Crystal growth, structure, spectroscopy, and lasing of NaLu$_{1-x}$Yb$_x$(WO$_4$)$_2$

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Tetragonal double tungstate NaT$_{1-x}$Yb$_x$(WO$_4$)$_2$ crystals emerged in the past year as efficient hosts for tunable Yb$^{3+}$ lasers. A tuning range larger than 60 nm was achieved for T=Gd, promising for sub-50 fs mode-locked pulses. The previously studied hosts, T=La and Gd, have congruent melting character and were grown by the Czochralski method. Since NaYb(WO$_4$)$_2$ melts incongruently there should be some upper $x_{\text{MELT}}$ limit for congruent melting of NaT$_{1-x}$Yb$_x$(WO$_4$)$_2$ compounds. For the above two hosts it is $x_{\text{MELT}}\approx$20 mol %. Higher Yb doping or alternative hosts with incongruent melting require different growth methods. In this work we used Na$_2$W$_2$O$_7$ flux and the top seeded solution growth method to prepare NaLu$_{1-x}$Yb$_x$(WO$_4$)$_2$ single crystals with $x_{\text{MELT}}=0$, 0.1 and 0.5.

From single crystal x-ray diffraction we conclude that these crystals have the $I\bar{4}$ space group symmetry. The refined formula for $x_{\text{MELT}}=0.5$ is Na$_{0.99(2)}$Lu$_{0.54(4)}$Yb$_{0.47(4)}$(WO$_4$)$_2$. Yb$^{3+}$ occupies two non equivalent crystallographic sites (also shared by Na and Lu) with occupancy factors 0.16 (2$d$-site) and 0.31 (2$b$-site). This multiple occupancy of the same crystallographic sites by Na, Lu and Yb cations contributes to the Yb$^{3+}$ linewidths.

At 300 K, the maximum absorption cross section for the $\pi$-polarization, $\sigma_{\text{GSA}}=2.1\pm0.1\times10^{-20}$ cm$^2$ at $\lambda=974.1$ nm, is slightly higher than for the $\sigma$-polarization, $\sigma_{\text{GSA}}=1.9\pm0.1\times10^{-20}$ cm$^2$. The 300 K photoluminescence spectral profile is similar for both polarizations at longer wavelengths up to $\approx$1100 nm. Laser operation was achieved at 300 K in a Z-shaped optical cavity using an uncoated sample of NaLu$_{1-x}$Yb$_x$(WO$_4$)$_2$ with $x_{\text{MELT}}=0.1$ under Brewster angle and without special cooling. Preliminary results with Ti:sapphire laser pumping are shown in Figure 1. The 0.94 mm thick sample absorbed roughly 60% of the pump radiation for the three output couplers used. A slope efficiency of $\eta=51.6\%$ was obtained for an output coupler transmission of $T_{OC}=5\%$ and $\sigma$-polarization. The maximum output power achieved with $T_{OC}=1\%$ was 282 mW for an absorbed power of $P_{\text{abs}}\approx 1.1$ W, however, at $P_{\text{abs}}\approx 900$ mW, roll-off of the output power dependence occurred. This was attributed to thermal effects because using a chopper the same slope efficiencies as those shown in the figure were obtained for peak powers up to $P_{\text{abs}}\approx 1.1$ W.

![Figure 1. Continuous-wave laser performance of NaLu$_{0.9}$Yb$_{0.1}$(WO$_4$)$_2$ at room temperature. $\lambda_{\text{PUMP}}=974.9$ nm.](image-url)