UV-INDUCED LOSS MECHANISMS IN A Ce$^{3+}$:YLiF$_4$ LASER

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The optical pumping of Ce$^{3+}$:YLiF$_4$ at 308 nm leads to the formation of transient and stable color centers due to an excited-state absorption which photoionizes the cerium ions. Measurements of the temperature dependence of the transient center lifetime and the dependence of the single-pass gain on pump repetition rate are presented.

1. Introduction

The optical absorption and luminescence spectra of Ce$^{3+}$:YLiF$_4$ (YLF) are due to the parity allowed 4f$^1$-5d transitions of the trivalent cerium ion. The broad vibronic emission from the lowest 5d state should make Ce$^{3+}$:YLF an ideal tunable solid state laser material which would operate in the near UV from 310 to 350 nm. Although laser action has been demonstrated in this material$^1$, the results indicate less than ideal performance characteristics. The limitations include an early onset of gain saturation and rolloff as well as a pump repetition rate maxima of 0.5 Hz. We have recently demonstrated$^2$ that a two-step photoionization of the Ce$^{3+}$ ions due to an excited-state absorption to the conduction band is responsible for the formation of transient and permanent color centers in Ce$^{3+}$:YLF. The color centers, which are absorptive at the Ce$^{3+}$ emission band are the origin of the performance limitation for an optically pumped Ce$^{3+}$:YLF laser.

2. EXPERIMENTAL RESULTS

The experimental technique used to measure the transient gain and loss utilizes a coaxial pump-probe geometry. The output of a pulsed 308 nm XeCl laser was focused to provide a 2 mm diameter spot along the 13 mm length of a 1% doped sample. The probe beams were from a He-Cd laser (325 and 442 nm) and a He-Ne laser (633 nm) and were focused to a 0.3 mm spot concentric with the pump beam. The transmitted probe intensity was monitored by a photomultiplier tube and care was taken to filter out the Ce$^{3+}$ fluorescence and the scattered pump light. A LeCroy 3500 transient digitizer and signal averager was used to record the time dependence of the photomultiplier signal.

Although the time dependence of the transient absorption following 308 nm pumping is highly nonexponential, the decay can be characterized by distinct and well separated multiple lifetime components. The values of these lifetime components at room temperature are 50 ns, 200 $\mu$s, 50 ms and 30 s. The longer time components grow in strength, at the expense of the shorter components, as the probe wavelength moves to the blue. We have been able to follow the temperature dependence of these lifetime components as displayed in Fig. 1. The lifetimes fall into four distinct groups, each of which can be fit to an Arrhenius function of the form $\tau^{-1} = A \exp(-\Delta E/kT)$. Thus the decay of the configurations responsible for the transient...
absorptions are thermally activated. The values of the rate parameter $s$ and the activation energy $\Delta E$ are displayed in Table 1.

<table>
<thead>
<tr>
<th>group</th>
<th>$s$ (sec$^{-1}$)</th>
<th>$\Delta E$ (cm$^{-1}$)</th>
</tr>
</thead>
<tbody>
<tr>
<td>A</td>
<td>$2.8 \times 10^{10}$</td>
<td>1450</td>
</tr>
<tr>
<td>B</td>
<td>$1.2 \times 10^{11}$</td>
<td>4200</td>
</tr>
<tr>
<td>C</td>
<td>$1.2 \times 10^{10}$</td>
<td>4200</td>
</tr>
<tr>
<td>D</td>
<td>$1.2 \times 10^{6}$</td>
<td>4500</td>
</tr>
</tbody>
</table>

The transient color centers as well as the more permanent centers are detrimental to the lasing characteristics of Ce$^{3+}$:YLF in that they are absorptive at the Ce$^{3+}$ emission wavelengths. The role of the transient centers is evident from our measurements of the transient single-pass gain as a function of the 308 nm pump repetition rate, as displayed in Fig. 2. For these measurements, the transmitted 325 nm probe is sampled immediately after the 8 ns duration pulse from the XeCl laser. The 325 nm wavelength is near the peak of the gain profile for the Ce$^{3+}$ stimulated emission. For pump repetition rates greater than 1 Hz, the sample is absorptive at 325 nm, with the losses due to the transient centers exceeding the stimulated optical gain. If the pump repetition rate is much less than 1 Hz, then there is sufficient time for the transient color center population to relax, and the gain exceeds the loss. The 1 Hz repetition rate is nearly the 0.5 Hz value mentioned by Ehrlich et al.\textsuperscript{1} for which there is a significant drop in the Ce$^{3+}$:YLF laser output.

FIGURE 1. Logarithm of the transient center lifetime as a function of $1000/T(K)$.

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REFERENCES