Visible laser emission of Pr^{3+} in various hosts

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Abstract.

We report on the first observation of low temperature pulsed blue and orange laser emissions in $\text{Pr}^{3+}:\text{YAG}$. Furthermore, we compare these results with similar observations in YLiF_4 and YAlO_3 doped crystals. Pulsed laser oscillations were obtained in crystals with uncoated surfaces by direct excitation of one of the relevant Stark components of the $^3\text{P}_0$, $^3\text{P}_1$ or $^3\text{P}_2$ states. A careful spectroscopic analysis of all these systems allows to clearly identify the laser transitions as arising from the $^3\text{P}_0$ metastable state, the lifetime of which has been measured using a nanosecond pumped dye laser.

Introduction.

The renewal of interest in studying praseodymium (Pr^{3+})-doped solid state materials is due to the emission spectrum of this ion extending from ultraviolet [1] to near infrared [2]. Laser action in several Pr^{3+} -doped crystals: YLF [3], YAP [4], YAG [5] as well as Pr^{3+} -doped fibres has recently been reported. The need for compact, integrated laser sources in the blue-green part of the spectrum has led to development of up-conversion laser using two-photon pumping schemes, excited state absorption or energy transfer, to generate laser emission at wavelengths shorter than those used for pumping [6]. Clearly, of practical significance are one-colour IR laser diode pumped devices of this type. Our recent studies put into evidence efficient one-colour up-conversion of IR photons to blue emission in Pr^{3+} -doped YAG and YLF crystals [7]. In this paper, we compare the first observation of low temperature pulsed blue and orange laser emission in $\text{Pr}^{3+}:\text{YAG}$ reported recently [8] with similar observations in YLF and YAP doped crystals.

Experimental.

The samples used are $\text{YAG}: 0.6\text{at}\%\text{Pr}^{3+}$, $\text{YAP}: 0.1\text{at}\%\text{Pr}^{3+}$ and $\text{YLF}: 0.24\text{at}\%\text{Pr}^{3+}$. Their thickness is 2.53 mm, 4.97 mm and 4 mm respectively. They are optically polished without

any coating on the surfaces. They are mounted in a liquid He cryostat with a gas heating system to allow temperature variation from 4.4 K to 300 K. The crystals were excited longitudinally by a pulsed Coumarin 480 dye laser (10 ns pulse, 0.1 cm^{-1} band width) pumped by a tripled Nd: YAG or an excimer laser.

Results.

Each crystal was excited selectively in one of the Stark components of the 3P_2 , 3P_1 or 3P_0 states. Then fluorescence occurs from the 3P_0 metastable level which decay is mostly radiative. The gain was high enough that only with optically polished plane parallel ends, providing $\approx 4\%$ Fresnel reflections from the crystal/He interface, laser emission was achieved at cryogenic temperatures. The table 1 summarizes the 3P_0 spectroscopic properties (position and lifetime), the excitation and laser emission wavelengths for each

	3P_0 lifetime (μs)	3P_0 energy (cm^{-1})	Excitation wavelength (nm)	Blue laser wavelength (nm)	Orange laser wavelength (nm)
YAG : Pr $^{3+}$ 0.6at%	15	20538	475 $^3H_4(1) \rightarrow ^3P_1(2)$ $\Gamma_3 \rightarrow \Gamma_2$	487.9 $^3P_0(1) \rightarrow ^3H_4(3)$ $\Gamma_1 \rightarrow \Gamma_4$	616 $^3P_0(1) \rightarrow ^3H_6(1)$ $\Gamma_1 \rightarrow \Gamma_4$
YAP : Pr $^{3+}$ 0.1at%	11	20418	473 $^3H_4(1) \rightarrow ^3P_2(1)$ $\Gamma_1 \rightarrow \Gamma_2$	491 $^3P_0(1) \rightarrow ^3H_4(2)$ $\Gamma_1 \rightarrow \Gamma_2$	613.9 $^3P_0(1) \rightarrow ^3H_6(1)$ $\Gamma_1 \rightarrow \Gamma_1$
YLF : Pr $^{3+}$ 0.24at%	50	20866	479.2 $^3H_4(1) \rightarrow ^3P_0(1)$ $\Gamma_2 \rightarrow \Gamma_1$	-	604.3 $^3P_0(1) \rightarrow ^3H_6(1)$ $\Gamma_1 \rightarrow \Gamma_2$

Table 1: 3P_0 spectroscopic properties and excitation and laser emission wavelengths.

crystal as well as assignment for each transition. The blue lasers obtained in YAG: Pr and YAP: Pr operated at temperatures up to 32 K. In the fluoride, only the orange laser was observed. For the three crystals, this orange laser emission, which was extremely intense at 4.4 K, started to decrease around liquid nitrogen temperature and ceased at about 140 K. The figure 1 shows the output energy of these orange lasers measured at one end of the crystal only as a function of pump energy at 4.4 K. Temporal analysis revealed instantaneous build-up of the laser oscillations after the excitation pulse. In the time scale of our apparatus, the temporal behaviour of the laser emission follows the excitation pulse profile. In the cases of YAG: Pr and YAP: Pr, which are pumped by selective excitation in

the 3P_1 and 3P_2 manifold respectively, this is in agreement with fast relaxation between these levels and the 3P_0 state.

Fig. 1: Output energy of the orange laser emission as a function of pump energy at 4.4 K

Discussion.

A careful spectroscopic analysis of all these systems allowed us to build the energy level scheme of Pr^{3+} ion in these crystals, and to clearly identify the laser transitions as it is reported in the table 1.

In YAG:Pr^{3+} , the blue laser transition terminates on the third 43 cm^{-1} Stark component of the ground state manifold. In YAP:Pr^{3+} , the blue laser transition terminates on the second 52 cm^{-1} Stark component of the ground state manifold. These levels start to be substantially populated at higher temperatures explaining the increase of threshold and disappearance of laser emission at 32 K.

In YAG:Pr^{3+} , YAP:Pr^{3+} and YLF:Pr^{3+} , the transitions corresponding to the orange laser terminate at the lowest Stark component of the $^3\text{H}_6$ multiplet fulfilling the four-level operating scheme. In this case the thermal line broadening resulting in the reduction of the peak absorption and emission cross sections is responsible for the increase of threshold at higher temperatures.

Conclusion.

It is evident that after optimizing the crystal length and using appropriate mirrors it should be possible to improve laser performances of the investigated systems. The influence of doping level on laser efficiency should also be considered. Due to the quasi-four-level scheme for the blue laser, its low threshold operation could be limited to low temperatures. However, this work demonstrates the potential of these Pr^{3+} -doped systems for a blue laser. Recently, we observed such laser oscillations from the $^3\text{P}_0$ level in other crystals as $\text{BaY}_2\text{F}_8:\text{Pr}^{3+}$, GGG:Pr^{3+} , Further studies of up-conversion pumping in several Pr^{3+} systems are in progress.

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