Rapid Communication

Pulsed Laser Action of Pr:GdLiF₄ at Room Temperature

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Abstract. We realized, to our knowledge for the first time, laser emission in Pr:GdLiF₄ at seven wavelengths: 522 nm, 545 nm, 604.5 nm, 607 nm, 639 nm, 697 nm, and 720 nm. The crystal was pumped with an excimer laser pumped dye laser at 468 nm. All laser experiments were carried out at room temperature. We also achieved pulsed room temperature laser oscillation of Pr:KYF₄ at 642.5 nm pumped at 465 nm wavelength.

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Pr³⁺ is an interesting ion for many visible laser transitions because of the large number of energy levels (Fig. 1, data for Pr:YLF after Adam et al. [1]). After the first report of a Pr laser [2] many Pr-doped crystals have been investigated. An overview has been given by Kaminskii [3]. Recently CW laser action was obtained in Pr-doped LaCl₃ [4], fibres [5–8], and YAlO₃ [9, 10]. Due to the large number of laser channels in Pr:LiYF₄ (Pr:YLF) [11–13] and Pr:LiLuF₄ [14] in the pulsed mode, we tried to obtain laser action in other Pr-doped fluoride crystals, GdLiF₄ (GLF) and KYF₄ (KYF).

As the Y site in YLF seems to be too small for the Pr³⁺ ion, these crystals easily crack when stressed mechanically, thermally or by depositing high pump energies. Therefore, we looked for a new host material with a larger site for the Pr³⁺ ion expecting a higher damage threshold. For this purpose we have chosen GLF which has the same tetragonal scheelite (CaWO₄) structure as YLF [15] and which is an interesting host material for Nd³⁺ lasers [16]. The investigated crystal had a Pr concentration of 0.3%.

The laser experiments were performed using a nearly concentric resonator with mirrors of 5 cm radius of curvature. The crystal of 9.3 mm length was pumped with an excimer laser pumped dye laser at 468 nm where it absorbed 75%. The roughly elliptical pump beam was focussed onto the crystal with a lense of 5 cm focal length to a spot size of approximately 50×100 µm². The pump pulse duration was about 50 ns. We observed laser action at seven wavelengths. The results obtained so far are shown in Fig. 1 and summarized in Table 1. The excitation density nₜ₉ at threshold was estimated by the following equation:

\[ n_{\text{thr}} = \frac{E_{\text{thr}}}{E_p V}. \]

Eₜ₉ is the absorbed energy at threshold, Eₚ = hc/λₚ is the energy of the pump photons. The pumped volume V was roughly estimated from the pump focus and the crystal length.

It was even possible to achieve simultaneous oscillation of up to three transitions, e.g., at 522 nm, 545 nm, and 639 nm. The laser wavelengths indicate that the en-

Fig.1. Energy levels and laser transitions of Pr:GLF
Table 1. Results of the laser experiments on Pr:GLF. The values for the threshold $E_{\text{thr}}$ and $N_{\text{thr}}$, the maximum output energy $E_{\text{out}}$, and the slope efficiency $\eta$ are not optimized. They strongly depend on the crystal quality and the transmittance of the output mirror.

<table>
<thead>
<tr>
<th>Transition</th>
<th>$\lambda$ [Ã]</th>
<th>Polarization</th>
<th>$E_{\text{thr}}$ [mJ]</th>
<th>$N_{\text{thr}}$ [cm$^{-3}$]</th>
<th>$E_{\text{out}}$ [mJ]</th>
<th>$\eta$ [%]</th>
</tr>
</thead>
<tbody>
<tr>
<td>$^{3}P_{1} \rightarrow ^{2}H_{5}$ 522</td>
<td>E</td>
<td></td>
<td>c</td>
<td>197</td>
<td>$1.3 \times 10^{19}$</td>
<td>83</td>
</tr>
<tr>
<td>$^{3}P_{0} \rightarrow ^{2}H_{6}$ 545</td>
<td>E</td>
<td></td>
<td>c</td>
<td>49</td>
<td>$3.2 \times 10^{18}$</td>
<td>2</td>
</tr>
<tr>
<td>$^{3}P_{0} \rightarrow ^{2}H_{9}$ 604.5</td>
<td>E</td>
<td></td>
<td>c</td>
<td>144</td>
<td>$9.3 \times 10^{18}$</td>
<td>32</td>
</tr>
<tr>
<td>$^{3}P_{0} \rightarrow ^{2}F_{4}$ 639</td>
<td>E</td>
<td></td>
<td>c</td>
<td>4</td>
<td>$2.6 \times 10^{17}$</td>
<td>98</td>
</tr>
<tr>
<td>$^{3}P_{0} \rightarrow ^{2}F_{4}$ 679</td>
<td>E</td>
<td></td>
<td>c</td>
<td>73</td>
<td>$4.7 \times 10^{18}$</td>
<td>31</td>
</tr>
<tr>
<td>$^{3}P_{0} \rightarrow ^{2}F_{4}$ 720</td>
<td>E</td>
<td></td>
<td>c</td>
<td>6</td>
<td>$3.9 \times 10^{17}$</td>
<td>80</td>
</tr>
</tbody>
</table>

Energy levels of Pr:GLF are comparable to those of Pr:YLF [1]. This is also confirmed by the absorption spectra which are very similar for both crystals. The lifetime of the thermally coupled $^{3}P_{0}$ and $^{3}P_{1}$ levels is 40 Ìs for Pr(0.3%):GLF and 36 Ìs for Pr(0.8%):YLF.

The estimated damage threshold of Pr:GLF was approximately 400 MW/cm$^2$ which is twice as high as for Pr:YLF. Furthermore, neither the crystal length nor the transmission of the output coupler was optimized.

Input-output diagrams of the laser transitions at 522 and 639 nm are shown in the Figs.2 and 3. At 639 nm we obtained laser action even without an output mirror. In this case the feedback was only given by the crystal surface. The threshold of about 170 ÌJ yields an estimated emission cross-section of the order of 10$^{-19}$ cm$^2$.

A more detailed investigation of the spectroscopic properties of Pr:GLF is in progress.

The pulse duration of the Pr laser at 639 nm, pumped with a pulse of 50 ns at 468 nm, was of the same order as the pump-pulse duration (Fig.4). The delay time between pump and laser pulse depends on the pump energy and the transmittance of the output coupler. In our experiments it was of the order of a few hundred nanoseconds.

The relative heights of pump and laser pulse in Fig.4 are arbitrary. Without an output mirror, i.e., only using the reflection of the crystal surface ($T = 96\%$), the laser-pulse duration was only 6 ns with a delay of 60 ns. For the laser transition at 522 nm we obtained a pulse duration of about 140 ns with a delay of the order of a hundred nanoseconds.

We also investigated Pr:KYF with a doping level of 0.4%. The lifetime of the $^{3}P_{0}$ level is about 50 Ìs. Pumping at 465 nm we obtained laser action at 642.5...