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Two step pumped YLF:Tm blue upconversion laser

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Abstract
Several fluoride and oxide Tm and (Tm, Yb) doped crystals such as YLF, GLF, YAG, and YAlO₃ were tested for pulsed lasing at the ¹D₂ → ³F₄ transition (453 nm blue) at room temperature under two step pumping by a Ti:sapphire and a dye laser. Laser operation was obtained for the fluoride crystals. The input-output curves exhibit a saturation effect of emission with respect to both pump lasers. This can be explained by a depletion of the ³H₆ ground state of Tm³⁺ and the competition between the excited state absorption from the ³H₄ intermediate level and the mechanisms depopulating this level.

Introduction
Blue laser radiation is of practical importance for many purposes, such as information processing or medical applications. Up-conversion or excited state absorption (ESA) pumping schemes are of interest in this respect because in principle powerful laser diodes in the red or infrared spectral region could be used as pump sources.

Trivalent thulium presents interesting properties due to the existence of two strong blue emission transitions, ¹D₂ → ³F₄ and ¹G₄ → ³H₆. Recently blue laser emission of this ion was reported at cryogenic temperatures [1, 2], but there are only very few data on the emission characteristics at 300 K [2]. We investigated the possibilities to obtain blue laser emission at the transition ¹D₂ → ³F₄ from several fluoride- (YLiF₄, GdLiF₄) or oxide- (YAlO₃, Y₃Al₅O₁₂) crystals doped with Tm or double doped with Tm and Yb, by two step pumping using a pulsed Ti:sapphire laser (tuned to 780 nm, pulse duration 60 µs) and a dye laser (650 nm, 40 ns). Under this excitation laser operation could be obtained in the fluoride crystals while the oxide crystals were destroyed before the laser threshold was reached.

Fig. 1. The Tm³⁺ energy levels and the transitions for pumping and laser-emission at 453 nm

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Results
The excitation scheme is shown in Figure 1. The Ti:sapphire laser populates the level $^3H_4$ and the dye laser subsequently raises the excitation to $^1D_2$. The wavelength of the dye laser was chosen to match the strongest ESA transition $^3H_4 \rightarrow ^1D_2$. Since the level $^3H_4$, which serves as the intermediate state, could be efficiently depopulated by the cross-relaxation process $(^3H_4, ^3H_6) \rightarrow (^3F_4, ^3F_4)$, the proper choice of setting the delay time of the dye laser pulse with respect to the Ti:sapphire pulse would be crucial for the pumping process. Our measurements of luminescence emission $^1D_2 \rightarrow ^3F_4$ as a function of the delay time between the dye laser pulse and the beginning of the Ti:sapphire pulse show, that in case of Tm(1%):YLF the maximum emission intensity is obtained when the delay time is about 70 $\mu$s (Figure 2).

Using the experimental set-up presented in Figure 4 we obtained room temperature blue emission at 453 nm on the transition $^1D_2 \rightarrow ^3F_4$ in Tm(1%):YLF, Tm(1%), Yb(10%):YLF and Tm(1%):GLF. The resonator was nearly concentric and formed by 5 cm radius of curvature mirrors with a transmission of 0.5 % at the blue laser wavelength. The input coupler had high transmission and the output coupler had high reflection for both wavelengths of pumping in order to enhance the absorbed pump power. The best results were obtained for the Tm,Yb:YLF-crystal (Figure 3), about twice the output of the YLF-crystal without Yb. The positive influence of Yb on this emission is not yet clearly understood.

![Fig. 2. $^1D_2 \rightarrow ^3F_4$ emission in Tm:YLF as a function of the delay time between the dye laser pulse and the on-set of the Ti:sapphire laser](image)

![Fig. 3. Emission energy of the up-conversion pumped blue laser in Tm(1%), Yb(10%):YLF as a function of absorbed Ti:sapphire radiation for three values of absorbed dye laser energy](image)
From the measured input-output curves one can see a saturation effect with respect to both pump lasers. Figure 3 shows clearly this effect as a function of absorbed Ti:sapphire radiation. In order to explain the observed behaviour, we performed a computer modelling of the laser emission using rate equations with parameters determined from luminescence experiments. The resulting temporal evolution of populations of the Tm levels involved in this process is shown in Figure 5. It can be clearly seen, that the saturation of emission with respect to the Ti:sapphire laser originates essentially from the severe depletion of the Tm ground state $^3H_6$: for 50 mJ of pump energy the population of $^3H_6$ drops to about 20 % of its initial value. The modelling also shows that in case of the dye pump the saturation process is governed by the balance between the dye laser excited state absorption process from $^3H_4$ and the rates of the other depopulation mechanisms for this level.

In summary, we have investigated two step laser pumping of Tm doped $\text{YLiF}_4$, $\text{GdLiF}_4$, $\text{YAlO}_3$, and $\text{Y}_3\text{Al}_5\text{O}_{12}$. Pulsed laser operation at 453 nm could be obtained in case of the fluorides at 300 K.
Fig. 5. The temporal evolution of populations predicted by the rate equation modelling.

References
