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High-power diode-pumped cryogenically-cooled Yb:CaF$_2$ laser with extremely low quantum defect

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Doped fluoride crystals have been known and identified as attractive laser media since the very beginning of the lasers [1]. In the field of ultrahigh-power laser with high repetition rate, very interesting laser development at cryogenic temperature has been done with Yb:YLF [2-3] and Yb:CaF$_2$ [4,5]. In the case of Yb:CaF$_2$, crystallographic and luminescence properties were already known a long time ago [6]. However, the characteristics of these materials have not been fully exploited, especially at high dopant concentration [7-10]; a broad emission band, a long emission lifetime of 2.3 ms, with moreover a good thermal conductivity [11,12] and at last, but not least, a well-mastered growing process for high-quality and large single crystals. Thus, Yb doping now allows calcium fluoride to strike back for high power laser applications; they indeed become in few years among the most promising laser materials for high-power/high-power diode-pumped laser systems [13-16].

In this letter we present another typical property of Yb:CaF$_2$, which allows for efficient laser emission in a “quasi-two-level” laser scheme without any ultra wavelength-selective or narrow-linewidth intracavity element. This could represent a breakthrough for ultrahigh-power lasers since it could lead both to efficient diode-pumped laser operation and to a minimal thermal load [5,17-19]. Moreover, as this quasi-two-level laser system operates at cryogenic temperature, it brings another positive advantage for high power laser devices, which consists in improving the thermal properties of the laser element such as its thermal conductivity.

Yb$^{3+}$ has a simple electronic-level structure based on only two manifolds ($^{7}F_{7/2}$ and $^{7}F_{9/2}$) which splits into different crystal-field Stark sublevels whose number and energy separation depend on the symmetry and the strength of the local crystal-field environment. Due to charge compensation and minimum-energy arrangements of the ions in this system [6], the case of heavily doped Yb:CaF$_2$ is very particular. Indeed, the luminescent centers, responsible for the laser properties of the material, give rise to a typically relatively weak crystal-field and reduce crystal-field splittings of the Yb$^{3+}$ energy levels (see in Fig. 1a).

Consequently, in addition to the broad-band vibronic structure, which extends from about 1000 to 1060 nm, and to the zero-phonon lines (corresponding to the so-called “zero-line” around 981 nm), there is another set of clear lines at 987 nm and 991.5 nm with substantial emission/absorption cross sections. They correspond to zero-phonon transitions from the lowest $^{7}F_{9/2}$ (emitting level at about 10200 cm$^{-1}$) to the second and third energy levels of the $^{7}F_{7/2}$ ground multiplet (around 50 and 110 cm$^{-1}$), respectively.

**Fig. 1a** Spectroscopic lines of Yb:CaF$_2$ at 77 K. (b) Experimental measurements of pump and laser wavelengths for pumping at 981 nm (blue curve) or 986 nm (red curve) and gain cross section of the Yb:CaF$_2$ at 77 K and for $\beta=0.1$ (purple curve).

Thus, laser operation at short-wavelength and “ultra-low quantum-defect” is possible by cooling the laser crystal down to Liquid Nitrogen (LN$_2$) temperature, as presented in the letter.

The experiments were performed with a 2.2%-Yb-doped, 5-mm-long fluoride crystal. The experimental setup is displayed in Fig. 2. In order to pump the crystal longitudinally and to allow simultaneously an extremely short wavelength separation between pump and laser, a
broadband HR mirror of 2-mm-diameter glued on a 25-
mmdiameter AR plate is implemented. This pump-beam-
occultering mirror uses the advantage of the large diameter
pump beam (collimated fiber-coupled laser diode with NA
= 0.22) compared to the laser beam inside the Yb:CaF$_2$
resonator, forming as a so-called modal multiplexer.
The corresponding losses observed on the pump beam do
not exceed 4%. Moreover, the laser is free from any
spectral selection and operates very efficiently at its
maximum spectral gain without additional losses.

Fig. 2: Experimental setup.

According to the emission and absorption spectra of Yb:CaF$_2$
registered at low temperature [5], the laser should have naturally operated, without extra wavelength
selector, at 992 nm for an inversion population ($\beta$) higher
than 10%. However, it is not the case, since laser
operation remains fixed at 1034 nm, even for an average
inversion population higher than 40%. This is probably
due to uncertainty on the temperature elevation in the
crystal. In order to favor the short wavelengths emission
with a minimum of losses, we insert in the cavity (Fig. 2) a
wavelength selector consisting of two high-pass dichroic
mirrors highly reflective between 980 and 1000 nm with
losses per bounce <0.1% at 992 nm, <1% at 997 nm and a
high transmission (>95%) around 1030 nm. Consequently,
the impact of this selector is acceptable and allows
efficient laser at low wavelengths. In these conditions,
laser operation occurs between around 992 nm and 997
nm.

Figure 3a displays the output power obtained for different output couplers. Thereby, we determine the
different gain of the crystal at varying population
inversion levels, estimated using the following equation:

$$\exp(\frac{1}{N} (\beta \sigma_{\text{em}} + (1 - \beta \sigma_{\text{abs}}) l) - 1) = 1$$  \hspace{1cm} \text{Eq. 1}

where $N$ is the Yb-dopant concentration, $\sigma_{\text{em}}$, $\sigma_{\text{abs}}$ are the emission and absorption cross sections at the laser
wavelength ($\lambda_L$), $l$ is the length of the crystal, $T_{OC}$ is the
output coupler transmission and $L$ the losses (vs $\lambda_v$).

At low inversion ($\beta < 0.05$, e.g. $T_{OC} = 5\%$), only 997 nm is
observed, whereas at intermediate levels ($0.05 < \beta < 0.08$
the gain is flat between 993-997 nm, e.g. for a 10% output
coupler ($\beta = 0.065$), the laser operates simultaneously at
997.1, 994.2 and 993.0 nm. At higher inversion levels
($\beta > 0.08$) the laser operates around 992 nm for an output
coupler of 15% ($\beta = 0.093$) or higher the laser wavelength
lies between 989.7 and 992.0 nm.

The best CW laser performance at 992 nm has been
obtained with the 15% OC ($\beta = 0.11$) with a laser emission
of 33 W for 93 W absorbed pump power (under laser
operation). The laser efficiency is then 35 % (Fig. 3b). The
measured small signal gain is found to be equal to 2.7.

The laser and pump emission wavelengths were
measured simultaneously. Figure 1b reports these pump
and laser wavelengths at the maximum output power. On
the same graph the gain cross section is also plotted for
the value $\beta = 0.1$ corresponding to the optimal power. The
predominance of the gain at 992.0 nm clearly appears,
corroborating the experimental results. The mean
emission wavelength is 992.7 nm and the mean pump
wavelength is 980.7 nm, which corresponds to a very low
laser quantum defect $\eta_{QD} = 1 - \lambda_L/\lambda_p = 1.2\%$. Those results
clearly indicate the strong potential of Yb:CaF$_2$ used at
cryogenic temperature for high power laser developments
where efficiency and heat load are an issue.

It is interesting to note that for broadband amplification
(992-997 nm), the optimal operation should be obtained at
low inversion levels.

Exploiting such small quantum defect configuration
might be challenging and it is worth considering a
number of points. It is important to identify that thermal
loads in ytterbium-doped laser materials come from three
types of non-radiative relaxations: the laser quantum
defect ($\eta_{QD} = 1.2\%$) between pump and laser photons,
the fluorescence quantum defect ($\eta_{QD} = 3.1\%$) between
the pump and fluorescence photons and non
radiative desexcitations from $^2$F$_{5/2}$ to $^2$F$_{7/2}$ levels, evaluated in our case to 0.7 % of the absorbed pump photons [12].
Then, 1.2%×35% (35%=laser efficiency) of these absorbed
pump photons heats the crystal by laser quantum defect
and 3.1%×64% by fluorescence quantum defect.
Consequently, for a total absorbed pump power of 93W,
this leads respectively to 0.65 W, 0.39 W and 1.85 W (total
of 2.9 W).

This clearly indicates that in a small quantum defect
laser the thermal loads due to the fluorescence quantum
defect cannot be neglected. Therefore, the laser efficiency
directly impacts on the thermal loads. In our experiment,
the efficiency is limited by the losses providing from the
not fully-ideally-coupled cavities of the uncoated facet of
the crystal and/or to residual pollution due to our non-
perfect cryostat vacuum.

The second issue to be considered, especially at high
pump power, is the thermal conductivity of the laser
material. From this point of view, Yb:CaF$_2$ is particularly
interesting since its thermal conductivity at LN$_2$
temperature rises up to 68 W/m/K for an undoped
This means that the thermal loads can be efficiently evacuated and that the thermal lensing effects should be greatly reduced. This is clearly what we noticed in our experiments since no thermal lensing effect was observed even at full pump power (Fig. 3b).

![Diagram of theoretical emission wavelength and theoretical percentage of absorbed pump power at 986 nm versus β.](image)

Fig. 4: Theoretical emission wavelength and theoretical percentage of absorbed pump power at 986 nm versus β. Experimental laser wavelength and laser efficiency have been plotted for the optimal value β=0.09 (15 % OC).

A last point which can be noticed by examining the gain cross section for β=0.1 reported in the figure 1 is the absorption line at 986 nm in order to decrease further the laser quantum defect. As plotted in the figure 4, the theoretical absorption of our crystal at 986 nm (for a single pump pass) is only 30% at maximum (and without saturation) and becomes null for β=0.21 (or equivalently for a laser at 992 nm and with Tc=39%). On the other hand, we have to remember that there is also a constraint for β to emit at 992 nm. As a matter of fact, the optimal inversion population was found around 0.09 which corresponds in our case to an output coupler of 15 %. The experiment was performed and we obtained a laser output power of 4 W for an absorbed pump power of 35 W (efficiency of 11 %). The laser wavelength (Fig. 1b and Fig. 4) was 992.9 nm, leading to a low quantum defect of 0.7 %. In conclusion, we have demonstrated, simultaneously for the first time, a low quantum defect, highly efficient and high-power laser operation of an Yb:CaF2 laser crystal at cryogenic temperature. This represents an important step towards practical lasers based on Yb:CaF2 operating at very high power levels. As a matter of fact, by using the simple figure of merit given by the ratio (thermal conductivity)/(quantum defect), we find record values of 5700 W/m/K for pumping around 981 nm and 9700 W/m/K around 986 nm[14]. Moreover, Yb:CaF2 really appears as a favorable material for such a kind of low quantum-defect laser operation (more than any other material) because of the existence of this sharp emission peak around 992 nm with a substantially high emission cross section. Such a peak does not exist in a system like Yb:CALGO [15], Yb:KCGdLu(WO4)2 [17], materials which also gave rise to a very low-quantum defect laser operation, but with a much lower laser efficiency. Such a peak exists in the case of Yb:YLF [18]. However, the lowest quantum defect which could be (theoretically) obtained would be around 2% and the one achieved so far, by pumping around 960nm (which is not a common diode wavelength) was around 3.5%. Yb:CaF2 has then this rare property of having clear peaks very close to the zero-phonon-line, which is very auspicious to efficient ultra-

low-quantum-defect diode-pumped laser. Moreover concerning absorption improvement, this can be done using pump-recycling such as in thin disks.

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