ($^3H_4 \rightarrow {}^3P_2$ absorption transition) by using an especially developed intracavity frequency-doubled OPSL operating around 477 nm [10].

So, all-solid visible lasers based on Pr^{3+}-doped crystals and glasses are motivating again much attention and their utilization in the new generation of color displays, for data storage techniques and medical imaging is becoming a reality.

Nevertheless, the GaN semiconductor diodes, although offering the most compact and simple solution for the development of such lasers, are still limited to about 1W (without any improvement in the last two years) and their beam quality is seriously degraded for powers higher than 500 mW. On the other hand, frequency-doubled OPSLs, although offering the possibility of higher pump powers with a better beam quality, remain on-demand, non-commercially available thus rather expensive laser devices.

This is the reason why we have examined a third (alternative) solution, which is the purpose of the present communication, which consists in pumping the materials (see in the Fig. 1) around 469 nm with the aid of an easier to built, efficient and potentially powerful solid-state laser based on a diode-pumped and intracavity frequency-doubled 3-level Nd-doped crystal such Nd:YAG. This new pumping configuration, which addresses the $^3H_4 \rightarrow {}^3P_1$, $^1I_6$ absorption transition of the Pr^{3+} ions, has not been exploited so much, except by using an Argon ion laser at 465.8 nm [4], thus far from the considered absorption peak(s), and also by using a frequency-doubled Nd:YAG laser, but at 473 nm, thus again far from the absorption peak [11].

So, in this paper, we present a series of laser results obtained in the Red, Orange and Green spectral regions by pumping Pr^{3+}:$LiLuF_4$ (Pr:LLF), Pr^{3+}:$LiYF_4$ (Pr:YLF) and Pr^{3+}:KY_3F_{10} (Pr:KYF) crystals with the aid of an home-made and especially developed diode-pumped and intracavity frequency-doubled Nd:YAG laser with improved laser performance.

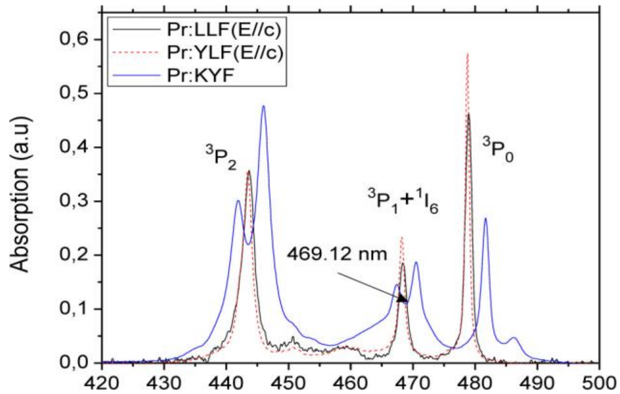

Fig. 1. Room temperature absorption spectra of Pr:LLF, Pr:YLF and Pr:KYF in the blue spectral range.

2. Experimental conditions

2.1 Pump source characteristics

Several blue solid-state laser sources based on the frequency-doubling of the infrared three-level laser emission transitions of Nd^{3+} around 900-950 nm have been reported in the recent years with output powers up to several Watts [12,13]. These laser sources, however, have been operated either around 473 nm [12] or 456 nm [13], thus at laser wavelengths where the Pr^{3+} absorption is very weak. Indeed, as it is shown in the Fig. 1 for Pr:YLF and Pr:KYF, but which remains true for most of the Pr-doped materials, the $^3H_4 \rightarrow {}^3P_1$, $^1I_6$ absorption transition have peaks between about 467 and 470.5 nm, which means frequency-doubling (SHG) of infrared laser emissions ranging between 934 and 941 nm, thus around 938 nm. The Nd-doped materials which could be used to obtain such a suitable lasing wavelength have been identified and are reported in the Table1.

Table 1. Nd^{3+} doped materials for infrared laser emission around 938 nm.

Nd^{3+} laser materials	$^4I_{9/2}(cm^{-1})$ Z1~Z5	$^4F_{3/2}(cm^{-1})$ R1, R2	$\Delta E(cm^{-1})$, Wavelength(nm)	References
YSGG {$Y_3Sc_2Ga_3O_{12}$}	0,97,168,252,778	11439,11489	10661, 938	[14] p137
YGG {$Y_3Ga_5O_{12}$}	0,79,179,246,784	11443,11478	10659, 938.2	[14] p138
YAG {$Y_3Al_5O_{12}$}	0,130,199,308,857	11427,11512	10651, 938.5	[14] p133
GGG {$Gd_3Ga_5O_{12}$}	0,93,178,253,772	11442,11485	10670, 937.2	[14] p149
GdCOB {$Ca_4GdO(BO_3)_3$}	0,72,302,441,662	11344,11539	10682, 936.2	[15]
YCOB {$Ca_4YO(BO_3)_3$}	0,73,300,451,680	11347,11537	10667, 937.5	[16]

Because of the wavelength matching but also because of the possibility to get cheap and commercially available crystals, the choice has been made, for this first approach, of exploiting the $^4F_{3/2}$ (R_2) $\rightarrow$ $^4I_{9/2}(Z_5)$ laser emission transition of the Nd:YAG laser system at 938.5 nm. Diode-pumped CW laser operation at this wavelength was first published by Koch et al [17] and extra- as well as intra-cavity frequency-doubling at 469 nm was demonstrated by Bjurshagen et al [18], but with a maximum output power of 200 mW, by using a PPKTP doubler and a Z-type laser resonator.

The laser used here for pumping our Pr-doped crystals is an optimized version which allowed us to reach nearly 1W at 469 nm. It is based on a V-shaped cavity in which diode-pumped intra-cavity SHG is achieved with a properly oriented LBO nonlinear crystal and the required emission transition at 938 nm is selected with the aid of an (glass plate) etalon. More details are given in a separated paper [19] in which we consider both intracavity frequency-doubling (SHG) and sum-frequency mixing. By adjusting the inserted glass plate and the phase matching angle of the LBO crystal, such a system allows us to tune the laser wavelength from about 469.05 to 469.34 nm and to get corresponding output powers of 800 up to 930 mW. Figure 2 shows this blue laser output power for different lasing wavelengths. In order to obtain a maximum absorbed pump power in the Pr-doped samples, we have set the pump wavelength at 469.12 nm, and maintained an output power of about 910 mW.

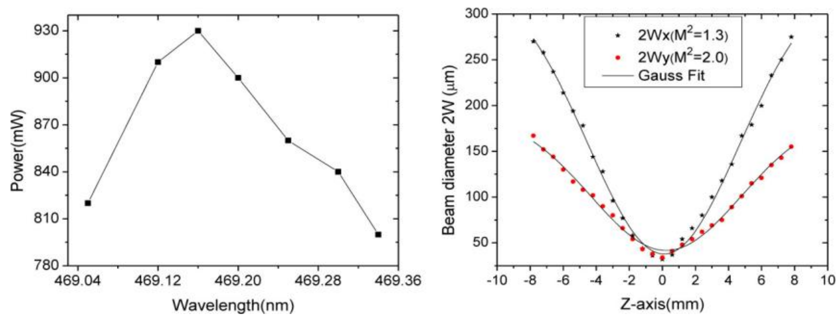

Fig. 2. Output power of the blue Nd:YAG pump laser versus laser wavelength (left) and beam waist diameter near the focal point (right) in the x and y directions

Power fluctuations are better than 3.4% rms and measurements of the M^2 factor with the aid of a Beamscope-P7 indicate an output beam with the transversal values $M^2_x = 1.3$ and $M^2_y = 2.0$ (see in Fig. 2).

2.2 Praseodynium laser cavity

The length and the nominal doping level of the Pr-doped crystals used in the experiments are reported in the Table 2. The table also gives the percentage of absorbed pump power at 469 nm for a single pass in the crystal. These absorbed powers could be larger for 4-levels laser systems; it means that the length and the doping concentration of the samples were not optimized.

Table 2. Parameters of the Pr^{3+}-doped fluorides in laser experiments

Material	Pr:LLF	Pr:YLF	Pr:KYF
Concentration	0.1 at%	0.8 at%	0.35 at%
Length (mm)	9	5	4.2
Absorbed power (Single pass)	33%	48%	34%

The schematic experimental laser setup is shown in Fig. 3. The laser cavity is a simple plano-concave cavity. The flat dichroic input mirror M_1 has a high transmission for the 469 nm pump wavelength and it is highly reflective either in red, the orange or in the green region around 640, 610 and 525 nm. The output curved mirror M_2 has a 50 mm radius of curvature and various transmissions depending on the considered laser wavelength. The pump laser is focused inside the laser crystals, without reshaping, with a lens having a focal length of 75 mm. The pump beam inside the crystals has a beam-waist radius of about 75 and 65 μm in the X and Y orientations respectively. By using the ABCD matrix formalism, a laser waist radius of 70 μm is found. The laser crystals having uncoated, flat, parallel and polished end-faces are mounted on a simple copper plate, without any additional cooling.

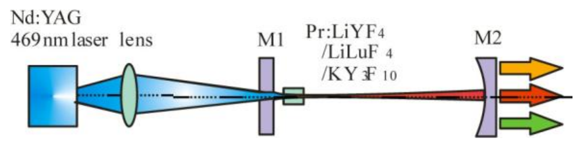

Fig. 3. Experimental setup for the Pr^{3+} cavity.

3. Laser results

3.1 Red emission

Figure 4(a) gives the maximum laser output power versus absorbed pump power for the $^3P_0 \rightarrow {}^3F_2$ red emission transition of the investigated samples. The emitting wavelength is around 640 nm for Pr:LLF and Pr:YLF, which corresponds to a σ polarization and around 645 nm for Pr:KYF (isotropic crystal). Several output couplers with different transmission values were experimented both to optimize the laser output power and to determine the optical losses in the cavity.

With a output mirror transmission of 4.2%, the power curves show maximum output powers up to 60, 137 and 65 mW for absorbed pump powers of about 290, 420 and 305 mW in the 9mm-long Pr (0.1at.%):LLF, 5mm-long Pr (0.8%):YLF and 4.2mm-long Pr (0.35%):KYF, crystals, respectively. The corresponding slope efficiencies are 41%, 45% and 40% (see in Fig. 4(a)).

It is worth noting that due to the relatively high transmission of the output coupler, it is found relatively high absorbed pump power thresholds, 148, 120 and 145 mW, respectively. With a 0.3% transmission output coupler, these threshold values reduce to 40, 25 and 33 mW. The laser beam at the maximum output power was found to be nearly diffraction-limited, with a M^2 factor equal to 1.1.

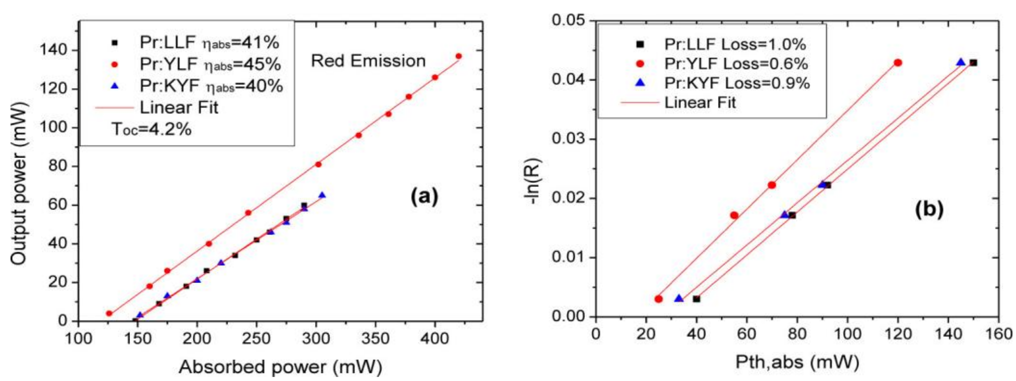

Fig. 4. (a) Optimized output power versus absorbed pump power curves for the $^3P_0 \rightarrow {}^3F_2$ red laser emissions of the investigated crystals. (b) Findlay-Clay analysis of the $^3P_0 \rightarrow {}^3F_2$ red laser emissions of the investigated crystals.

The optical losses in the cavity were estimated by using the Findlay-Clay method [20]. The total round-trip cavity losses, including absorption and scattering losses inside the laser medium and at the resonator mirrors as well as diffraction losses, are derived by measuring the absorbed pump power thresholds $P_{th,abs}$ for different output mirror transmissions T and by plotting -ln(R), with R = 1-T, versus $P_{th,abs}$. The curves are reported in the Fig. 4(b). From these curves, it is derived total round-trip losses of 1% for Pr:LLF, 0.6% for Pr:YLF and 0.9% for Pr:KYF.

3.2 Orange emission

Figure 5(a) gives the maximum output power versus the absorbed pump power for the $^3P_0 \rightarrow {}^3H_6$ orange laser transition of the three studied crystals. The emitting wavelength is around 607 nm for Pr:LLF and Pr:YLF, and around 610 nm for Pr:KYF. Using the output mirror transmissions of 2%, 2% and 1.8% for Pr:LLF, Pr:YLF and Pr:KYF, respectively, we measured maximum output powers of 37, 72 and 22 mW and associated slope efficiencies of 21%, 23% and 18%, respectively. To the best of our knowledge, it is the first demonstration of an orange laser emission in a Pr:KYF laser crystal. Lower transmission output couplers led to significantly lower output powers.

It is also worth noting that for the three considered Pr^{3+} doped crystals, there is a small but non-negligible spectral overlap between the $^3H_4 \rightarrow {}^1D_2$ absorption and the $^3P_0 \rightarrow {}^3H_6$ orange emission. These reabsorption effect likely accounts for the reduced efficiency of the orange laser emission compared to the red one.

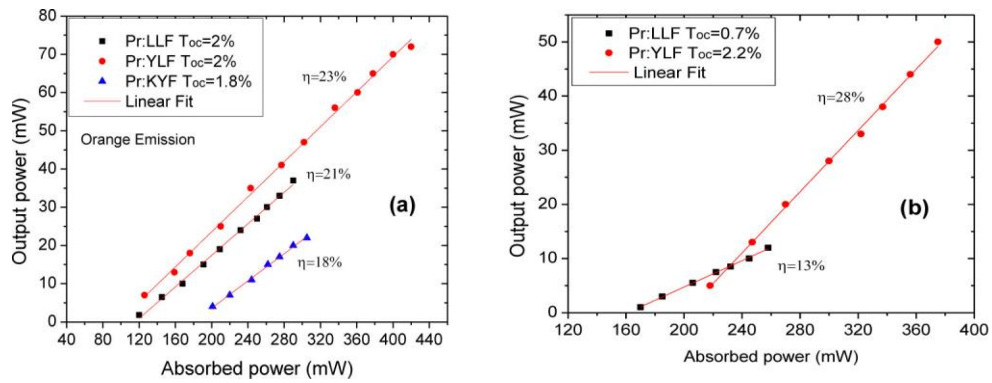

Fig. 5. (a) Optimized output power versus absorbed pump power curves for the $^3P_0 \rightarrow {}^3H_6$ orange emissions of the investigated crystals. (b) Optimized output power versus absorbed pump power curve for the $^3P_1 \rightarrow {}^3H_5$ green laser emissions of the investigated crystals.

3.3 Green emission

Figure 5(b) gives the maximum output power versus absorbed pump power for the $^3P_1 \rightarrow {}^3H_5$ green laser emission of the investigated samples. The emitting wavelength is around 522.5 nm for Pr:LLF and Pr:YLF. Here again better results were obtained with the Pr:YLF laser crystal. A maximum output power of 50 mW and a 28% slope efficiency were measured with a 2.2% transmission output mirror. For Pr:LLF, the best result, i.e. an output power of 12 mW and 13% slope efficiency, was obtained by using a 0.7% transmission output coupler.

A Findlay-Clay analysis was also performed in the case of Pr:YLF. However, due to the only 2 output mirrors used in the experiments, the Findlay-Clay plot was replaced by a calculation based on the following formula

$$\frac{P'_{th,abs}}{P_{th,abs}} = \frac{L - \ln(R')}{L - \ln(R)}$$

According to this calculation the round-trip cavity losses for the green laser emission in the case of Pr:YLF reaches a value of about 3%. Of course, the results obtained for this green emission in the case of Pr:LLF could be greatly improved considering the low Pr^{3+} concentration (0.1%) and the crystal length which was subsequently used, a length which is far too large to have a good overlap between the pump and the laser modes inside the crystal.

Finally, so far, we have not obtained any green laser emission from our Pr:KYF laser crystal. This is probably due to a significantly lower emission cross section and a threshold pump power exceeding the available pump power.

4. Summary

In conclusion, we have reported CW red, orange and green laser emissions of Pr:LLF, Pr:YLF and Pr:KYF crystals by pumping them with an especially developed diode-pumped and intracavity frequency-doubled CW Nd:YAG laser operating around 469 nm. The maximum red laser output powers were 60, 137 and 65 mW and the corresponding slope efficiencies were 41%, 45% and 40%. Orange laser emission was also obtained with the three crystals with maximum output powers of 37, 72 and 22 mW and corresponding slope efficiencies of 21%, 23% and 18%, respectively. Green laser operation was only achieved, however, with Pr:LLF and Pr:YLF, with maximum output powers of 12 and 50 mW, and slope efficiencies of 13 and 28%, respectively.

Further improvements both in terms of laser output powers and laser slope efficiencies can be definitely obtained and will be made by using shorter but concentration optimized laser crystals with AR-coated end-faces, a three-mirror resonator and better-adapted coatings for the input and output mirrors. In addition, better performance can be obtained by increasing the pump power and improving the pump beam quality. Thanks to the high power and high beam quality of the fiber-coupled IR laser diodes at 808 nm, together with the quality of the nonlinear crystals, SHG of Nd-based lasers (Nd:YAG as well as other Nd-doped materials which are currently studied for that application) should also rapidly improve and the proposed pumping scheme should open the way for efficient and compact high power Pr^{3+} solid-state lasers.

Acknowledgments

The authors wish to acknowledge the support from the French National Research Agency (ANR) within the framework of the FLUOLASE research program. They also wish to acknowledge Dr. A. Benayad and Mr. V. Ménard for their help in the preparation of the laser samples.