Orange avalanche upconversion for high-resolution laser spectroscopy

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Abstract. High resolution spectroscopy of quantum systems is a key point in the quantum information field. Rare earth ions in crystals are interesting candidates for this application because of the long coherence lifetime of some of their transitions. These ions can also be manipulated by optical excitation which has, however, to be produced by ultra-stable laser sources. This proves to be a very difficult task in the case of dye lasers which are necessary to excite Pr$^{3+}$ doped crystals and especially Pr$^{3+}$:Y$_2$SiO$_5$. This compound is by far the most used host in rare earth based quantum information studies. In this paper, we discuss the use of Pr$^{3+}$,Yb$^{3+}$ codoped materials to build an infrared pumped solid state laser suitable to excite Pr$^{3+}$:Y$_2$SiO$_5$ around 606 nm. We show by the analysis of a rate equation model that avalanche upconversion is not very efficient to obtain a high power laser. This is more easily obtained if a second laser is set to pump resonantly Yb$^{3+}$ ions. The spectroscopic properties of a new matrix Pr$^{3+}$:Y$_2$SiO$_5$:PbF$_2$ are also investigated. We found that this compound emits at 606.18 nm with a width of 5 nm and would be therefore suitable to excite Pr$^{3+}$:Y$_2$SiO$_5$. Moreover, it can be excited at 857 nm and 975 nm, i.e. in the range of high power laser diodes.

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1 Introduction

High-resolution laser spectroscopy, including hole-burning and photon echoes techniques, is very useful to determine the homogeneous linewidths of optical transitions and energy level structures hidden inside the inhomogeneous linewidths. These data are especially useful in the field of quantum information based on spin impurities in solids [1,2]. Recently, rare earth doped crystals have been actively studied as candidates for implementing solid state quantum computers or quantum memories. In the first case, a two-qubit control phase gate has been demonstrated as well as the selection of qubit within the inhomogeneous line-width [3,4]. On the other hand, a pulse of light as been stored for more than one second using a homogeneous linewidth [3,4]. On the other hand, a pulse of light as been stored for more than one second using a homogeneous linewidth [3,4].

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As an alternative approach, we propose to use a Pr$^{3+}$ laser emitting around 606 nm on the $^3P_0 \rightarrow ^3H_6$ transition. As a matter of fact, an orange laser has been demonstrated using a fluoride crystal, BaY$_2$F$_8$, codoped with praseodymium and ytterbium ions, with output powers of about 50 mW [11]. The pump was a Ti-Sa laser delivering up to 3.42 W (corresponding to 500 kW/cm$^2$) at
A process called avalanche upconversion [12,13] is used in this material to populate high energy levels from the infrared excitation. Previous works have shown that avalanche upconversion is very efficient in Pr$^{3+}$, Yb$^{3+}$ doped materials [14–16]. A laser based on this process, properly stabilized and diode pumped, could replace dye lasers in quantum information applications based on Pr$^{3+}$ doped crystals. However, Pr$_3$Yb$_2$F$_8$ emission peaks at 607.5 nm with a 1 nm linewidth (FWHM) and is therefore non resonant with Pr$^{3+}$:Y$_2$SiO$_5$ absorption. Moreover, the pumping scheme used in this work results in very high excitation densities which are close to the crystal and mirrors damage thresholds. This is particularly true of fluoride hosts which are usually fragile. Such high pumping intensities are also difficult to obtain with laser diodes because of focusing problems.

In this paper, we address these two points. First of all, the avalanche process in Pr$^{3+}$, Yb$^{3+}$ crystals is discussed and a double wavelength pumping scheme is proposed to achieve higher excited state populations and stimulated emissions with the same total pumping intensity. Then, basic spectroscopic properties of a new crystal, Pr$_3$Yb$_2$:PbF$_2$ are described. This material emits a strong orange emission in resonance with Pr$^{3+}$:Y$_2$SiO$_5$ absorption at 605.977 nm.

2 Experimental

Absorption spectra were recorded on a Varian Cary 5 spectrophotometer. Emission spectra were obtained using a Jobin-Yvon HRD 600 mm double monochromator and a Hamamatsu H6240-01 photon counting head. Wavelength and relative intensity calibrations were performed using a Ne spectral lamp and a reference quartz-halogen lamp. Excitation was provided by a CW Coherent 890 Ti-Sa laser pumped by a Coherent Sabre argon laser. Excitation spectra were corrected for Ti-Sa output power variations as a function of wavelength.

β-PbF$_2$ single crystals doped with YbF$_3$ and PrF$_3$ were synthesized using a modified Bridgman method [17]. PbF$_2$ and LnF$_3$ powders were put into a vitreous carbon crucible and placed into the strong thermal gradient (20 °C/cm) of a vertical furnace, heated at 1000 °C. The slow decrease of the temperature of the furnace (from 1000 °C to 820 °C) induces the displacement of the thermal gradient and the slow crystallisation of the PbF$_2$, LnF$_3$ mixture.

3 Optimal pumping scheme for Pr,Yb upconversion lasers

3.1 Rate equation modeling

Avalanche upconversion mechanisms in Yb$^{3+}$, Pr$^{3+}$ systems have been studied in several papers [15,18,19]. In this work, we focus on the effect of a non negligible ground state absorption, following our previous work on three-level systems [20].

Pr$^{3+}$ and Yb$^{3+}$ energy levels involved in the orange avalanche upconversion process are shown in Figure 1 as well as the excitation paths and energy transfers. The main characteristic of avalanche upconversion is an excitation beam (around 850 nm) which is strongly resonant with a transition from an excited state, in this case $^1G_4 \rightarrow ^3P_0$. On the other hand, the excitation beam is only very weakly resonant with transitions starting from the ground states and therefore ground state absorption (GSA) is very small. In the Pr,Yb system, non resonant GSA occurs through ytterbium $^2F_{7/2} \rightarrow ^2F_{5/2}$ transition. Such a scheme would normally lead to a negligible population in the highest level but strong energy transfers between Yb$^{3+}$ and Pr$^{3+}$ can dramatically change this conclusion.

Avalanche starts from a weak absorption from ytterbium ground state in a $^2F_{7/2} \rightarrow ^2F_{5/2}$ phonon side band or by multiphonon assisted absorption [21]. After the energy transfer ($^2F_{5/2}$,$^2H_4$) $\rightarrow$ ($^2F_{7/2}$,$^1G_4$), $^1G_4$ is populated and by resonant absorption of the pump beam, it is possible to find an excited Pr$^{3+}$ ion in the $^3P_0$ level. The latter can relax towards the $^1G_4$ level promoting at the same time an Yb$^{3+}$ ion from the $^2F_{7/2}$ to the $^2F_{5/2}$ levels: ($^3P_0$,$^2F_{7/2}$) $\rightarrow$ ($^1G_4$,$^2F_{5/2}$). Then, the excited Yb$^{3+}$ ion can transfer again its energy to another Pr$^{3+}$ ion according to: ($^2F_{5/2}$,$^2H_4$) $\rightarrow$ ($^2F_{7/2}$,$^1G_4$). At the end of these two energy transfers or cross-relaxations, two Pr$^{3+}$ ions are in the $^1G_4$ level and are ready to absorb the excitation beam, leading to two ions in the $^3P_0$ level. Emissions from the latter to levels located at lower energies than $^1G_4$ will result in upconversion of the excitation beam. The whole process is characterized by a sharp threshold in the upconverted emission intensity as a function of the excitation intensity which explains the term “avalanche”.

![Fig. 1. Energy levels and processes involved in Pr,Yb avalanche upconversion. Thick arrows: pump transitions (rate $R_1$ and $R_2$); thin arrows: energy transfers (rates $r$ and $s$); thick dashed line: orange laser transition.](image-url)
The complete Yb,Pr system shown in Figure 1 involves 11 levels and is clearly too complex for a rate equation analysis. The system was therefore reduced to 5 levels with several assumptions. First of all, we considered samples with small Pr$^{3+}$ concentrations, so that Pr$^{3+}$-Pr$^{3+}$ interactions are neglected. Moreover, Yb$^{3+}$-Pr$^{3+}$ energy transfers are non resonant and back transfers are therefore neglected.

Then, since efficient avalanche in Pr$^{3+}$ requires a long lived $^{1}G_4$ level, only low-energy phonon hosts (cut-off frequency $<550$ cm$^{-1}$) are of interest: the energy gap $\Delta E$ between $^{1}G_4$ and $^{3}F_4$ is about 3000 cm$^{-1}$ and is too small to avoid strong multiphonon relaxation when phonon frequency are higher as in many oxide hosts. However, even in BaY$_2$F$_8$ where the phonon cut-off frequency is only 350 cm$^{-1}$ [22], $^{1}G_4$ lifetime is only 38 $\mu$s at low concentration [23] whereas its radiative lifetime is $\approx$ 1.06 ms [24]. This corresponds to a quantum efficiency of 36%. The non-radiative relaxation rate $W_{NR}$ is given by:

$$W_{NR} = C \exp(-\alpha\Delta E)$$  \hspace{1cm} (1)

where $C$ and $\alpha$ are phenomenological parameters, with $\alpha$ typically on the order of $3-5 \times 10^{-3}$ cm$^{-1}$ [25] (approx. $5 \times 10^{-3}$ cm in BaY$_2$F$_8$ [24]). The $^{3}F_4$, $^{3}F_5$, $^{3}F_2$, $^{3}H_6$ and $^{3}H_5$ levels all exhibit energy gaps to the level immediately below lower than 2200 cm$^{-1}$, the latter value being obtained for the $^{3}H_6$ level. According to the above equation, the non-radiative relaxation rates will be at least 10 to 50 times larger for these levels than for the $^{1}G_4$ one. Their lifetimes will therefore be at most a few $\mu$s. We can therefore safely ignore these levels populations in the rate equations. This analysis is also confirmed by the fact that no self terminating behavior has been reported for the orange laser transition $^{3}P_0 \rightarrow ^{3}H_6$ in BaY$_2$F$_8$.

Finally, the population of the $^{1}D_2$ level can also be neglected since the latter is not populated directly by the excitation beam or the energy transfers. Moreover, the $^{3}P_0$-$^{1}D_2$ energy gap is 4000 cm$^{-1}$ which results in very weak non-radiative relaxation. Radiative relaxation from the $^{3}P_0$ is also negligible because of the small corresponding $U^{(2)}$ reduced matrix element : 0.0134 (see Sect. 4).

In conclusion, the rate equations describing the system have been limited to the $^{3}H_4$, $^{1}G_4$, $^{3}P_0$, $^{2}F_{7/2}$ and $^{2}F_{5/2}$ levels which are denoted the numbers 1 to 5 in Figure 1. The time-dependent system reads:

$$\frac{dn_1}{dt} = w_{21}n_2 - rn_1n_5 + w_{31}n_3$$  \hspace{1cm} (2)

$$\frac{dn_2}{dt} = -w_{21}n_2 + wn_1n_4 + wn_3n_4 + wn_5n_2 - R_2n_2 + R_2n_3$$  \hspace{1cm} (3)

$$\frac{dn_3}{dt} = -w_{31}n_3 - w_{32}n_3 - wn_3n_4 + R_2n_2 - R_2n_3$$  \hspace{1cm} (4)

$$\frac{dn_4}{dt} = -R_1n_4 + R_1n_5 + wn_1n_5 - wn_4n_3$$  \hspace{1cm} (5)

$$\frac{dn_5}{dt} = -w_{51}n_5 - wn_1n_5 + wn_3n_4 + wn_5n_2 - R_5n_5$$  \hspace{1cm} (6)

where $n_i$ is the population of level $i$, $w_{ij}$ is the relaxation rate from level $i$ to level $j$ and $r$ (respectively $s$) the $^{3}H_4$-$^{2}F_{5/2} \rightarrow ^{1}G_4$-$^{2}F_{7/2}$ (respect. $^{3}P_0$-$^{2}F_{7/2} \rightarrow ^{1}G_4$-$^{2}F_{5/2}$) energy transfer rates. $R_1$ and $R_2$ are the $^{2}F_{7/2} \rightarrow ^{2}F_{5/2}$ and $^{1}G_4 \rightarrow ^{3}P_0$ pumping rates. Pr$^{3+}$ and Yb$^{3+}$ populations are related to their total respective concentrations $N_{Pr}$ and $N_{Yb}$ by $n_1 + n_2 + n_3 = N_{Pr}$ and $n_4 + n_5 = N_{Yb}$. In the above equations, stimulated emissions induced by the excitation are included to take into account population saturations. These effects can occur at high pumping rates as those used to obtain orange avalanche laser action.

All parameters used in the following simulations are given in Table 1 and correspond to a 1.25 at.%Pr, 6 at.%Yb:BaY$_2$F$_8$ sample [18]. The rate equations have been solved numerically using Matlab software to give the steady state populations.

A plot of level 3 population as a function of the total pump intensity $I_p = I_{P_1} + I_{P_2}$ is shown in Figure 2, curve (a). $I_{P_i}$ is related to the pump rate $R_i$ by:

$$I_{P_i} = \frac{\hbar c R_i}{\lambda \sigma}$$  \hspace{1cm} (7)

where $\lambda$ is the transition wavelength and $\sigma$ its cross-section. The fraction of ground state pumping, $f = I_{P_2}/I_{P_1}$, as well as the maximal pump intensity were taken according to reference [18]. On curve (a), the avalanche threshold is clearly seen around 150 kW/cm$^2$ and is characterized by a very fast variation of $n_3$ with $R$. For higher pumping rates, this variation is much lower, which is more suitable to a stable laser operation.

Before turning to numerical simulations, it is worthwhile to look for analytical solutions of the steady-state solutions of the above rate equations. However, this is only possible for a zero ground state absorption ($R_1 = 0$), i.e. ideal avalanche upconversion. In this case, two conditions have to be fulfilled to get a population in level 3:

1. The cross-relaxation rates $s$ and $r$ have to be large enough compared to the $w_{31}$ relaxation rate.
2. The condition is satisfied, population appears in level 3 when the pumping rate $R_2$ exceeds a threshold value.

The first condition can be found by assuming that there is initially some small populations in levels 3 and 5 and a very strong $R_2$ pumping rate so that all ions in level 2 are promoted to level 3. The condition for level 3 and 5 populations to sustain or grow is found by setting population gains higher than losses in these levels (emissions stimulated by the pump are neglected):

$$sN_{3}n_3 + w_{32}n_3 + rN_{Pr}n_5 \geq n_3(w_{31} + w_{32}) + sN_{Yb}n_3$$  \hspace{1cm} (8)

$$sN_{5}n_5 \geq n_5w_{51} + rN_{P}n_5$$  \hspace{1cm} (9)

where $n_1$ (respect. $n_4$) have been set to $N_{Pr}$ (respect. $N_{Yb}$) assuming that the system is close to threshold. This gives finally the condition:

$$srN_{Pr}n_5 \geq w_{31}n_3$$  \hspace{1cm} (10)

which shows that when $w_{31}$ is too large no avalanche upconversion can be observed. If the above condition is fulfilled, solving equation (2) in the steady state gives the
matrix elements of some Pr\textsuperscript{3+} elements effectively increase emission from level 3 to levels lower than level 2 (i.e. up-dramatically reduce level 3 population. Since stimulated threshold pumping rate:

$$R_{2t} = \frac{w_{31} s N_{\gamma} r N_{\gamma} + s w_{51} + r w_{31} + w_{31} w_{51}}{s N_{\gamma} r N_{\gamma} - w_{31} r N_{\gamma} - w_{31} w_{51}}$$

(11)

showing that the avalanche pumping threshold increases when $w_{31}$ increases. For a given pumping rate, this can dramatically reduce level 3 population. Since stimulated emission from level 3 to levels lower than level 2 (i.e. up-converted emissions) will effectively increase $w_{31}$, this effect is of particular concern and will be discussed in the next section.

### 3.2 Influence of ground state pumping

Figure 2 (curve (b)) shows level 3 population as a function of the total pump intensity with a ground state pumping fraction $f$ of 0.23. This could be achieved by a second pump beam at 980 nm in addition to the main one at 840 nm. Since there is a substantial ground state absorption, the system is no more in an avalanche regime. The threshold has completely disappeared and level 3 population is higher than in the case of avalanche upconversion regardless of the total pump intensity. However, the difference is much larger at low pump powers, when the avalanche process has not yet occurred. The effect of the ground state pumping is due to the possibility to populate level 2 even if level 3 population is very small. In the avalanche regime this is not possible because the (3,4)$\rightarrow$(2,5) cross-relaxation, which is the only way to populate level 2, is inefficient for low level 3 populations. This also explains that at high pump powers, the difference between the two pumping schemes is reduced, because level 3 population is high and the cross-relaxation becomes roughly as efficient as the ground state pumping. In Figure 2, for a total pump intensity of 500 kW/cm\textsuperscript{2}, level 3 population is only 1.19 times larger with the ground state pumping compared to avalanche but at 100 kW/cm\textsuperscript{2}, this ratio reaches 150.

For a given total pumping rate, there is an optimal ground state pumping fraction which gives the highest level 3 population, since excited state absorption along the 2-3 transition is always necessary to reach this level. The corresponding curves are shown in Figure 3 (labels a,b,c) for three total pump intensities. As expected from the preceding paragraph, a more pronounced maximum (at $f \approx 0.19$) is observed at low pump intensities (curve (c)). For curves (b) and (a), the maxima shift towards lower ground state pumping fractions ($f \approx 0.1$) for increasing $I_P$ and the overall variations of population are smaller. These high pump intensities are more likely to be found in a laser configuration so that it may seem that the ground state pumping is of limited interest. However, when laser action occurs, a strong intra-cavity stimulated emission rate has to be sustained. In the rate equation model, the latter will simply add to the $w_{31}$ rate and will substantially increase the avalanche threshold and reduce level 3 population. In this case, an additional ground state pumping will have a large effect. This is seen in Figure 3 where the curves (d) and (e) which include stimulated emission rates, show again pronounced maxima around a

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**Table 1.** Parameters used in the Pr,Yb rate equation model (corresponding to 1.25%Pr\textsuperscript{3+}, 6%Yb\textsuperscript{3+}:BaY\textsubscript{2}F\textsubscript{8} [18]) and reduced matrix elements of some Pr\textsuperscript{3+} transitions.

<table>
<thead>
<tr>
<th>Relaxation rates (s\textsuperscript{-1})</th>
<th>Transition from $^3P_0$ to:</th>
<th>$(U^{(2)})^2$</th>
<th>$(U^{(4)})^2$</th>
<th>$(U^{(6)})^2$</th>
</tr>
</thead>
<tbody>
<tr>
<td>$w_{31}$ 18181.8</td>
<td>$^3\text{H}_4$</td>
<td>0</td>
<td>0.1714</td>
<td>0</td>
</tr>
<tr>
<td>$w_{32}$ 1027 s\textsuperscript{-1}</td>
<td>$^3\text{H}_6$</td>
<td>0</td>
<td>0</td>
<td>0.0726</td>
</tr>
<tr>
<td>$w_{2}$ 26315.8 s\textsuperscript{-1}</td>
<td>$^3\text{F}_2$</td>
<td>0.2943</td>
<td>0</td>
<td>0</td>
</tr>
<tr>
<td>$w_{5}$ 500 s\textsuperscript{-1}</td>
<td>$^3\text{F}_4$</td>
<td>0</td>
<td>0.1213</td>
<td>0</td>
</tr>
<tr>
<td>Transfer rates (s\textsuperscript{-1} cm\textsuperscript{3})</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>$r$ 1.6 x 10\textsuperscript{-16}</td>
<td>$^1\text{G}_4$</td>
<td>0</td>
<td>0.0425</td>
<td>0</td>
</tr>
<tr>
<td>$s$ 2.3 x 10\textsuperscript{-16}</td>
<td>$^1\text{D}_2$</td>
<td>0.0134</td>
<td>0</td>
<td>0</td>
</tr>
</tbody>
</table>

Transitions cross-sections (cm\textsuperscript{2})

- $\sigma_a$ (840 nm): 10\textsuperscript{-19}
- $\sigma_a$ (980 nm): 10\textsuperscript{-19} (estimated)
- $\sigma_a$ (607 nm): 5 x 10\textsuperscript{-19}

Concentrations (cm\textsuperscript{-3})

- $N_{P_3}$: 8 x 10\textsuperscript{19}
- $N_{Yb}$: 3.8 x 10\textsuperscript{20}

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**Fig. 2.** $^3P_0$ level population as a function of the pumping rate. Fraction of ground state pumping: 10\textsuperscript{-4} (a), 0.23 (b).
ground state pumping fraction of 0.21. These curves were computed by replacing $w_{31}$ by $w_{31} + \sigma_c \lambda I_l/hc$ where $I_l$ (respect. $\sigma_c$) is the intensity (respect. emission cross-section) at the laser wavelength $\lambda$, around 607 nm. The laser intensity used for curve (d), 250 kW/cm$^2$, corresponds to an intra cavity power of about 1.7 W in the setup of reference [11] and therefore to 17 mW of output power with a 1% transmitting output mirror, as observed in the same paper. An interesting point is that the optimal value of the ground state pumping fraction is similar between the case of high excitation and stimulated emissions and of low excitation intensity without stimulated emission. This may allow one to optimize the pumping scheme without a laser cavity.

The reduction of level 3 population when the stimulated emission rate increases has important consequences for an Pr$^{3+}$/Yb$^{3+}$ upconversion laser: an increase in the laser threshold and a reduction of the output power. The latter effect can be roughly accounted for by adding a photon rate equation to the population ones. The former reads:

$$\frac{d\phi}{dt} = c \sigma_n n_3 \phi - \frac{\phi}{\tau_c}$$

where $\phi$ is the photon density at the laser wavelength and $\tau_c$ is the photon lifetime inside the cavity. The latter is given by:

$$\tau_c = \frac{2l}{c} \frac{1}{\ln(r_1 r_2)}$$

where $l$ is the cavity length, $c$ the light speed and $r_1$ (respect. $r_2$) is the input (respect. output) mirrors reflectivity. At steady-state, level 3 population, $n_3$, is fixed by equation (13) to the value $(c \sigma_c \tau_c)^{-1}$ and satisfies as well the other rate equations (Eq. (2)) where $w_{31}$ has been replaced by $w_{31} + \sigma_c \lambda I_l/hc$ (see above). For a given total pump intensity and ground state fraction pumping, we can compute the photon density in the cavity which will give the above level 3 population. The output intensity is then deduced from the output mirror transmission. The corresponding curves are shown in Figure 4 with a cavity length of 10 cm [11]. The transmissions used in the computations are quite high compared to those used in usual laser experiments (a few %). Accordingly, the calculated output intensities are also higher by roughly an order of magnitude than those reported in reference [11]. This is due to our very crude model which does not take into account any losses apart from the mirrors, the pump beam absorption within the crystal, and other parameters like the beam profiles. The computed curves have therefore to be considered as mainly qualitative. The conclusions we draw from them should however still remain valid if all factors were taken into account. Curves (a–c) correspond to a 15% output transmission and three different pump intensities. They show again a clear maximum at a ground state pumping fraction of $f = 0.23$. At this value, the output intensity is between 68 and 28 times higher than in the avalanche scheme ($f = 10^{-4}$) when the total pump power is varied from 250 to 1000 kW/cm$^2$. As discussed above, the effect of the ground state pumping is stronger for lower excitation and larger laser intensities. This is clearly seen on curve (d) where the output transmission has been increased to 30%. The intra cavity laser intensity is then lower and the optimal ground state pumping gives an output intensity only 18 times larger than in the avalanche case. For the same reason, it can be seen that at low ground state pumping fraction, a larger transmission gives a better result (compare curves (b) and (d) for $f < 10^{-3}$).

In conclusion, the different simulations show that an additional excitation beam tuned to Yb$^{3+}$ absorption offers considerably improved performance for a Yb$^{3+}$/Pr$^{3+}$ upconversion laser. This pumping scheme recycles efficiently the ions which relax to the ground state through stimulated emission. This allows one to obtain higher

**Fig. 3.** Level 3 population as a function of the ground state pumping fraction. (a–c): no stimulated emission, total pump power density 500 kW/cm$^2$, 250 kW/cm$^2$, 100 kW/cm$^2$. (d, e): 1–3 stimulated emission 100 kW/cm$^2$, 250 kW/cm$^2$, total pump power 500 kW/cm$^2$.

**Fig. 4.** Laser output intensity as a function of the ground state pumping fraction. (a–c): output transmission 0.15, total pump power 1000 kW/cm$^2$, 500 kW/cm$^2$, 250 kW/cm$^2$; (d): output transmission 0.3, total pump power 500 kW/cm$^2$. 

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laser output intensities or to use lower total pump power. This last point may be advantageous given the very high pump intensities necessary to obtain laser emission in the avalanche regime [11].

We now address the second important point to be able to use an orange avalanche upconversion laser for high-pump intensities necessary to obtain laser emission in the avalanche regime.

### 4 Avalanche upconversion in Pr,Yb:PbF₂

β-PbF₂ is a fluorite type cubic crystal corresponding to the Fm3m space group. Rare earth ions substitute Pb²⁺ in a clearly established cubic site and, especially at higher concentrations in other sites which natures are not completely known [26, 27].

Figure 5 shows the absorption spectrum of a 1 at.%Pr³⁺, 2 at.%Yb³⁺:PbF₂ sample. The Pr³⁺(respect. Yb³⁺) atomic concentrations used to compute the cross-sections were 1.9 × 10⁻²⁰ cm² (respect. 3.8 × 10⁻²⁰). The peak cross-section for Yb³⁺ (0.9 × 10⁻²⁰ cm²) is quite high compared to other fluoride hosts including Yb³⁺:BaY₂F₈ (maximum value: 0.75 × 10⁻²⁰ cm² for E//z [28]) and Yb³⁺:CaF₂ (0.4 × 10⁻²⁰ cm² [29]). This confirms that at high concentrations, Yb³⁺ do not occupy sites of cubic symmetries [27] for which electric dipole transitions are forbidden. No data were found to compare Pr³⁺: absorption cross-sections, but emission values quoted for Pr³⁺:BaY₂F₈ suggest that the peak transitions cross-sections are significantly weaker in PbF₂. This may be due to the large linewidths of the transitions (see below) which is also common in this type of fluoride crystals where several sites are possible for the rare earth ion.

The emission spectrum obtained under pumping at 856 nm, i.e., in avalanche regime, is reported in Figure 6. Besides the intense peak in the blue region corresponding to the ³P₀ → ³H₄ transition, the emission is dominated by the ³P₀ → ³H₆ orange transition. The latter peaks at 606.0 nm (606.18 nm in vacuum) with a FWHM of 5 nm.

It is therefore in very good resonance with the Pr³⁺ transition from the ground state which lies at 665.977 nm (in vacuum) in Y₂SiO₅ [30]. Another striking point is the very low intensity of the ³P₀ → ³F₂ transition around 635 nm compared for example to LiYF₄ in σ polarization [15].

These differences in emissions intensities are well accounted for by the Judd-Ofelt theory. The transition intensity is proportional to the line strength S given by:

\[
S(J, J') = \sum_{\lambda=2,4,6} \Omega_{\lambda} \left( J \right| \left( U^{(\lambda)} \right| J' \right)^2
\]

where the Ω parameters are usually determined from absorption experiments and the \(( J \left| U^{(\lambda)} \right| J' \) reduced matrix elements tabulated [31] and independent of the host. Their values for the transitions from the ³P₀ level are given in Table 1. Due to the selection rule \( J = \lambda \) for the final level of the transitions, most of the reduced matrix elements vanish which explains the selective enhancement of some transitions. For example, the weak intensity of the ³P₀ → ³F₂ transition in PbF₂ corresponds to a very small Ω₂ parameter but this has no effect on the ³P₀ → ³H₆ transition which is solely determined by Ω₆. Given the large variations of the Ω parameters among Pr³⁺ doped crystals, it should be possible to find hosts with maximal orange emission, i.e. weak Ω₂ and Ω₁ parameters. This may be important since the ³P₀ → ³H₆ transition has a rather small reduced matrix element.

The excitation spectrum of the orange emission corresponding to the avalanche regime is shown in Figure 7. Two broad bands are recorded peaking at 857 nm and 828 nm and with respective FWHM of 20 and 13 nm. The main band is suitable for pumping by commercially available GaAs/AlGaAs high power laser diodes and its width should favor a stable absorbed power versus pump wavelength shifts. This is especially important for stable single mode operation.
Fig. 7. Excitation spectrum of Pr,Yb:PbF$_2$ monitoring emission at 605.8 nm.

Fig. 8. Emission intensity at 605.8 nm as a function of the excitation power at 856 nm in Pr,Yb:PbF$_2$.

The orange emission intensity was recorded as a function of the pump power at 856 nm (Fig. 8). The “S” shape is characteristic of avalanche upconversion with a threshold of about 750 mW ($\approx 15$ kW/cm$^2$) and a saturation regime which begins to appear at the highest pump power. These results are very close to those obtained with Pr$^{3+},$Yb$^{3+}$:BaY$_2$F$_8$ [18].

5 Conclusion

Pr$^{3+},$Yb$^{3+}$-co-doped materials could be used to build a single frequency orange laser pumped in the infrared region and suitable for high resolution spectroscopy. Although an orange Pr$^{3+},$Yb$^{3+}$ avalanche upconversion laser has been demonstrated, simulations based on a rate equation model show that the use of a second pumping source in resonance with Yb$^{3+}$ transition (and therefore also in the infrared) leads to dramatic improvement in the laser performance. This is explained by the inefficiency of the avalanche process to excite back to the upper levels the ions which have decayed to the ground state. Since stimulated emission strongly populates the ground state level at the expense of the upper level, avalanche pumping is unable to maintain large upper state populations in the laser regime. In opposition, a resonant pumping from the ground state to an intermediate state allows the pumping between excited states to efficiently repopulate the upper level. Computations also show that the optimal fraction of the ground state pumping can be determined without laser action, by simply recording upconverted fluorescence intensity under low pumping rate.

A new host, Pr$^{3+},$Yb$^{3+}$PbF$_2$, is also proposed to meet the requirement that the orange laser emission must be in resonance with the transition probed. In particular, the most used rare earth doped crystal for quantum information application is Pr$^{3+}$:Y$_2$SiO$_5$ in which the $^3$H$_4 \rightarrow ^1$D$_2$ absorption occurs at 605.977 nm (in vacuum) with a linewidth of a few GHz. This wavelength lies outside the emission spectra of the crystal used so far for orange laser (Pr$^{3+},$Yb$^{3+}$BaY$_2$F$_8$). This is not the case with Pr$^{3+}$ emission in PbF$_2$ (along the $^3$P$_0 \rightarrow ^3$H$_6$ transition) which is centered at 606.18 nm (vacuum value) with a FWHM of 4 nm. Avalanche upconversion has been observed in this compound under pumping at 856 nm. This wavelength is in the range of high power laser diodes. Further investigations, including addition of a resonant pumping and laser tests, are in progress.

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References


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