Direct Comparison of Yb^{3+}:CaF_2 and heavily doped Yb^{3+}:YLF as laser media at room temperature

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Abstract: We report an extensive comparison of the laser performances of diode-pumped Yb^{3+}:YLF (30% at.) and Yb^{3+}:CaF_2 (5% at.) crystals, lasing at room-temperature and operating in two different operation mode, i.e. Continuous Wave (CW) and quasi-CW. An in-depth investigation of the crystals behavior by changing the pump power, clearly shows the crystal absorption depends on the lasing conditions. Therefore, we report an unambiguous definition of the slope efficiency calculated taken into account the real measured crystal absorption under laser action. Finally, we present a study of problems related to thermally induced losses which are expected influencing the laser performance.

OCIS codes: (140.0140) Lasers and laser optics; (140.3580) Lasers, solid-state; (140.5680) Rare earth and transition metal solid-state lasers; (140.3600) Lasers, tunable; (160.3380) Laser materials.

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1. Introduction

The development of new classes of short-pulses and high-peak-power laser sources, playing a fundamental role across many research fields, including high-resolution spectroscopy, non-linear frequency conversion or medical surgery, is deeply related to the capability to produce innovative lasing materials with peculiar emission and thermo-optical properties. The most important requirements are: first, a broad emission spectrum which allows for the generation of short pulses; second, a long upper-state lifetime that depending on the host materials, influences the energy storage capability; last but not the least, a high thermal conductivity, in order to handle the pump-generated thermal load in high power applications.

To comply with all requirements, in recent years there has been an increasing interest in Yb-doped materials, i.e. crystals, glasses or ceramics. The simple laser energy level scheme of Yb^{3+} ion (consisting of only two manifolds i.e. $^2F_{5/2}$ and $^2F_{7/2}$, with a relatively small quantum defect between absorption and emission), allows a very efficient lasing in the near infrared, with a broad emission band which permits to obtain a wide range of tunability in CW and Q-switching regime or short duration pulses in mode-locking. Moreover the small diameter of the ion permits a heavy doping of host materials without a significant reduction of the emission lifetime. The main drawback in Yb-doped materials is a not negligible absorption at the lasing wavelength due to the thermal population of the lower levels of the laser transition. This requires a high pump intensity to reach the laser threshold.

Several host material (e.g. oxides, fluorides, vanadates, borates or tungstates) are reported in literature, each of them with some particular and complementary characteristics. The direct comparison between the literature data regarding the laser performances achievable with different hosts materials and different doping levels is somehow complicated by the differences in the various experimental set-up, such as variations in the pump wavelength and geometry, cavity schemes, crystal cooling systems, mirrors reflectivity spectra, tuning devices.

The aim of this study is to provide a direct comparison of the laser performances with the same experimental set-up to reduce the influences of the apparatus differences of two fluorinated crystals, namely Yb^{3+}:CaF_2 [1,3,4] and Yb^{3+}:YLF [2,5–7], to establish a clear comparison of the achievable laser performances. These materials are very interesting because of their very wide tuning range as well as for their long upper level life-time (>2 ms), which is much higher respect to the majority of the mostly used Yb doped hosts (see for instance [8] and references therein). This makes them very attractive for amplifier laser systems and generation of ultrashort pulses [11].

Therefore, the experiments were carried out on samples having similar geometries, with the same cavity layout and elements, and pumping device. Great attention was placed in the assessment of the effect of the pump-induced thermal load on the laser performance, by analyzing the behavior of the energy conversion and slope efficiency of the two laser devices under CW and quasi-CW pumping mode, with various Duty Factors (DF). Finally, we present the explored tunability range.

2. Crystals Growth and their Properties

Among the fluoride host crystals, CaF_2 shows good thermal properties [9]. The incomplete charge compensation of the dopant (Yb^{3+}) and the smaller replaced ion diameter (Ca^{2+}), hampers the use of high doping levels. The rearrangement of the crystal structure for the compensation of the charge defect results in a diversified site structure, which leads to a broader band emission fluorescence. This in turns determines a wide tunability [9], but on the other hand it determines the formation of clusters which can produce alternative de-excitation paths. Moreover, CaF_2 is easily fabricated in bulk by standard method, *i.e.* Bridgman and

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Czochralski, or in thin films by Molecular Beam Epitaxy [10]. Due to its optical isotropy a simpler laser cavity is required.

Conversely YLF allows for heavy Yb doping because of the exact ion charge substitution. The upper state lifetime is comparable with CaF_2, *i.e.* 2 ms [8]. Its thermal conductivity is lower than that of the CaF_2, and it further decreases for increasing Yb^{3+} doping, see [12]. Concerning the polarization properties, YLF is an uniaxial crystal which permits two possible polarization states, *i.e.* π-pol and σ-pol, with different absorption and emission cross-section spectra. A preliminary investigation of the spectroscopic and laser properties of heavily doped samples, performed by our group, is reported elsewhere [6].

As for the pumping wavelength and geometry, both crystals feature a moderate and relatively constant value of absorption cross section around in the 920-950 nm window. With heavily doped Yb:YLF it is possible to obtain high absorption constants (of the order of 10-20 cm^{-1}), well suited for longitudinal pumping of the short (few mm) crystals with fiber-coupled diode lasers; conversely, with CaF_2 (which allows only for moderate doping) it is more difficult to obtain a strong pump absorption over a short crystal length in this wavelength region. Both Yb:YLF and Yb:CaF_2 can be pumped on the zero-phonon line, respectively at 958 and 976 nm. The Table 1 resumes the main spectroscopic and thermal properties of these crystals.

Table 1. Spectroscopic and thermal properties of Yb:YLF and Yb:CaF_2

Parameter	Yb:YLF	Yb:CAF_2
Absorption cross section @ 940 nm	$1.5x10^{-21}cm^2$ (π) [6] $2.7x10^{-21}cm^2$ (σ) [6]	$1.8x10^{-21}cm^2$ [4]
Peak absorption cross section and λ	$12.3x10^{-21}cm^2$ (π, 958 nm) [6]	$4.1x10^{-21}cm^2$ (980 nm) [4]
Absorption cross sect. @ 1050 nm	$0.057x10^{-21}cm^2$ (π) [6]	$0.11x10^{-21}cm^2$ [4]
Emission cross sect. @ 1050 nm	$0.78x10^{-21}cm^2$ (π) [6]	$1.5x10^{-21}cm^2$ [4]
Upper laser level lifetime	2 ms [6]	2.4 ms [13]
Thermal conductivity (5% doping)	4.7 $Wm^{-1}K^{-1}$ [12]	5.2 $Wm^{-1}K^{-1}$ [14]

Crystal growth of Yb-doped CaF_2 was performed in a vacuum-tight Czochralski system equipped with an automatic diameter control system. The resistive heater and thermal insulators were made of high-purity graphite. The starting materials were prepared from high-purity commercial (Stella Chimifa, Japan) fluoride powders of CaF_2 and YbF_3 (>99.99%). The concentration YbF_3 in the starting material was 5 mol%. For further details about the CaF_2 crystal growth, see [15].

The Yb^{3+}:YLF lattice investigated in the experiment has been well described in [6]. Briefly, it was grown by a Czochralski technique employing LiF and YF_3 powders (from AC Materials, Tampa, Fla, Usa) as raw material for the lattice and adding a proper amount of YbF_3 (>99.99%) powder to achieve a concentration of ytterbium as high as 30% at.

3. Experimental Set-Up

The experimental apparatus is sketched in Fig. 1. The laser cavity is V-shaped with a folding half angle of 10° and with arm lengths of 78 mm (between the mirrors FM and EM) and 430 mm between the mirrors FM and OC. We have experimentally tested several values of this length (*i.e.* between 200 and 600 mm) and we have found that for both crystals the laser output has only a weak dependence from the arm length. The chosen value maximizes the output power for both crystals. Several flat output couplers, OC, with transmission ranging from 1.5% to 20% were used. The crystal was carefully oriented with the facets perpendicular to the cavity axis, in order to re-inject the Fresnel reflection of the uncoated crystal faces into the cavity itself. The crystals (1.5 mm length for YLF, 2 mm for CaF_2) are welded with Indium on a copper heat sink. The heat sink is cooled with a Peltier device and stabilized at 18°C. The pump source is a laser diode emitting at 940 nm, coupled into an unpolarised fiber with 200 μm core diameter and a numerical aperture of 0.44 (full angle). The measured pump intensity distribution in the focal plane results about Gaussian with a spot radius around 150 μm @ $1/e_2$. The laser was tuned by placing a tuning prism made of SF10 glass, with an apex

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angle of 60 degrees into a cavity arm between the output coupler and the folding mirror. The laser emission is tuned by tilting the OC-mirror around an axis perpendicular to the prism dispersion plane. The emission wavelength was measured with a fiber coupled, 60 cm focal length spectrometer equipped with a multichannel detector (spectral resolution 0.4 nm).

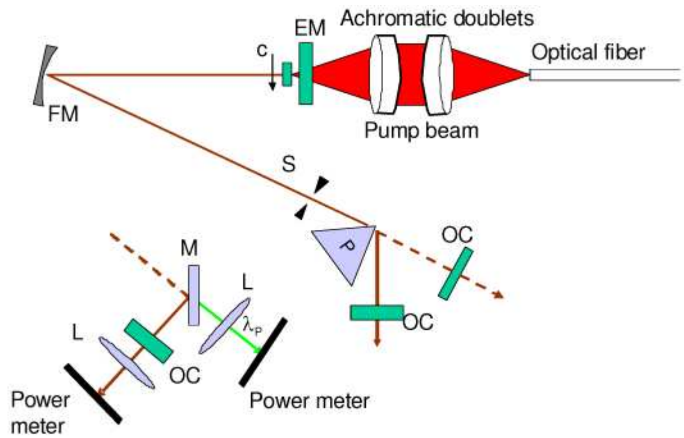

Fig. 1. Tunable Laser Cavity. EM: End Mirror (flat); FM: Folding Mirror (radius of curvature 155 mm); OC: Output Mirror (flat); C denotes the direction of the crystal optical axis; P: tuning prism; S: slit. The inset shows the non-tunable cavity configuration used to measured the fraction of the pump power absorbed from crystals. M: flat mirror, F: pump beam filter, L: convergent lens

4. Experimental Results and Discussion

In quasi-three level systems the pump absorption under high intensity levels and in lasing condition can be different from the linear (*i.e.* low intensity) absorption, due to the combination of two competing effects: the saturation of the absorption because of the depletion of the lower laser level and the population draining from the upper laser level caused by the laser action [10]. Moreover the heating of the pumped volume can modify the thermal population distribution in the lower manifold. On the other hand the accurate assessment of the crystal absorption is needed for the determination of the laser threshold and efficiency. To clarify these aspects, we have investigated the behavior of the crystals absorption at high pump intensities, when the laser action is on and off, and under different thermal load conditions.

In absence of the laser action, we have measured the fraction of power absorbed by the crystal for different DF (from 20% to 100%), with the same pump power range and focusing conditions used in the laser experiments. The measurement of the absorbed pump power when laser action is switched on was obtained by measuring the residual pump power transmitted through the crystal. To carry out this measurement, we modified the non tunable cavity of Fig. 1 by adding a flat folding mirror in the arm between the FM and the OC mirrors, which allowed the extraction of the pump fraction transmitted by the crystal and reflected by FM. The switching off of the laser action is achieved by obstructing the cavity arm between the folding mirror and the output coupler. The residual pump beam is collected by a converging lens and focused on a power meter. A short pass filter rejects the small amount of the laser radiation that leaks from the coating of the FM.

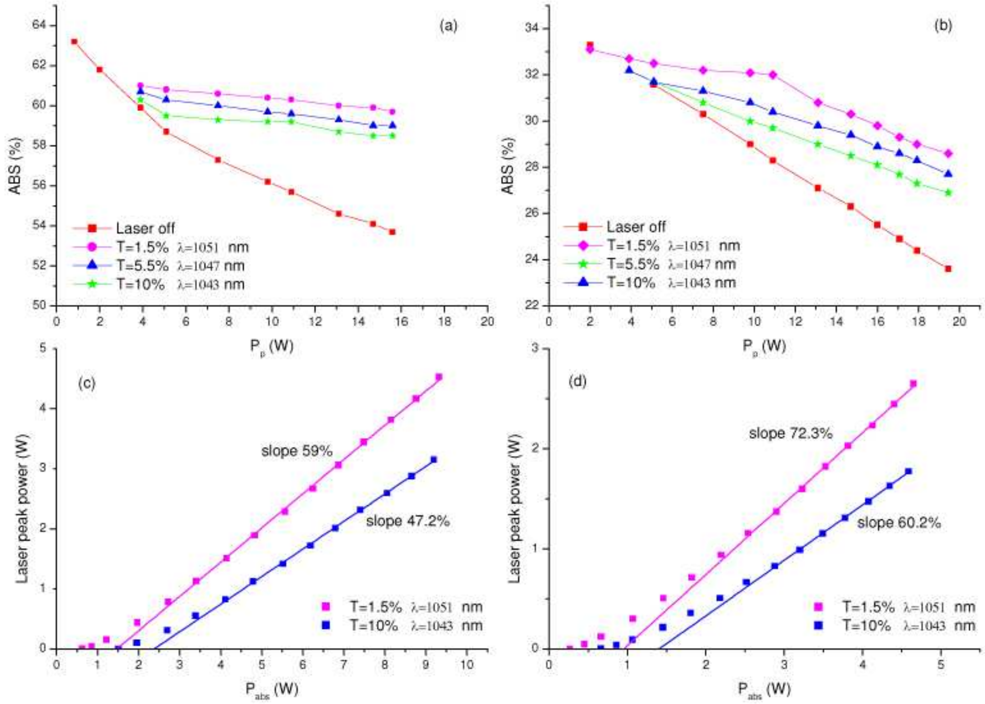

Fig. 2. (a)-(b) Fraction of the pump peak power (ABS) absorbed from Yb^{3+}:YLF (a) and Yb^{3+}:CaF_2 (b) either when the laser is off (red points) or active by employing an output coupler mirror with a transmission ranging from 1.5% to 10%. (c)-(d) report the slope efficiency for Yb^{3+}:YLF and Yb^{3+}:CaF_2 respectively, obtained taken into account the results in (a)-(b), with different output coupler mirrors.

Figure 2(a) and 2(b) report the results obtained by using various output couplers. In non-lasing condition we observed a decrease of several percent of the absorbed pump peak power fraction for increasing incident peak power, due to the absorption saturation. In the case of CaF_2 the absorption decreases of about 10%, and for YLF the absorption decreases of about 7%. For both crystals, when the laser action is turned on the absorption increases to a level between the saturated absorption for the same incident pump peak power and the unsaturated absorption.

For the Yb^{3+}:YLF, the absorption in lasing conditions is closer to the unsaturated absorption, while it remains fairly constant when the pump peak power increases; for CaF_2 the absorption monotonically decreases for increasing pump level.

This behavior can be clarified by considering the dependence of the saturated absorption in presence of both an intense pump and laser field. It can be shown [10] that if the laser intensity I_L in the crystal is much higher than the saturation intensity at the laser wavelength λ_L, defined as

$$I_{Lsat} = hc/\left[\lambda_L \tau\left(\sigma_{emL} + \sigma_{abL}\right)\right] \tag{1}$$

(σ_{emL} and σ_{abL}: emission and absorption cross sections at the laser wavelength respectively), then the crystal absorption coefficient tends to the asymptotic value

$$\alpha_{asym} = N_0 \sigma_{abL}\left(I_{Lsat}/I_{P\min}\right) \tag{2}$$

where N_0 is the dopant density, and I_{Pmin} is the pump intensity needed to bleach the absorption at the laser wavelength, given by

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$$I_{P\min} = hc/\left[\lambda_L \tau\left(\sigma_{emP} - \left(\sigma_{abP}\sigma_{emL}/\sigma_{abL}\right)\right)\right] \quad (3)$$

(σ_{emP} and σ_{abP} : emission and absorption cross sections at the pump wavelength respectively). It is worth to note that for a given pump wavelength this asymptotic absorption coefficient depends on the laser wavelength through σ_{emL} and σ_{abL}.

For Yb:YLF one has to consider separately the contribution of the π and σ polarization for the pump, and the fact that the laser is π-polarized, so that Eq. (2-3) modify as it follows

$$_{\pi,\sigma}\alpha_{asym} = {}_{\pi,\sigma}\sigma_{abL} N_0 \left({}_{\pi}I_{Lsat} / {}_{\pi,\sigma}I_{P\min}\right) \quad (4)$$

$$_{\pi,\sigma}I_{P\min} = hc/\left[\lambda_L \tau\left({}_{\pi,\sigma}\sigma_{emP} - {}_{\pi,\sigma}\left(\sigma_{abP}\sigma_{emL}/\sigma_{abL}\right)\right)\right] \quad (5)$$

where $_{\pi,\sigma}\sigma_{emP}$ and $_{\pi,\sigma}\sigma_{abP}$ are the emission and the absorption cross sections at the pump wavelength for the π and σ polarization respectively.

The saturation intensity for CaF_2 at 1049 nm is 41 kW/cm^2, in comparison with a circulating laser intensity of the order of few MW/cm^2. The asymptotic absorption coefficient for CaF_2 in presence of an intense laser field at 1049 nm has then a value of 1.7 cm^{-1}, that correspond to an absorption of 28.8% over a crystal length of 2 mm. For YLF, the saturation intensity at 1051 nm is about 80 kW/cm^2. Asymptotic absorption coefficients are respectively 9.9 cm^{-1} for the σ component and 4.2 cm^{-1} for the π component, under a laser field at 1050 nm with π polarization. The polarization-averaged pump absorption over the crystal length is then 59.6%. Both these values are in agreement with the experimental findings.

According to experimental results, we conclude that the laser efficiency with respect to the absorbed pump peak power of Yb^{3+}:CaF_2 and Yb^{3+}:YLF must be calculated by accounting for the actual crystal absorption at each lasing configuration.

For the assessment of the laser threshold and of the laser efficiency we measured, without any wavelength or polarization selective element in the cavity, the output peak power as a function of the absorbed peak power. To determine the effect of the thermal load on the laser performance, we pumped the crystals either in CW mode or in quasi-CW mode, *i.e.* by modulating the pump laser, that is the DF, with square pulses at low repetition frequency, 10 Hz, and with a pulse lengths going from 20 ms to 80 ms. The rise and the fall time of the pump (50 µs) are much shorter than the pulse duration, as well as the rise and the fall time of the laser emission, for both crystals. Therefore, during the pump-on periods the Yb laser behaves substantially as it would in CW mode under the same instantaneous pump power, but with respect to the pure CW case the period averaged pump power (and therefore the period averaged thermal load) are reduced to a fraction equal to the Duty Factor. Regarding the polarization, with the Yb^{3+}:YLF the laser output is linearly polarized along the crystal optical axis, with a polarization ratio greater than 100:1; Yb^{3+}:CaF_2 output has a random polarization.

We have found that the efficiency of the heavy-doped Yb^{3+}:YLF are lower than the low-doped Yb^{3+}:CaF_2, see Fig. 2(c)-(d). Under quasi-CW pumping, we measured for Yb^{3+}:YLF crystal a threshold absorbed pump peak power P_{th} = 0.62 W, a maximum output peak power P_{out}= 4.5 W, a slope efficiency of 59% respect with an absorbed peak power P_{abs}= 9.5 W, while for Yb^{3+}:CaF_2 we found P_{th}= 0.26 W, a maximum output peak power P_{out}= 2.6 W with a slope efficiency of 72.6%, close to the to the quantum defect, with P_{abs}= 5 W. The lower threshold of Yb:CaF_2 with respect to the Yb:YLF can be ascribed to the fact that the emission cross section at the lasing wavelength for Yb:CaF_2 is about twice than Yb:YLF (see Table 1). Therefore, for a given population inversion density the effective small signal gain is correspondingly higher.

The trends of crystal efficiency are confirmed in CW-operating mode as well. The discrepancy at lower value of absorbed pump peak power between the experimental and theoretical curves, which are expected linear, may be addressed to a re-absorption effects due to the presence of population at the lower level of the laser transition

In principle, thermal load effects of the gain medium may play a significant role on the laser performance because it induces thermal lensing effects which can increase the resonator

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geometrical losses. Furthermore the temperature rise in the pumped region increases the thermal population of the lower laser level, thus enhancing re-absorption effects. The situation is worsened by the fact that the thermal conductivity of the crystal decreases for increasing temperature.

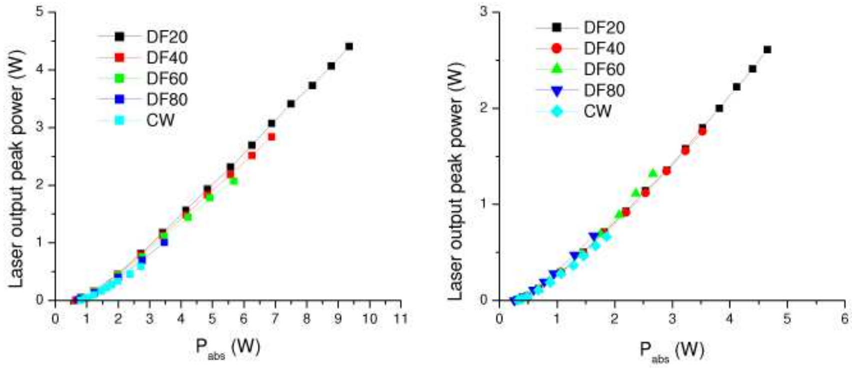

Fig. 3. Laser peak power as a function of the absorbed pump peak power from Yb^{3+}:YLF (a) and $Yb^{3+}:CaF_2$ (b) obtained by changing the Duty Factor from 20% to 100%. OC transmission is 1.5%.

We investigated the cumulative result of these effects by measuring the laser peak power with several pumping DF. We remind that using a quasi-CW operating mode the heating of the crystal addressed to the pump beam may be reduced increasing the overall laser efficiency. Yb^{3+}:YLF slope efficiency decreases dramatically from 59% to 31% by increasing the duty factor while $Yb^{3+}:CaF_2$ slopes appear to be unaffected by the thermal load, see Fig. 3(a)-(b). For both crystals the threshold values result unchanged. Unfortunately, a numerical modeling of the thermal behavior of these two crystals, which could provide useful hints to explain the observed differences, is hampered by the fact that the relevant thermo-mechanical and thermo-optical coefficients for the heavily doped YLF are unknown.

Regarding the emission range of the samples, we have measured the output peak power as a function of wavelength. The largest tuning band, measured using an OC-mirror with an overall transmission of 1.5% and a pump peak power of 5.95 W (DF of 20%), was obtained

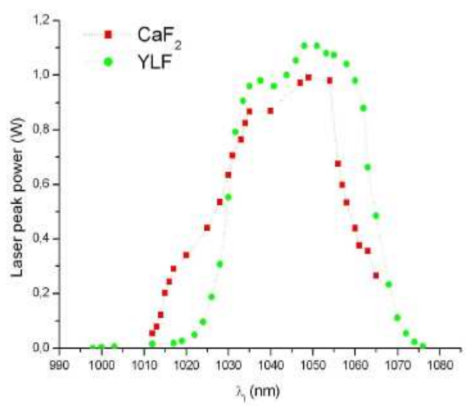

Fig. 4. Laser peak power versus the laser wavelength is reported. For both crystals, the pump peak power was 5.95 W, while the transmission of the output coupler mirror was 1.5%.

with Yb^{3+}:YLF, exceeding 78 nm, *i.e.* from 997 nm to 1075 nm. It is worth to point out that the overall cavity mirrors reflectivity (calculated over the cavity round trip) was almost constant between 97% and 98% in the interval 997-1100 nm. Therefore the tuning curves shown in Fig. 4 are not distorted by the mirror reflectivity curves. Unfortunately, the

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maximum pump power is severely limited by the damaging threshold of active media preventing the achievement of an even wider tunability. In principle, considering the FWHM of the tuning curves (31.7 nm for Yb:CaF_2 and 34.2 nm for Yb:YLF), pulses as short as 36 fs for Yb:CaF_2 and 34 fs for Yb:YLF could be generated.

5. Conclusions

In this paper we present an extensive study of the laser properties of Yb^{3+}:YLF and Yb^{3+}:CaF_2 crystals pumped in quasi-CW and CW mode. The samples have a similar geometry but different concentration of dopants. The performances were compared using the same experimental apparatus and pumping device. With the Yb^{3+}:YLF we were able to obtain a output peak power of 4.5W at 20% of pump duty factor, and a slope efficiency of 59%. With Yb^{3+}:CaF_2 the maximum output peak power was 2.6 W, but the slope efficiency with respect the absorbed pump peak power was as high as 72.6%. This is one of the highest ever reported for this laser material and close to the quantum defect limit. We have to keep in mind that in this work, we used a clearly defined slope efficiency unambiguously calculated starting from the absorption as measured when the crystals are lasing. In fact, we showed that the crystal absorption depends on the lasing condition. Special care was placed in the evaluation of the effects of the pump-induced thermal loads on the laser performance. In this sense, the CaF_2 was found much less sensitive to thermal effects than YLF.

Table 2. Performance and Comparation of Yb:YLF and Yb:CaF_2

Crystal *	Yb:YLF (30% at.)	Yb:CaF_2 (5% at.)
Laser Peak Power (W)	4.53 (P_{abs} = 9.32 W)	2.65 (P_{abs}= 4.65 W)
Slope Efficiency (%)	59	72.3
Absolute Efficiency (%)	47	52
Range Tuning (nm)	78	55
Laser Threshold, P_{th}, (W)	0.62	0.26

* In both Crystals: T_{OC} = 1.5%, λ_l = 1051 nm, DF = 20%

Finally, we explored the potentiality of the tunability of the two crystals. We have found that for equal conditions of absorbed pump peak power and cavity losses the Yb:YLF shows both a wider tuning range and a higher output peak power than CaF_2 (see Table 2).

In conclusion, these measurements show unambiguously that Yb:CaF_2 can be better suited than Yb:YLF for the applications where the crystal is subjected to a high thermal load (*e.g.* when a high CW power output is sought), because of its lower threshold, higher slope efficiency, and relative insensitivity to thermal effects. To take full advantage of this output power capabilities, probably it is better to pump CaF_2 on the zero phonon line at 980 nm, rather than at 940 nm, in order to obtain a large pump absorption even with low doping level.

On the other hand, Yb:YLF shows a clear advantage with respect to Yb:CaF_2 in the applications where a broad emission spectrum is needed, for instance for the development of broadly tunable sources, or for the generation of ultrashort pulses. Furthermore, the possibility of high doping levels allows for more flexibility in the choice of the pumping wavelength.

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