Facilitated by recent progress in blue InGaN laser diodes, Pr3+ lasers are the most efficient diode-pumped visible solid-state lasers. Simple frequency doubling enables efficient generation of deep ultraviolet (DUV) coherent radiation which is sought for many applications.

Various materials have been investigated as saturable absorbers (SAs) for Pr3+-based lasers. Among these, we previously demonstrated the Co2+ doped spinel crystal (Co2+ :MgAl2O4) to be an outstanding SA for passively Q-switched visible Pr3+:LiYF4 (Pr:YLF) lasers, yielding a simple approach for pulsed visible and UV sources demanded for many applications.15) Pulsed UV lasers are also needed in plasma spectroscopy. Previous work must maintain attribution to the author(s) and the title of the work, journal citation and DOI. Content from this work may be used under the terms of the Creative Commons Attribution 4.0 license. Any further distribution of this work must maintain attribution to the author(s) and the title of the work, journal citation and DOI. © 2022 The Author(s). Published on behalf of The Japan Society of Applied Physics by IOP Publishing Ltd

Figure 1 shows the laser setup. The resonator shorter than 1 cm reduced the resonator round trip time and thus pulse duration in all experiments. Compared to previous work,14) we minimized any air spacing between optical elements, maintaining only the necessary room for independent alignment of each component. We used a 5 mm long and 5 mm diameter a-cut cylindrical Pr3+:YLF crystal (Optogama UAB). The uncoated crystal was plane parallel, wrapped in indium foil and held in a water-cooled copper heatsink at 20 °C. The pump source was a linearly polarized cw 2ω-OPSL (Coherent, Inc.) with a maximum output power of 5 W at 479.05 nm and an M2 of 3. The incident pump power was adjusted by a combination of a λ/2-plate and a polarizing beam splitter. The π-polarized (E∥c) pump beam, which maximizes the absorption in Pr:YLF, was focused into the crystal by a spherical lens (f = 50 mm) to a beam waist diameter of 41 μm. The laser resonator was formed by a concave dichroic pump mirror (PM) with a radius of curvature of 10 mm and a plane output coupler (OC). The PM is highly transmissive for the pump at 479 nm and highly reflective for the laser wavelengths of 640, 607, and 523 nm. The lasing wavelength was selected by the respective OC-coating. We adopted OCs featuring transmissions of 2.3%, 3.5%, 5.7%, and 10.4% for 640 nm operation, 3.4%, 14.4%, and 23.5% for 607 nm operation, and 1.3%, and 1.9% for 523 nm operation.

For passive Q-switching we used four Co:MgAl2O4 crystals of different thickness and similar doping concentration. Their unbleached initial transmission values were calculated using the absorption coefficients determined in10) (see Table I). We selected Co:MgAl2O4 crystals with a 0.75 mm thick sample is antireflection-coated for the visible spectral range, yielding lower insertion losses.

An aluminum mount held the SAs between the Pr:YLF and the plane OC. The resonator shown in Fig. 1 yields a higher laser intensity in the SA than in the Pr:YLF, which is required for stable Q-switching.10,12) The single-pass pump absorption efficiency was obtained with OC transmissions of 2.3%, 2.5%, 5.7%, and 10.4% for 640 nm operation, 3.4%, 14.4%, and 23.5% for 607 nm operation, and 1.3% and 1.9% for 523 nm operation.12)

First, we characterized the laser in cw operation. The highest slope efficiency was obtained with OC transmissions (Toc) of 2.3, 14.4, and 1.3% at wavelengths of 640, 607, and 523 nm, respectively. The respective beam profiles are shown in Figs. 2(a)–2(c). Figure 2(d) shows the corresponding output characteristics. The highest slope efficiency of 62% was obtained at 640 nm with a maximum output power of 2.5 W at 4.2 W of absorbed power. This is comparable to the highest efficiencies of 68% obtained from a 2ω-OPSL-pumped Pr3+:YLF laser,4) proving an excellent mode matching and low losses despite the restrictions of our resonator.

At 607 nm the output power reached 1.5 W at a lower slope efficiency of 40% and high Toc of 14.4% owing to...
ground-state absorption $^3\text{H}_4 \rightarrow ^1\text{D}_2$ causing additional resonator losses by reabsorption of laser photons. This efficiency is below the highest efficiency of 60% reported for this transition,\textsuperscript{4)} which may be caused by a lower quality of our crystal, becoming in particular relevant at the high inversion densities induced by such high $T_{\text{OC}}$.\textsuperscript{4)}

The cw performance of the green laser at 523 nm was limited to 0.4 W of output power at 21% slope efficiency. Here, the negative thermal lens of YLF for $\pi$-polarized light\textsuperscript{20)} prevented fundamental-mode laser operation [see Fig. 2(c)]. A strong roll-over started at absorbed pump powers around 2 W and could not be compensated by alignment. The efficiency of the 523 nm laser strongly decreases at higher inversion levels arising from the increased resonator losses caused by the thermal lens.\textsuperscript{4)}

To analyze the observed trend of the peak power $P_{\text{peak}}$ and the pulse duration $t_{\text{p}}$ in the Q-switched laser operation, we adopted equations resulting from a rate equation model\textsuperscript{21,22)} describing the dependency of these parameters as well as pulse energy $E_p$ and repetition rate $f_{\text{rep}}$ on the modulation depth $\Delta T$ of the SA assuming a Gaussian pulse shape:

$$E_p = c_1 \cdot \Delta T$$
$$f_{\text{rep}} = c_2 \cdot \Delta T^{-1}$$
$$\tau_p = c_3 \cdot \Delta T^{-1}$$
$$P_{\text{peak}} = 0.94 \cdot \frac{P_{\text{avg}}}{f_{\text{rep}}} \propto c_4 \cdot \Delta T^2,$$

where $c_1, c_2, c_3, c_4$ are fitting parameters representing properties of the gain medium and resonator. Assuming negligible non-saturable losses $L_{\text{n}}$, one can rewrite Eqs. (1)–(4) as a function of the initial transmission $T_0$ with $\Delta T = 1 - T_0$. Note that this assumption is useful because the initial transmission of a SA can be directly measured. The fits to the experimental data for the Q-switched 640 nm laser versus the SA initial transmission $T_0$ at maximum pump power are shown in Fig. 3.

The repetition rate at fixed pump power decreases by an order of magnitude with increasing resonator losses (i.e. decreasing $T_0$) due to the higher laser threshold. This also allows more energy to be stored, increasing the pulse energy for smaller $T_0$. Simultaneously, the pulse duration shortens from $>10$ ns at $T_0 = 96\%$ to $<3$ ns at $T_0 = 80\%$, which in combination with the lower $f_{\text{rep}}$ significantly enhances the peak power.

The model predicts even shorter pulse durations at lower $T_0$, but practically this is infeasible for three reasons: first, the laser thresholds increase, which reduces the average power and causes inefficient or unstable laser operation. Second, the efficiency of Pr$^{3+}$-lasers decreases at high inversion densities\textsuperscript{4)} required to overcome the high insertion loss of SAs with lower $T_0$. Third, with the available Co$^{2+}$ doping concentration, longer SAs are required, counteracting our short resonator design.

The experimental data at $T_0 = 0.93$ in Fig. 3 deviate from the fitting curves for $f_{\text{rep}}$ and $E_p$ caused by the AR coating on this SA. Additionally, alignment limitations of the resonator

\begin{table}[h]
\centering
\caption{Initial transmission of the Co:MgAl$_2$O$_4$ SAs used in our experiments based on absorption coefficients from Ref. 10.}
\begin{tabular}{|c|c|c|c|c|}
\hline
Thicknes & Initial transmission $T_0$ & 640 nm & 607 nm & 523 nm \\
[mm] & & & & \\
\hline
0.40 & 0.962 & 0.954 & 0.989 \\
0.75a) & 0.930 & 0.916 & 0.979 \\
1.67 & 0.850 & 0.823 & 0.954 \\
2.30 & 0.800 & 0.764 & 0.938 \\
\hline
\end{tabular}
\begin{tablenotes}
\item a) Sample is antireflection-coated.
\end{tablenotes}
\end{table}

Fig. 1. (Color online) Schematic of the passively Q-switched Pr:YLF laser.

Fig. 2. (Color online) (a) Red, (b) orange, (c) green beam profile at the respective highest output power. (d) CW and (e) Q-switched laser characteristics of the different Pr:YLF laser at the respective best OC transmission. The corresponding pulse trains at highest output power are shown in Fig. 4.

Table I. Initial transmission of the Co:MgAl$_2$O$_4$ SAs used in our experiments based on absorption coefficients from Ref. 10.
caused minor differences of the beam diameter in the resonator for SAs of different thickness, which explains further deviations of the experimental results from the model.

As expected, we obtained the shortest pulses and highest peak power with the respective SA with the highest modulation depth that still allowed to reach the laser threshold. For the orange laser at 607 nm, where the SA-absorption is higher than at 640 nm (see Table I), this was the 1.67 mm thick SA, while the lower gain at 523 nm limited the maximum SA thickness to 0.75 mm.

Figure 2(e) shows the Q-switched laser characteristics for all three transitions, representative pulse trains for all lasers at maximum pump power in absolute power scale are shown in Figs. 4(a)–4(c).

At 640 nm an average output power of 285 mW was obtained at 11% slope efficiency for $T_{\text{oc}} = 10.4\%$. The pulse duration was as short as $2.4 \pm 0.1$ ns at a repetition rate of $97 \pm 12$ kHz, yielding a pulse energy of $2.9 \mu$J. Despite unexplained minor sub-pulses [see Fig. 4(a)], the pulse peak power of the main pulses reaches 1.1 kW. Figure 4(d) shows one of the shortest single pulses with a duration of 2.3 ns, which is the shortest pulse duration from any Q-switched Pr$^{3+}$ laser.

For the 607 nm laser we achieved an average power of 340 mW at 22% slope efficiency. Lower repetition rates of $48 \pm 18$ kHz and pulse durations of $5.3 \pm 1.5$ ns yielded a high average peak power of 1.3 kW at 7 $\mu$J pulse energy.

The low gain at 523 nm did not allow for operation at the high modulation depth required for shortest pulse durations. Consequently, the pulse durations of $102 \pm 14$ ns were significantly longer compared to those at 640 and 607 nm and the repetition rate reached 534 $\pm 65$ kHz. At such high repetition rates, the pulse interval of $\sim 1.9 \mu$s approaches the recovery time of the SA of 0.7 $\mu$s, causing the irregular pulse train in Fig. 4(c). The average power reached 145 mW at a slope efficiency of 6%; the corresponding average peak power remained as low as 2.5 W.

Table II summarizes the most relevant laser parameters of passively Q-switched Pr:YLF lasers. Compared to the first report utilizing a V-shaped longer resonator, we reduced the pulse duration and increased the peak power for the 640 and 607 nm lasers by orders of magnitude and for the 523 nm operation the pulse duration was decreased by a factor of two. Compared to our recent work with a similar compact resonator configuration, we further reduced the pulse duration by a factor of more than three and increased the peak power by more than a factor of five at 640 nm. Compared to the report of 20 W-direct diode pumping with a longer V-shaped resonator, our miniaturized resonator reached an order of magnitude shorter pulses, and thus comparable (640 nm) or even higher (607 nm) peak power levels.

Further improvement of the output parameters is straightforward by utilizing antireflection coatings on the SA- and Pr:YLF crystals. New and improved SAs may also result in shorter pulse durations and improved laser performance. Likewise, shorter resonators may contribute to further reduce the pulse duration. This can be achieved by utilizing thinner SA materials with stronger absorption, e.g., higher Co$^{2+}$-doped MgAl$_2$O$_4$ or a shorter Pr:YLF. To preserve the high pump absorption efficiency
Table II. Comparison of Pr:YLF lasers passively Q-switched with Co:MgAl2O4 SAs. The respective record values are marked in bold letters. All peak powers were calculated by Eq. (4).

| λ [nm] | 640 | 607 | 523 |
|--------|-----|-----|-----|
| P [W]  | 1.4 | 2.1 | 1.0 |
| f [MHz]| 0.17| 0.06| 0.78|
| Ppeak [kW]| 0.08| 1.1 | 0.16|
| τ [ns] | 103 | 30.9| 7.5 |

References
11 12 13
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11 12
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for this case requires a higher Pr3+ doping concentration, which results in a degradation of the laser efficiency.21) Maintaining a reasonable absorption efficiency in the gain material will therefore ultimately limit the resonator length, and the use of sub-mm gain materials as reported for Nd-doped gain materials seems impractical.21) Hence, we believe that the remaining possibilities to shorten the resonator will yield no further significant reduction of the pulse durations.

The higher thresholds when using a SA of low initial transmission, thus larger modulation depth, could be overcome by the use of high-power blue laser diode pump sources. However, the achieved pulse duration of 2.4 ns at TOC = 10.4% is only a factor of three above the limit given by the cavity lifetime, so that even at highest modulation depths sub-ns-pulses are hardly expected. Furthermore, the currently applied pump power is limited to few 10 W,23) and further pump power scaling requires to mitigate thermal effects and adopt improved cooling concepts.24) Finally, laser operation at reduced or even cryogenic temperatures may result in an improved performance, in particular for the orange laser.25)

In summary, we investigated the influence of the Co:MgAl2O4 SA’s modulation depth on the performance of the passively Q-switched Pr:YLF laser operated at 640 nm. Subsequently we also operated the laser at 607 nm and 523 nm. We obtained peak power levels exceeding 1 kW at pulse durations of 2.4 ns and 5.3 ns for the red and orange transition, respectively. The performance of the green laser was affected by the low gain at 523 nm and we did not obtain sub 100 ns pulse durations for this transition.

Overall, we presented the shortest pulse durations and highest peak powers reported for any passively Q-switched Pr:YLF laser, making our lasers useful sources to drive efficient nonlinear frequency conversion into the DUV spectral range.

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References
1) S. Nakamura, M. Senoh, S. I. Nagahama, N. Iwasa, T. Yamada, H. Kiyoku, and Y. Sugimoto, Jpn. J. Appl. Phys. 35, L74 (1996).
2) A. Richter, E. Heumann, E. Osiac, G. Huber, W. Seelert, and A. Diening, Opt. Lett. 29, 2638 (2004).
3) C. Kränkel, D. T. Marzahl, F. Moglia, G. Huber, and P. Metz, Las. Photonics Rev. 10, 548 (2016).
4) P. W. Metz, F. Reichert, F. Moglia, S. Mueller, D. T. Marzahl, C. Kränkel, and G. Huber, Opt. Lett. 39, 3193 (2014).
5) H. Tanaka, S. Kalusniak, M. Badtke, M. Demesh, N. Kuleshov, F. Kannari, and C. Kränkel, Prog. Quantum Electron. 84, 100411 (2022).
6) A. Richter, E. Heumann, G. Huber, V. O. Ostrooumov, and W. Seelert, Opt. Express 15, 5172 (2007).
7) T. Günt, P. Metz, and G. Huber, Appl. Phys. Lett. 99, 181103 (2011).
8) L. B. Zhou, T. S. Zhang, B. Xu, X. D. Xu, A. A. Lyapin, Z. F. Yu, and J. Xu, IEEE Photonics Technol. Lett. 33, 1151 (2021).
9) H. Tanaka, R. Kariyama, K. Iijima, K. Hirasawa, and F. Kannari, Opt. Express 23, 19382 (2015).
10) H. Tanaka, C. Kränkel, and F. Kannari, Opt. Mat. Express 10, 1827 (2020).
11) M. Demesh, D. T. Marzahl, A. Yasukevich, V. Kisel, G. Huber, N. Kuleshov, and C. Kränkel, Opt. Lett. 42, 4687 (2017).
12) S. Fujita, H. Tanaka, and F. Kannari, Opt. Express 27, 38134 (2019).
13) M. Badtke, H. Tanaka, L. J. Olffenburg, S. Kalusniak, and C. Kränkel, Appl. Phys. B. 127, 83 (2021).
14) M. R. H. Knowles, A. I. Bell, G. Rutherford, H. Booth, G. Foster-Turner, and A. J. Kearsley, Proc. SPIE 3888, 210 (2000).
15) C. Wagner and N. Harmed, Nat. Photonics 4, 24 (2010).
16) D. B. You, J. H. Park, B. S. Kang, D. H. Yun, and B. Shin, Sci. Adv. Mater. 12, 516 (2020).
17) C. A. Aguilar, Y. Lu, S. Mao, and S. C. Chen, Biomaterials 26, 7642 (2005).
18) S. E. Jackson, N. J. Pearson, W. L. Griffin, and E. A. Belousova, Chem. Geol. 211, 47 (2004).
19) O. S. Kazasidis and U. Wittrock, Opt. Express 22, 30683 (2014).
20) M. Pollnau, P. J. Hardman, M. A. Kern, W. A. Clarkson, and D. C. Hanna, Phys. Rev. B. 58, 16076 (1998).
21) G. J. Spühler, R. Paschotta, R. Fluck, B. Braun, M. Moser, G. Zhang, E. Gini, and U. Keller, J. Opt. Soc. Am. B. 16, 376 (1999).
22) J. J. Degnan, IEEE J. Quantum Electron. 31, 1890 (1995).
23) H. Tanaka, S. Fujita, and F. Kannari, Appl. Opt. 57, 5923 (2018).
24) W. A. Clarkson, J. Phys. D: Appl. Phys. 34, 2381 (2001).
25) N. O. Hansen, PhD-Thesis Universität Hamburg (2011).