Laser performance of diode-pumped Yb:CaF2 optical ceramics synthesized using an energy-efficient process

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Due to their naturally improved toughness and also to a flexible and cheap production process, laser ceramics could provide an interesting alternative to single crystals to meet the challenges posed by the demanding domain of ultrafast and high power laser operations. In this frame, we are investigating the laser performance of a $Yb:CaF_2$ ceramics developed with a simple and green synthesis process. A 4 % at. doped ceramic sample was diode-pumped to deliver an output power of 1.6 W with an optical-to-optical efficiency reaching 25%, a slope efficiency of 43 %, a gain of 1.4 and wavelength tunability from 1015 to 1060 nm. Results are comparable to typical single $Yb:CaF_2$ crystal performances therefore opening many pertinent applications for these greener and low-cost $Yb:CaF_2$ ceramics in high power diode-pumped lasers.

OCIS codes: (140.3380) Laser materials; (140.3480) Lasers, diode-pumped; (140.3615) Lasers, ytterbium; (140.3600) Lasers, tunable

A renewed and still growing interest in ytterbium-doped media has recently emerged with the apparition of high power and high brightness pump sources at 980 nm such as InAsGa laser diodes. The simple electronic structure of Yb^{3+}, composed of only two multiplets, forbids most of the parasitic effects of up-conversion encountered in Nd^{3+}-doped materials. As an amplifying ion, Yb^{3+} also allows a quasi-three level laser scheme, leading to low quantum defects and low thermal loads. Moreover, Yb^{3+} ions have a significantly longer fluorescence lifetime and broader absorption and emission bands than most of the rare-earth ions operating in the near infrared; these properties ensure efficient energy storage and broader wavelength tunability for laser operation. Given these reasons, Yb^{3+}-doped materials are nowadays well-established in the industrial and academic laser community, specifically for high power, 100s-of-fs laser systems [1,2].

Yb^{3+} has been used as dopant in various crystalline materials and glasses [1,2]. Amongst them, ytterbium-doped calcium fluoride offers the advantage of being at the frontier of glasses and crystalline materials, possessing properties inherent to both material types. The crystalline nature of calcium fluoride (Space group, $Fm\overline{3}m$) provides higher thermal conductivities than optical glasses and with a thermal conductivity of 9.7 $W.m^{-1}.K^{-1}$ [3,4] is even similar to crystalline optical materials (YAG: 11 $W.m^{-1}.K^{-1}$). On the other hand, in terms of optical properties, rare-earth (RE)-doped calcium fluorides are known for providing atypical spectroscopic properties that can be mainly explained by the charge compensation of a trivalent RE^{3+} substitution on a Ca^{2+} site [5]. Electron spin resonance analysis confirmed that RE^{3+} dopant ions and charge compensating interstitial F^- were spontaneously organized as clusters [6] (whether dimers, tetramers or most commonly hexamers) instead of isolated dopant ions, even at a rather low doping content (clusters become predominant around 0.5 % at.) [4,7]. This preferential organization in clusters is the source of the specific spectroscopic behavior of RE-doped calcium fluoride. This effect can be either detrimental by favoring up conversion energy transfers for dopants allowing it (Nd^{3+}, Er^{3+}, Tm^{3+}…etc.), thus reducing excited levels lifetimes [5,8], or productive in the case of Yb^{3+}, by broadening the absorption and emission bandwidths [6,9-11]. The absorption spectrum of $Yb:CaF_2$ extends from 880 nm to 1040 nm and the emission one from 1000 nm to 1070 nm, approaching then the spectral broadness of Yb-doped glasses obtained by inhomogeneous broadening. Supplementary advantages of calcium fluoride include having a low phonon energy (~495cm^{-1}, almost twice as low as YAG one) and a negative thermo-optic coefficient ($dn/dT=-11.5.10^{-6}K^{-1}$) [12] which can be used for compensating positive geometric distortions [13].

The combined properties of Yb^{3+} and CaF_2 are promising for high energy, high power, ultrafast laser operation and as a result, Yb:CaF_2 single crystals have recently been utilized in a TW-level, femtosecond high power laser chain [14]. The long lifetime of Yb:CaF_2 favors energy storage and makes this material very attractive for high energy diode-pumped laser amplifiers. On the other hand, the broad spectral bandwidth of Yb:CaF_2 can be exploited to produce a pulse duration among the shortest obtainable with Yb-doped materials[15].

Nevertheless, issues related with single crystals such as depolarization, low thermo-mechanical resistance and high production costs still limit the performance and the wide dissemination of Yb:CaF_2. In high power laser systems, depolarization due to the stress can strongly degrade the laser performance by inducing losses. Moreover, polishing of highly doped crystals (typically > 3%) remains difficult [16], often resulting to poor surface quality and a lower damage threshold. Thermal shock sensitivity of the crystals also requires particular precautions.

Improving the crystal growth process can only partially address the aforementioned issues. Consequently, ceramics becomes an attractive, and economical, alternative for enhancing the properties of Yb:CaF_2. The polycrystalline character of ceramics and the random orientation of the grains make the material more isotropic and less sensitive to stress and depolarization. Moreover, utilizing ceramics presents benefits such as: no macroscopic dopant segregation, a better thermo-mechanical resistance, almost no size or shape restrictions, and the ability to produce complex doping architectures [16,17]. However, only a few studies on the performance Yb:CaF_2 laser ceramics have been reported up to date [18-20].

The ceramics used in this study were based on Yb:CaF_2 nanopowders obtained through a previously described soft chemistry process [7,21]. A green body was formed from these powders using a newly developed strainless packing method [22] and sintered at moderate temperatures (typically < 60% of the melting temperature) in air and without any pressure assistance. Thus, unlike a single-crystal based ceramic synthesis process, as reported in [19], this method does not require an energy-intensive processing step such as a preliminary single-crystal growth, or a high temperature pressure-assisted sintering step.

The samples were then cut up according to the desired dimensions and polished with colloidal silica (~100 nm particles). The UV-Visible-NIR scattering coefficients, α_s, of 4 % at. Yb^{3+}-doped ceramic samples were calculated according to the following equation,

$$\alpha_s(\lambda) = -\frac{1}{\varepsilon}.ln\left(\frac{T(\lambda)}{(1-R(\lambda))^2}\right)$$

in which ε is the sample thickness in cm, T(λ) the transmittance and R(λ) the sample reflectivity at the considered wavelength, and results are shown in figure 1.

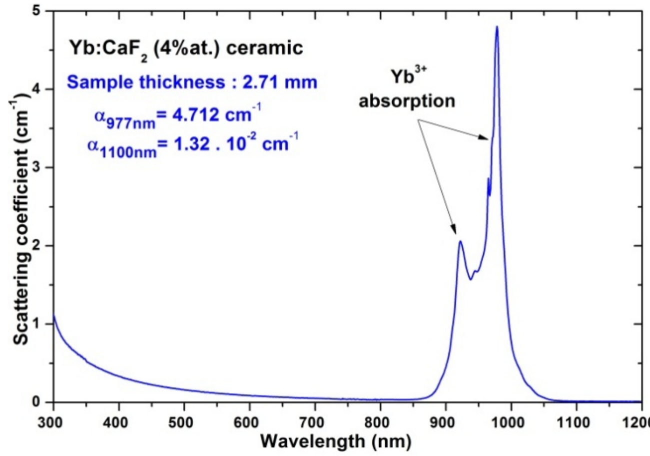

Fig. 1. Room temperature scattering coefficient of a 4 % at. doped Yb:CaF_2 ceramic. (Strong absorption in the 875 nm-1050 nm range is observed due to Yb^{3+} ions)

A significant short-wavelength scattering (below 500 nm), caused by the ceramic nanometer-sized residual defects was observed. However, the ceramic optical losses around 1100 nm were found to be quite low at 1.32 10^{-2} cm^{-1}, testifying the good optical quality of the ceramic samples.

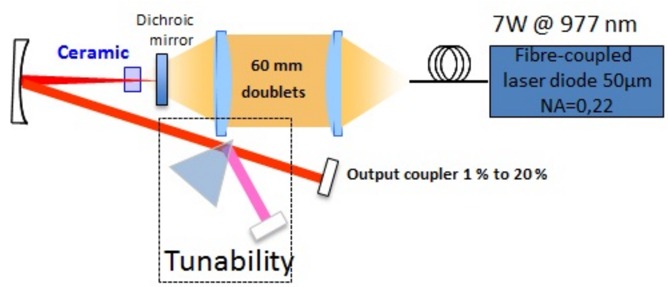

Fig. 2. Experimental cavity setup used for samples test

The laser performances of the ceramics were evaluated at room temperature in a V-shaped cavity setup, illustrated in figure 2. The setup was composed of a flat dichroic mirror, a concave mirror (100 mm) and an output coupler (O.C.) with a transmission ranging from 1 % to 30 %. The samples did not have antireflection coatings but Fresnel reflections were coupled back in the cavity to reduce losses. The samples were pumped by a continuous wave fiber-coupled laser-diode (50 µm core diameter, numerical aperture of 0.22) delivering up to 7.5 W power, with maximum wavelength emission intensity at 977 nm). The pump beam was imaged 1:1 to match the cavity laser beam. The maximum calculated incident power on the sample, taking into account the pump-optics and the Fresnel losses of the input facet, was 6.9 W. The 4 % doped ceramic used was 2.71 mm long and, at full power, absorbed 3.2 W, indicating absorption saturation. Figure 3 shows the obtained laser powers as a function of the incident pump power for different O.C. transmission coefficients.

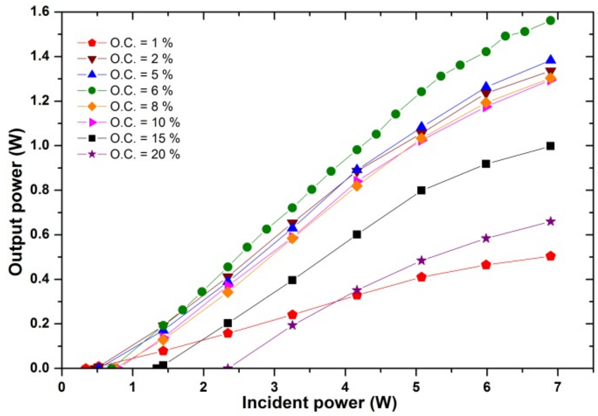

Fig. 3. Laser power for a 4 % at. Yb^{3+}-doped CaF_2 ceramic (thickness: 2.71 mm) as a function of the incident pump power depending on the O.C. transmission.

A maximum power of 1.6 W was reached for an O.C. transmission value of 6% with an oscillation threshold of 680 mW. For comparison, a 3.8 mm long, 4.5 %at. doped single-crystal was tested under the same conditions and a maximum power of 2.3 W was obtained with a threshold at 600 mW. At a first glance, the single crystal seems to have a better performance than the ceramic. However, for a fair assessment, the samples must be compared at similar absorption considering saturation issues. To quantify the effect of the pump saturation absorption, the transmitted-pump variation has been measured with and without laser operation. Fig 5. shows the output power of both samples with respect to the uncorrected absorbed power, while these results are corrected for saturation effects in Fig. 6. The (longer) single crystal absorbs almost 6 W in laser operation and the ceramic only 4 W. For a more accurate comparison with the single-crystal and with previously published works, we compared the slope efficiencies of three cases: versus the incident pump power (fig. 4), versus the absorbed pump power without absorption saturation correction (fig. 5) and versus the exact absorbed pump power taking this last effect into account (fig. 6). The different curves for the single-crystal and ceramic are plotted in fig. 4, 5, 6. For the ceramic, the slope efficiency versus the incident pump power is 26 % compared to 38% for the single-crystal. The "common" slope efficiency -versus the uncorrected absorbed pump power- is 58 % for the ceramic compared to 79 % for the single-crystal.

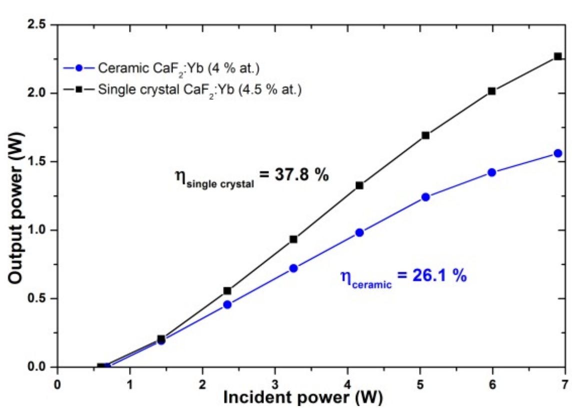

Fig. 4. Comparison between a 4 %-doped $Yb:CaF_2$ ceramic and a 4.5 % $Yb:CaF_2$ single crystal laser efficiency curves versus incident pump power.

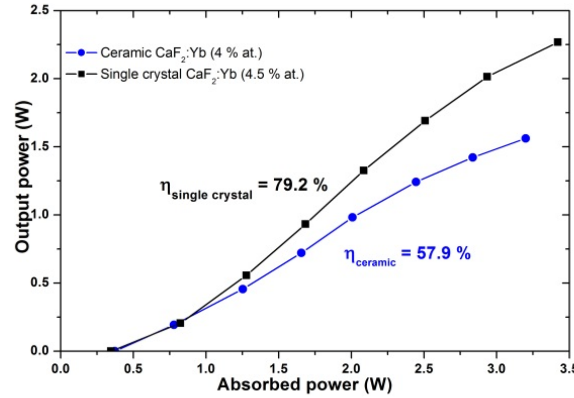

Fig. 5. Comparison between a 4 % doped $Yb:CaF_2$ ceramic and a 4.5 % $Yb:CaF_2$ single crystal laser efficiency curves versus absorbed pump power neglecting saturation effects

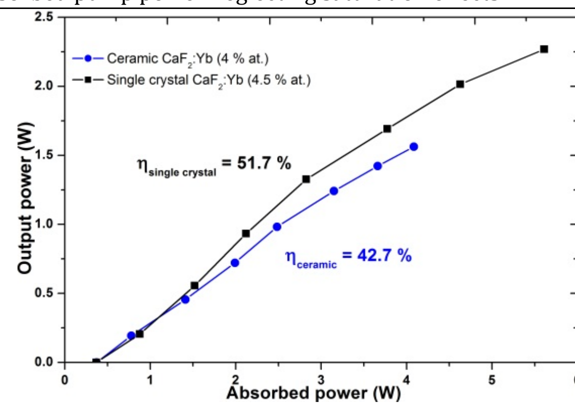

Fig. 6. Comparison between a 4 % doped $Yb:CaF_2$ ceramic and a 4.5 % $Yb:CaF_2$ single crystal laser efficiency curves versus "real" absorbed pump power (*id est* considering saturation effects)

These values are only interesting to be compared to some previous works where the absorption saturation evolutions are rarely taken into account. And finally, the slope efficiency versus the real absorbed pump power for the ceramic is 43 %, compared to 51 % for the single-crystal. In this last case, one can clearly evaluate the good performance of the ceramic in terms of laser gain. The results are also advantageously comparable to previously reported ones, whether for single-crystal-based ceramics [19], or nanoparticles-based ceramics [20].

Laser operation was obtained with various output couplers. The maximum laser power versus output coupler transmission coefficients is plotted in figure 7. The maximum O.C. transmission permitting laser operation was 20% and we can then extrapolate the gain in our condition at 1.4. By inspecting the guideline in fig. 7, laser operation was observed with the 30 % OC, but only for few seconds due to thermal issues, clearly indicating the gain limit.

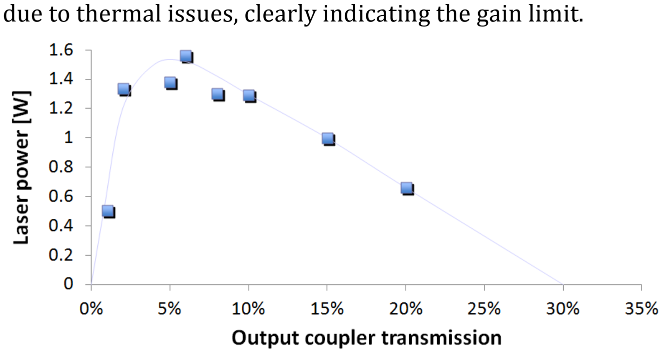

Fig. 7. Maximum laser powers versus O.C. values

The natural wavelength emission varied with the output coupler from 1030 nm to 1052 nm depending of the gain cross sections. With the 6 % O.C., the emission was around 1051 nm as presented in figure 8.

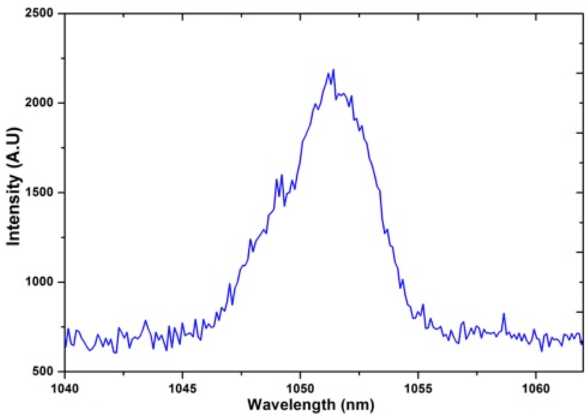

Fig. 8. Natural laser emission spectrum for 1.6 W obtained with the 6 % O.C.

The tunability of the laser wavelength was measured by inserting a dispersive prism in the collimated arm of the cavity, as presented in figure 2. The results of the measurements are shown in figure 9. The ceramic sample was continuously tunable over a range of 45 nm, from 1015 nm to 1060 nm, with a maximum output power at 1051 nm. The ceramic sample and single crystal exhibited similar tunability behaviors indicating *a priori* the same spectral signature of clusters in the spectrum. [23].

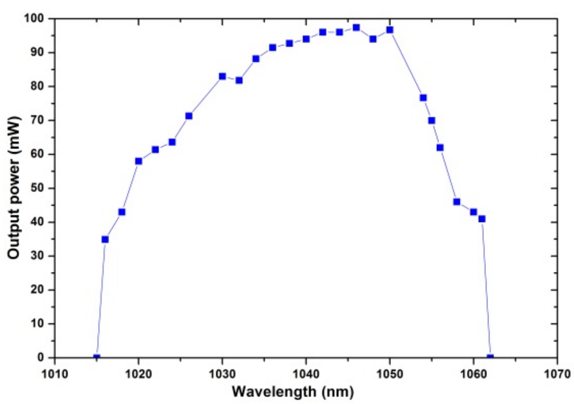

Fig. 9. Room temperature tunability measured on a 4% at. doped $Yb:CaF_2$ ceramic by prism dispersion of the cavity beam

In conclusion, we have demonstrated laser operation of nanopowder-based $Yb:CaF_2$ ceramics for the first time and have obtained exceptionally promising results including a significant slope efficiency (> 40 %) and threshold comparable to single crystal performance. A maximum output power of 1.6 W was achieved. This represents, to our knowledge, the best performance ever obtained with $Yb:CaF_2$ ceramics. Smooth and broadband wavelength tunability was verified, with a maximum power centered at 1051 nm and range of 45 nm. These results signify an important breakthrough for Yb-doped ceramics by having a similar performance with $Yb:CaF_2$ single-crystals. Furthermore, the viability of the ceramic process and production of $Yb:CaF_2$ laser ceramics is a very promising approach in the perspective of accessing alternative gain geometries and enhanced thermo-mechanical properties. $Yb:CaF_2$ ceramics together with our simple and low-cost synthesis process can open a new route for high power diode-pumped solid-state lasers. Lastly, our newly-developed, energy and cost effective synthesis process of ceramics offers a greener/more ecological approach to laser technology development as compared to single crystal growth.

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REFERENCES

1. F. Druon, F. Balembois, P. Georges, C.R. Physique **8,** 153 (2007).
2. F. Druon, F. Balembois, P. Georges, IEEE Photonics Journal **3,** 268 (2011).
3. G. A. Slack, Phys. Rev. **122,** 1451 (1961).
4. F. Druon, S. Ricaud, D.N. Papadopoulos, A. Pellegrina, P. Camy, J.L. Doualan, R. Moncorgé, A. Courjaud, E. Mottay, P. Georges, Opt. Mater. Express **1,** 489 (2011).
5. V. Petit, P. Camy, J-L. Doualan, X. Portier, R. Moncorgé, Phys. Rev. B. **78,** 085131-1 (2008).
6. P. J. Bendall, C. R. A. Catlow, J. Corish, P. W. M. Jacobs, J. Solid State Chem. **51**, 159 (1984).
7. A. Lyberis, G. Patriarche, P. Gredin, D. Vivien M. Mortier, J. Eur. Ceram. Soc. **31,** 1619 (2011).
8. S. A. Payne, J. A. Caird, L. L. Chase, L. K. Smith, N. D. Nielsen, W. F. Krupke, J. Opt. Soc. Am. B **8,** 726 (1991).
9. J.L. Doualan, P. Camy, R. Moncorgé, E. Daran, M. Couchaud, B. Ferrand, J. Fluor. Chem **128,** 459 (2007).
10. M. Siebold, S. Bock, U. Schramm, B. Xu, J. L. Doualan, P. Camy, R. Moncorgé, Appl. Phys. B **97,** 327 (2009).
11. R. Moncorgé, P. Camy, J. L. Doualan, A. Braud, J. Margerie, L. P. Ramirez, A. Jullien, S. Ricaud, F. Druon, D. N. Papadopoulos, P. Georges, Journal of Luminescence **133,** 276 (2013).
12. J. Boudeile, J. Didierjean, P. Camy, J. L. Doualan, A. Benayad, V. Menard, R. Moncorge, F. Druon, F. Balembois, P. Georges, *Optics Express* **16,** 10098 (2008).
13. F. Friebel, A. Pellegrina, D. N. Papadopoulos, P. Camy, J.L. Doualan, R. Moncorgé, P. Georges, F. Druon, Appl. Phys. B, DOI 10.1007/s00340-014-5872-4, (2014).
14. M. Siebold, M. Hornung, R. Boedefeld, S. Podleska, S. Klingebiel, C. Wandt, F. Krausz, S. Karsch, R. Uecker, A. Jochmann, J. Hein, M. C. Kaluza, Optics Letters **33,** 2770 (2008).
15. G. Machinet, P. Sevillano, F. Guichard, R. Dubrasquet, P. Camy, J.-L. Doualan, R. Moncorgé, P. Georges, F. Druon, D. Descamps, E. Cormier, *Optics Letters* **38,** 4008 (2013).
16. K. S. Wentsch, B. Weichelt, S. Günster, F. Druon, P. Georges, M. A. Ahmed, T. Graf, Opt. Express **22**, 1524 (2014).
17. A. Ikesue, Y. L. Aung, J. Am. Ceram. Soc. **89,** 1936 (2006).
18. A. Lyberis, A. J. Stevenson, A. Suganuma, S. Ricaud, F. Druon, F. Herbst, D. Vivien, P. Gredin, M. Mortier, Opt. Mater. **34,** 965 (2012).
19. M.Sh. Akchurin, T.T. Basiev, A.A. Demidenko, M.E. Doroshenko, P.P. Fedorov, E.A. Garibin, P.E. Gusev, S.V. Kuznetsov, M.A. Krutov, I.A. Mironov, V.V. Osiko, P.A. Popov, Opt. Mater. **35,** 444 (2013).
20. A. Shirakawa, H. Nakao, Y. Higashi, K. Ueda, Y. Ezura, H. Ishizawa, In : Post-deadline proceedings for ASSL - Advanced Solid-State Lasers 2013, Paris, France, 2013. DOI: 10.1364/ASSL.2013.JTh5A.7
21. P. Aubry, A. Bensalah, P. Gredin, G. Patriarche, D. Vivien, M. Mortier, Opt. Mater. **31,** 750 (2009).
22. European Patent Application N° 14.305.495.5 (filling date 04 April 2014).
23. A. Lucca, M. Jacquemet, F. Druon, F. Balembois, P. Georges, P. Camy, J. L. Doualan, R. Moncorgé, Optics Letters **29,** 1879 (2004).

REFERENCES (Full titles and pages)

1. F. Druon, F. Balembois, P. Georges, “New laser crystals for the generation of ultra-short pulses“, Article invité, Compte Rendu de l’Académie des Sciences, Recent advances in crystal optics, C.R. Physique **8,** 153-164 (2007).
2. F. Druon, F. Balembois, and P. Georges, "New Materials for Short-Pulse Amplifiers," IEEE Photonics Journal **3,** 268–273 (2011).
3. G. A. Slack, “Thermal conductivity of CaF2, MnF2, CoF2, and ZnF2 crystals”, Phys. Rev. **122,** 1451-1464 (1961).
4. F. Druon, S. Ricaud, D.N. Papadopoulos, A. Pellegrina, P. Camy, J.L. Doualan, R. Moncorgé, A. Courjaud, E. Mottay, P. Georges, “On Yb:CaF_2 and Yb:SrF_2: review of spectroscopic and thermal properties and their impact on femtosecond and high power laser performance [Invited]”, Opt. Mater. Express **1,** 489-502 (2011).
5. V. Petit, P. Camy, J-L. Doualan, X. Portier, R. Moncorgé, “Spectroscopy of Yb3+:CaF2 : From isolated centers to clusters”, Phys. Rev. B. **78,** 085131-1 (2008).
6. P. J. Bendall, C. R. A. Catlow, J. Corish, P. W. M. Jacobs, “Defect Aggregation in Anion-Excess Fluorites. II. Clusters Containing More Than Two Impurities Atoms“, J. Solid State Chem. **51**, 159-169 (1984).
7. A. Lyberis, G. Patriarche, P. Gredin, D. Vivien M. Mortier, “Origin of light scattering in ytterbium doped calcium fluoride transparent ceramic for high power lasers“, J. Eur. Ceram. Soc. **31,** 1619-1630 (2011).
8. S. A. Payne, J. A. Caird, L. L. Chase, L. K. Smith, N. D. Nielsen, W. F. Krupke, “Spectroscopy and gain measurements of Nd3+ in SrF2 and other fluorite-structure hosts”, J. Opt. Soc. Am. B **8,** 726-740 (1991).
9. J.L. Doualan, P. Camy, R. Moncorgé, E. Daran, M. Couchaud, B. Ferrand,” Latest developments of bulk crystals and thin films of rare-earth doped CaF2 for laser applications”, J. Fluor. Chem **128,** 459-464 (2007).
10. M. Siebold, S. Bock, U. Schramm, B. Xu, J. L. Doualan, P. Camy, R. Moncorgé, ”Yb:CaF2—a new old laser crystal”, Appl. Phys. B **97,** 327-338 (2009).
11. R. Moncorgé, P. Camy, J. L. Doualan, A. Braud, J. Margerie, L. P. Ramirez, A. Jullien, S. Ricaud, F. Druon, D. N. Papadopoulos, P. Georges, “Pure and Yb^{3+} doped fluorites (Ca, Sr, Ba)F_2: A renewal for the future high intensity laser chains”, Journal of Luminescence **133,** 276-328 (2013).
12. J. Boudeile, J. Didierjean, P. Camy, J. L. Doualan, A. Benayad, V. Menard, R. Moncorge, F. Druon, F. Balembois, P. Georges, “Thermal behaviour of ytterbium-doped fluorite crystals under high power pumping”, *Optics Express* **16,** 10098-10109 (2008).
13. F. Friebel, A. Pellegrina, D. N. Papadopoulos, P. Camy, J.L. Doualan, R. Moncorgé, P. Georges, F. Druon, “Diode-pumped Yb:CaF2 multipass amplifier producing 50 mJ with dynamic analysis for high repetition rate operation “, Appl. Phys. B, DOI 10.1007/s00340-014-5872-4, (2014).
14. M. Siebold, M. Hornung, R. Boedefeld, S. Podleska, S. Klingebiel, C. Wandt, F. Krausz, S. Karsch, R. Uecker, A. Jochmann, J. Hein, M. C. Kaluza, “Terawatt diode-pumped Yb:CaF2 laser”, Optics Letters **33,** 2770-2772 (2008).
15. G. Machinet, P. Sevillano, F. Guichard, R. Dubrasquet, P. Camy, J.-L. Doualan, R. Moncorgé, P. Georges, F. Druon, D. Descamps, E. Cormier, “High-brightness fiber laser-pumped 68W Kerr-lens mode-locked Yb:CaF2 oscillator”, *Optics Letters* **38,** 4008-4010 (2013).
16. K. S. Wentsch, B. Weichelt, S. Günster, F. Druon, P. Georges, M. A. Ahmed, T. Graf, "Yb:CaF2 thin-disk laser", Opt. Express **22**, 1524-1532 (2014).
17. A. Ikesue, Y. L. Aung, “Synthesis and Performance of Advanced Ceramic Lasers “, J. Am. Ceram. Soc. **89,** 1936-1944 (2006).
18. A. Lyberis, A. J. Stevenson, A. Suganuma, S. Ricaud, F. Druon, F. Herbst, D. Vivien, P. Gredin, M. Mortier, “Effect of Yb^{3+} concentration on optical properties of Yb:CaF2 transparent ceramics”, Opt. Mater. **34,** 965-968 (2012).
19. M.Sh. Akchurin, T.T. Basiev, A.A. Demidenko, M.E. Doroshenko, P.P. Fedorov, E.A. Garibin, P.E. Gusev, S.V. Kuznetsov, M.A. Krutov, I.A. Mironov, V.V. Osiko, P.A. Popov, « CaF2:Yb laser ceramics”, Opt. Mater. **35,** 444-450 (2013).
20. A. Shirakawa, H. Nakao, Y. Higashi, K. Ueda, Y. Ezura, H. Ishizawa, In : Post-deadline proceedings for ASSL - Advanced Solid-State Lasers 2013, Paris, France, 2013. DOI: 10.1364/ASSL.2013.JTh5A.7
21. P. Aubry, A. Bensalah, P. Gredin, G. Patriarche, D. Vivien, M. Mortier, “Synthesis and optical characterizations of Yb-doped CaF2 ceramics”, Opt. Mater. **31,** 750-753 (2009).
22. European Patent Application N° 14.305.495.5 (filling date 04 April 2014)
23. A. Lucca, M. Jacquemet, F. Druon, F. Balembois, P. Georges, P. Camy, J.L. Doualan, R. Moncorgé, “High-power diode-pumped Yb3+:CaF2 femtosecond laser”, Opt. Letters **29,** 2767-2769 (2004).