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Highly efficient mJ level laser amplifiers at 2 microns for frequency comb spectroscopy

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ABSTRACT

Over the last decade, frequency comb spectroscopy have led to significant developments in view of the identification of varied species and of the understanding of the structure of matter.
Highly efficient amplification of frequency comb femtosecond oscillators in the high pulse energies regime should allow future applications using this approach to Lidar-type measurements.
We report on the millijoule level design of femtosecond amplifiers near 2 µm wavelength having a great optical efficiency and compactness in order to be carrier in satellites. In addition to space applications, laser systems at 2 µm become more and more popular because they offer elegant solutions to generate ultra-broad band super-continuum in the mid-infrared and for material processing.
Our study helps to compare the optical performance of Tm:YAG, Tm:YAP and Tm:YLF crystals as active media, for designing ultrashort pulse regenerative amplifiers with a high gain and wall-plug efficiencies up to 10%. We will present our approach to ensure the conservation of the initial phase shift between the envelope and the carrier of pulses during amplification.
We primarily discuss an innovative model which proposes a gradual path towards the optimization of any regenerative amplifier using crystalline thulium-based, end-pumped doped rods. This also involves the analysis of sizing criteria based on the assumption of rod-based active media, including the doping content, the length of the rod and the beam size inside.

Keywords: frequency comb, Lidar, 2 µm, regenerative amplifier, thulium

1. INTRODUCTION

In the field of atmospheric sounding missions, space lidars were limited to one active wavelength (DIAL lidars). This restraints the concentration retrieval to one vertical element, namely the integrated column amount, and one molecule.

On the other hand, passive space instruments as spectrometers or interferometers provide complete spectra of the atmosphere in the UV, visible or infrared wavelengths. They allow the retrieval of temperature or humidity profiles (IASI), several molecule concentration and green house gazes as CO_2 (Microcarb). Nevertheless, thermal infrared wavelengths do not allow accurate measurements of [CO_2] down to the troposphere. As for visible or near infrared wavelengths, passive nadir sounders are limited to day-time measurements. They are also impinged by the aerosol and cloud diffusion of the optical flux that introduce some inaccuracy in the distance crossed through by the solar flux, hence in the concentration calculated from the attenuation measurement.

The concept of frequency comb lidar aims at joining the advantages of both active and passive instruments together. It is basically a lidar, but it features a set of wavelengths. Doing so allows to withdrawn the diffusion bias (the "lidar advantage") while providing a complete spectrum of the measured flux, from which several species can be retrieved. Complete frequency combs are a thrilling perspective for lidars, but their fitting to a space mission is beyond the state of the art of efficient short (fs) pulse optical amplifiers.

There are several different existing concepts for the generation of intense ultrashort laser pulses in the 2 µm wavelength region. A very prominent technique is optical parametric chirped-pulse amplification (OPCPA), which allows for wide wavelength tunability, short pulse duration, multi-mJ pulse energies, and GW-level peak powers [1]. However, this system is bulky and prone to low wall-plug efficiency, thus limiting its use for space applications.

Thulium (Tm) doped fiber laser systems are another attractive alternative. Indeed, Tm-doped silica fibers possess a very broad amplification band spanning from 1800 to 2100 nm. These fibers can be pumped efficiently by commercially available high power diode lasers emitting around 790 nm, and allow for slope efficiencies higher than 70% by exploiting cross-relaxation processes. The advantageous properties of fiber lasers allow for their average power scaling while maintaining an excellent beam quality. The amplification to high peak powers is challenging due to the high intensities found in the fiber core that, together with the long interaction length, promote the onset of limiting nonlinear effects. A pulse-peak of nearly 2 GW with an energy around 570 µJ has been reported in 2016 with a Tm-based fiber chirped pulse amplification (CPA) system [2]. Although having numerous advantages, the fibered architectures do not currently make it possible to easily obtain an energy higher than 1 mJ. In spite of all the progress, the ultimate performance of a single fibered amplifier might still not be sufficient to reach the desired laser parameters.

An idea to overcome this limit is to use several stages of fiber amplification. One example is the coherent combination of spatially separated amplifiers. In this concept, pulses emitted from the oscillator are split into different channels, they are amplified independently and then recombined. With this approach, Klenke *et al.* have amplified pulses at 1030 nm up to 5.7 mJ with four channels in 2014 [3]. In 2015, the same team has transposed this approach to 2 µm. The coherent combination of two Tm-doped fibered amplifiers allowed for an energy around 22 µJ and a peak power up to 25 MW with excellent pulse quality [4].
Energies higher than 1 mJ will be probably obtained in the coming years and this approach could meet the required parameters. However this amplifier architecture is very complex due to the need of heavy electronics especially if the number of combined beams are higher than two and so it is difficult to develop for a space application.

An alternative approach is to use solid-state amplifiers based on thulium or holmium (Ho) active ions. An interesting architecture consists to insert the doped-crystal in a regenerative amplifier (RA), which can boost nJ-pulses emitted by an ultrafast oscillator directly up to the mJ level. This was demonstrated for the first time at 2.1 µm with a holmium RA, producing pulse energies up to 990 µJ at 530 fs pulse durations [5]. This short pulse duration was achieved by spectral filtering of a broadband OPA seed inside the grating stretcher to compensate for the strong spectral gain shaping inside the RA ring cavity. This system has the drawback of a bulky, cost intensive and inefficient nonlinear parametric seed source, producing the required pulses at 2.1 µm. More compact Tm-Ho-doped ultrashort pulse fiber lasers with a narrower seed spectrum can be used, then a minimum pulse duration of 1 ps could be generated that is fairly enough for our targeted application.

Due to their spectroscopy, the Ho-doped material is typically pumped by a high power Tm:fiber laser, itself pumped by a high-power diode at 793 nm, because high brightness diode around 1.9 µm do not exist. This succession of pump-systems leads to a very low wall-plug efficiency for a holmium-doped RA. This is a limiting factor for space applications! A thulium-doped RA directly diode-pumped at 793 nm then appears as the most appropriate solution to achieve the required parameters. To the best of our knowledge, there is only one paper to Wienke *et al.* reporting a 380 fs Tm:YAP RA with pulse energy around 700 µJ at 1 kHz. Nevertheless this value seems to be limited by the damage threshold of the crystal [6]. As thulium-based RA architectures have not been deeply studied, we developed an innovative model which proposes a gradual path towards the ultimate optimization of any regenerative amplifier using various crystalline thulium-doped rods and to choose the most appropriate host.

The definition of the amplifier is the most difficult point to deal with, both in terms of basic technological choice and architecture. After introducing our innovative numerical model of thulium-doped rod amplifiers, we will discuss interesting results for Tm:YLF and Tm:YAG RA. The problem of controlling the relative phase of the carrier inside the pulses necessary for frequency comb spectroscopy is, initially, dissociated from the concept of amplification. Finally, the overall system concept will be presented.

2. PHYSICAL AND NUMERICAL MODELS OF THULIUM-DOPED ROD AMPLIFIERS

The standard energy diagram (Fig. 1) describing the fundamental processes of energy-transfer between neighboring Tm:ions is presented in Fig. 1. Please note that for the sake of simplification, the sub-structures of so-called Stark levels are not considered. The energy diagram then remains based on the coupling of elementary transitions between a unique set of four energy levels denoted {E1, E2, E3, E4}. They are defined as a fractional content of the total doping density of Tm^{3+} ions, named N_{tot}.
Considering direct diode-pumping around 790 nm, the transition is initiated from the fundamental level E1, towards the upper level E4. The lasing transition originates from the upper laser level E2 and must be related to a given probability of occurrence, to be determined by a very specific coefficient f_2.

The associated densities of population, respectively {N1, N2, N3, N4}, are determined by the kinetics of ionic interactions. In the same way, the actual probability of radiative decay towards E1 for lasing must be specified thanks to a second coefficient f_1. These two coefficients denote the so called "Boltzmann occupation factors". Their values are specified versus the material involved, assuming that the temperature of the medium remains near T=300-350 K. The actual balance between {N1, N2, N3, N4} is governed by the coupling of two kinds of resonant transitions: energy-transfers form cross-relaxation (mean rate p_{22}, m^3/s) from E2, and down conversion (mean rate p_{41}, m^3/s) from E4 to E1. The lifetimes of the three upper levels also participate in the definition of the equilibrium state for continuous-wave operation. This concerns t_2 for radiative decays in the form of fluorescence, but also t_3 and t_4 for the pumping and intermediate-trapping bands with non-radiative decays.

The complete resolution of the rate equations has been discussed in our latest paper [7]. Thanks to the resolution of these coupled equations, we can determine the efficiency of a single pass in a Tm:doped amplifier. Then, we developed an innovative model based on a gradual path towards the ultimate optimization of any regenerative amplifier using crystalline thulium-doped rods.

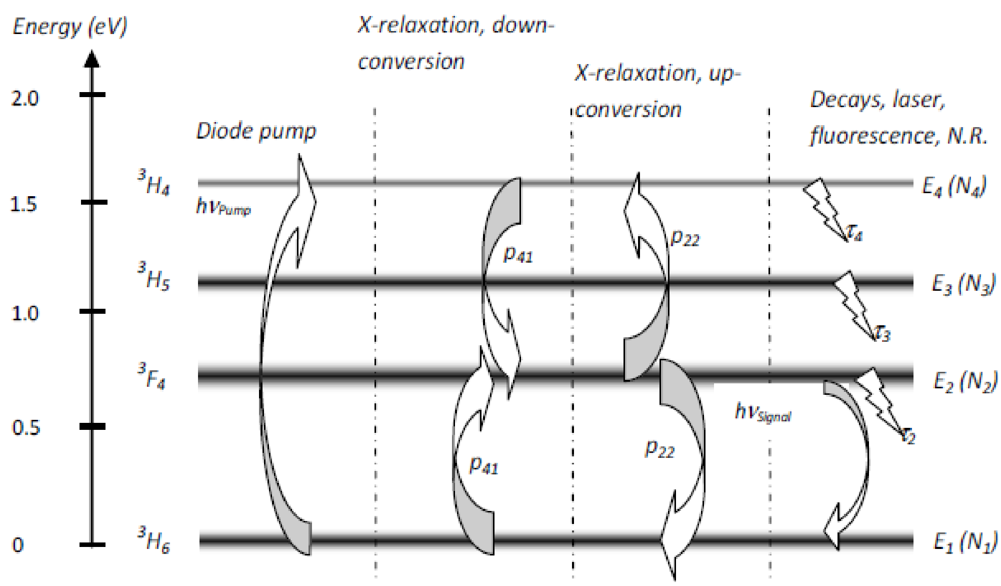

Figure 1 : Energy diagram for Tm^{3+}-Tm^{3+} interactions [7].

Our computational strategy is the following:

First, we define a set of three operating vectors figuring the input data, the sizing constraints and the final results for optimization, as follow:

- V_D = {spectroscopic material data, P_{Pump}, k_{opt}, Γ},

where P_{Pump}, k_{opt} and Γ represent the pump power, the optical losses and the overlap between the pump and the mode of the cavity.

- V_C = {$I_{Pump\ max}$, $L_{rod\ max}$, $N_{tot\ max}$, acceptable specifications of the beam},

where $I_{Pump\ max}$, $L_{rod\ max}$ and $N_{tot\ max}$ are respectively the maximum pump intensity, the maximum length of the crystal and the maximum percentage of Tm-ions.

- V_R = {P_{out}, Φ_{Beam}, L_{rod}, N_{tot}},

where P_{out} is the output power, Φ_{Beam} is the beam diameter, L_{rod} is the rod length and N_{tot} is the Tm-ions rate.

Then, we compute V_R thanks to scanning the coordinates of V_D throughout the affordable domains specified in V_C.

Finally, we determine the location of the ranges of peak values for the optical efficiency $\eta_{O/O}$, to identify the suitable coordinates for V_D in view of optimization.

Our optimization process is based on a series of embedded scanning loops in order to be implemented with appropriate routing conditions. Switching from a given loop to the other occurs as often as needed, for paying a particular attention to the criteria $L_{rod} = L_{opt}$. L_{rod} can be updated at the end of any step along the scan of a given regenerative amplifier configuration.

We have considered YLF and YAG, the two most commonly used materials for which physical properties are perfectly known (see Table 1). The YAP-host used by Wienke *et al.* in [6] is also interesting due to its great spectral gain bandwidth but difficult to model because of the lack of literature for certain spectroscopic parameters.

Table 1. Physical parameters for Tm:YAG and Tm:YLF crystals.

Parameter	Not	Unit	YAG	YLF
Pump absorption cross-section	σ_{aP}	m^2	$7\ 10^{-25}$, λ_S=785nm	$6\ 10^{-25}$, λ_S=792nm
Emission cross-section	σ_{eL}	m^2	$2.2\ 10^{-25}$, λ_S=2010nm	$3.9\ 10^{-25}$, λ_S=1940nm
Re-absorption cross-section	σ_{aL}	m^2	$1\ 10^{-26}$, λ_S=2010nm	$1\ 10^{-25}$, λ_S=1940nm
Lifetime @ N_2	τ_2	s	$12.3\ 10^{-3}$	$15\ 10^{-3}$
Lifetime @ N_3	τ_3	s	$7\ 10^{-6}$	$1\ 10^{-6}$
Lifetime @ N_4	τ_4	s	$1.42\ 10^{-5}$	$2\ 10^{-3}$
Up-conversion cross-relaxation	p_{22}	m^3/s	$2.4\text{-}3.2\ 10^{-24}$	$3.48\ 10^{-25}$
Down-conv. cross-relaxation	p_{41}	m^3/s	$3\text{-}3.5\ 10^{-23}$	$3.13\ 10^{-24}$
Boltzmann occup. factor @ N_1	f_1		0.0013-0.018	0.0273
Boltzmann occup. factor @ N_2	f_2		0.39-0.46	0.2817

3. DISCUSSION OF NUMERICAL RESULTS

Numerical results are provided in the form of 2D-maps to allow appropriate interpretation at first reading and evidence generic trends.Three maps are presented in each case as a function of Φ_{beam} along the y-axis and N_{tot} along the x-axis. These maps represent the evolution of:

- P_{out} for an incident power set at P_{in} = 1 µW (value typically issued from an oscillator, 1 nJ at a repetition rate equal to 1 kHz) and P_{Pump} = 30W (see Fig. 2/3 a)),
- L_{rod} (see Fig. 2/3 b)
- the number of passages required in the crystal for an extraction when the gain is saturated, k_{opt} representing the intra-cavity losses chosen around 10% (see Fig. 2/3 c)).

Figure 2 shows the optimization results for the YLF crystal. We observe that:

- P_{out} can reach 9 W corresponding to an energy around 9 mJ at 1 kHz. The optical-optical efficiency $\eta_{O/O}$ approaches 30%,
- the pump intensity defined as $I_{pump} = P_{pump} / S$ with $S = \pi \Phi^2_{beam} / 4$ should be in the range 4-8 kW/cm²,
- the doping should be around N_{tot} = 5-7.10^{26} m^{-3}, corresponding to approximately 3-5% (values consistent with the bibliographic data),
- the expected number of passages in the cavity varies from 10 to 20, this value is consistent with the data demonstrated with the YAP crystal.

These results also confirm the need to use a high brightness pump diode at 793 nm to deposit the 30 W over a total length likely to reach L_{rod} = 80 mm and in a diameter not exceeding Φ_{beam} = 1-1.5 mm. This may also mean that a double-pass pumping configuration have to be considered.

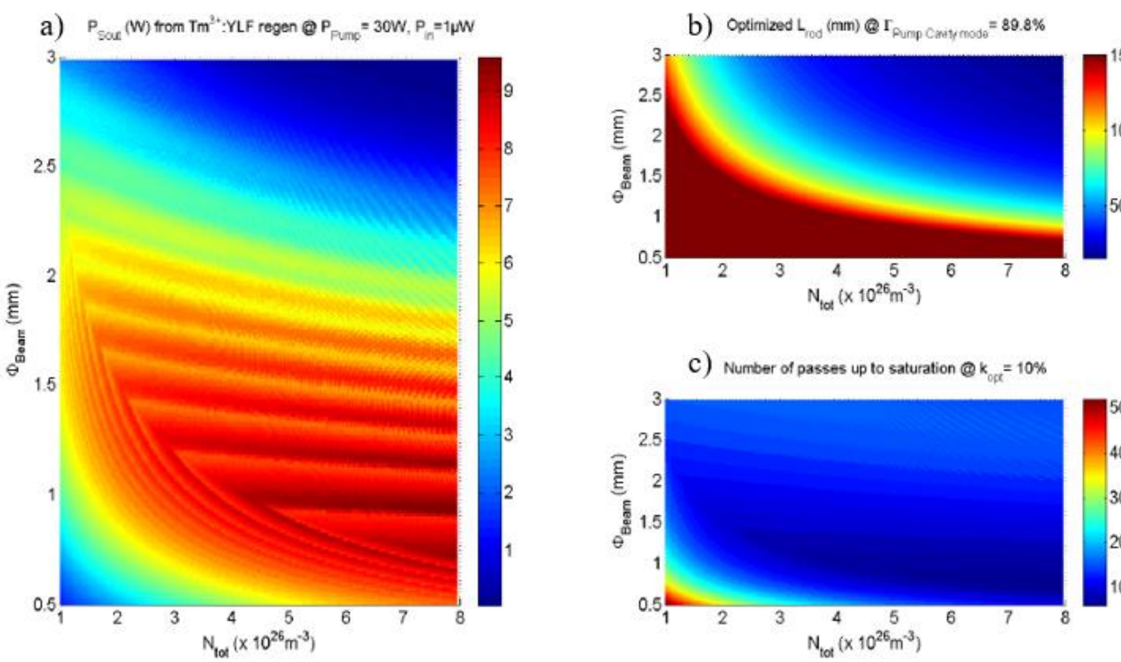

Figure 2: Optimization of the performances of a Tm:YLF regenerative amplifier according to Φ_{beam} (mm) and N_{tot} (m^{-3}) for P_{pump}=30W at 793 nm. On a color scale: evolution of P_{out} (W) a), of the length of the crystal (mm) b), and of the number of passages c). The recovery efficiency between the pump beam and the cavity mode is fixed to Γ = 90%, and the losses are k_{opt}=10%.

Figure 3 shows the same optimization results for the YAG crystal. In this case, we observe that:

- P_{out} can reach 7 W and thus the optical-optical efficiency $\eta_{O/O}$ approaches 23%,
- the pump intensity I_{pump} should be in the range 10-15 kW/cm²,
- the doping should be in the range N_{tot} = 5-7.10^{26} m^{-3}, corresponding to approximately 3-5% (values consistent with the bibliographic data),
- the expected number of passages in the cavity varies also from 10 to 20.

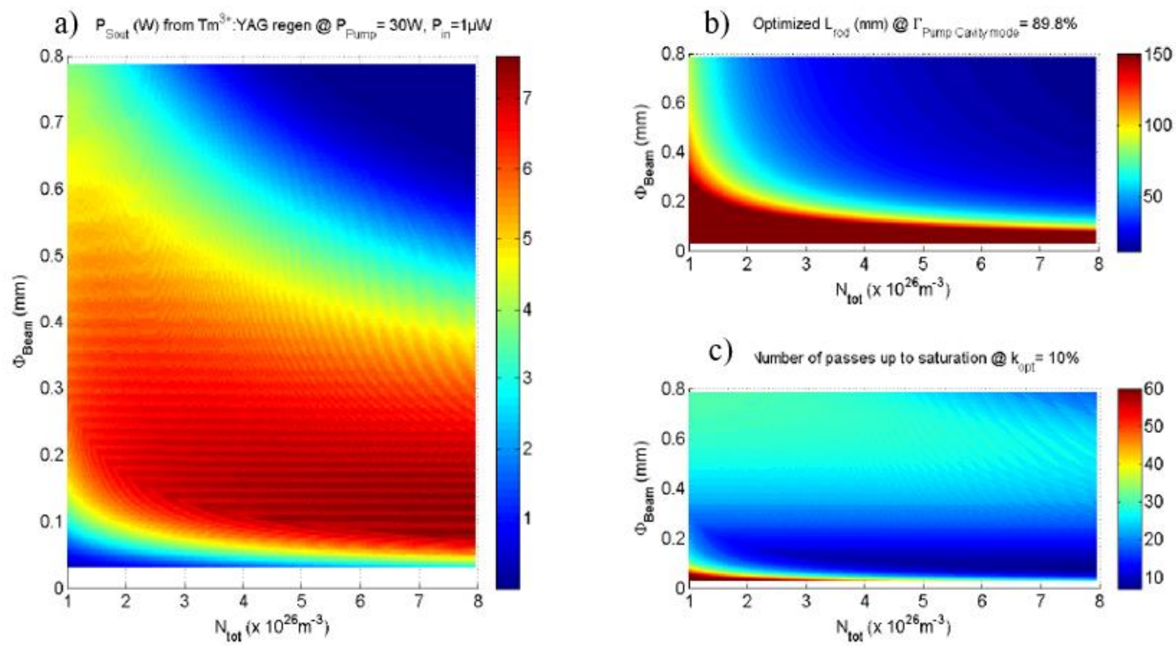

Figure 3: Optimization of the performances of a Tm:YAG regenerative amplifier according to Φ_{beam} (mm) and N_{tot} (m^{-3}) for P_{pump}=30W at 785 nm. On a color scale: evolution of P_{out} (W) a), of the length of the crystal (mm) b), and of the number of passages c). The recovery efficiency between the pump beam and the cavity mode is fixed to Γ = 90%, and the losses are k_{opt}=10%.

These results indicate that the use of a YAG crystal in a regenerative amplifier is possible but more delicate than the YLF. Indeed, for a crystal of similar maximum length, the necessary I_{pump} is three time higher for YAG than for the YFL. This observation is directly related to the spectroscopic of the two materials. This is an important constraint on the amount of heat deposited in the material and therefore the thermal focal length. The thermo-optical and thermo-mechanical gradients defining this thermal lens will thus be more important for the YAG than for the YLF and could strongly degrade the spatial quality of the output beam.

To summarize, the YLF and in the background the YAP used in [6] appear as the two possible candidates to achieve the required performances. The YAP can amplify ultra-short pulses (<400 fs) but the maximum accessible energy seems to be limited to 1 mJ [6]. Based on our simulations, the YLF should allow to generate multi-mJ energy with a good wall-plug efficiency required for spatial applications, but to amplify pulses of less than one picosecond duration it will be necessary to pre-compensate the spectral gain narrowing using, for example, a Lyot filter [8].

4. OVERALL SYSTEM CONCEPT

The proposed principle of amplification of ultra-short pulses (see Figure 4) is based on the classical Chirped Pulses Amplification (CPA technique). We start with a frequency comb femtosecond oscillator delivering sub-200 fs pulses with an energy around 1 nJ in the 2 µm spectral range. These pulses are then stretched up to 100 ps and the repetition rate is reduced down to few kHz with a pulse-picker. The stretched pulses are then injected into the RA cavity with a Tm:YLF or Tm:YAP crystal. A transverse rubidium titanyl phosphate (RTP) pockels cell with a quarter-wave voltage around 1kV, designed to space applications, is used as an optical switch. The pump-diode is a multi-mode fiber-coupled laser diode delivering 30 W at 793 nm with a wall-plug efficiency around 45%. We designed the RA to obtain amplified pulses with energy on the order of few mJ, corresponding to a net-gain around 10^6. Pulses are then compressed to sub-ps duration with a free-space compressor having an efficiency close to 85%. The wall-plug efficiency of this CPA is an important value for space applications. It is mainly determined by three components, more precisely the wall-plug efficiency of the pump diode, $\eta_{O/O}$ and the efficiency of the compressor, and should be greater than 10%. Despite of the estimated length of the cavity is around 2m, the expected maximum size will be 1.5x1.5x0.6 m^3 thanks to folding mirrors, and so the size is consistent with a satellite carrier.

For Lidar-type measurements planned, it is important to keep the carrier-envelope-phase (CEP) shift less than 600 mrad. Nevertheless, in the CPA architecture, many processes are non-negligible sources of phase shift as mechanical vibrations or material temperature vibration [9-10]. To limit these effects, we will implement a well-known technique that consists in measuring the phase shift with a f-to-2f interferometer [11] and set up a feedback loop on the system [12]. An adjustment parameter conventionally used in such a servo loop is the pump power delivered to the oscillator [13]. Typically, the signal emitted by the interferometer controls an acousto-optic modulator (AOM) which makes it possible to finely adjust the pump power and thus the phase shift related to the non-linear effects in the crystal in the oscillator.

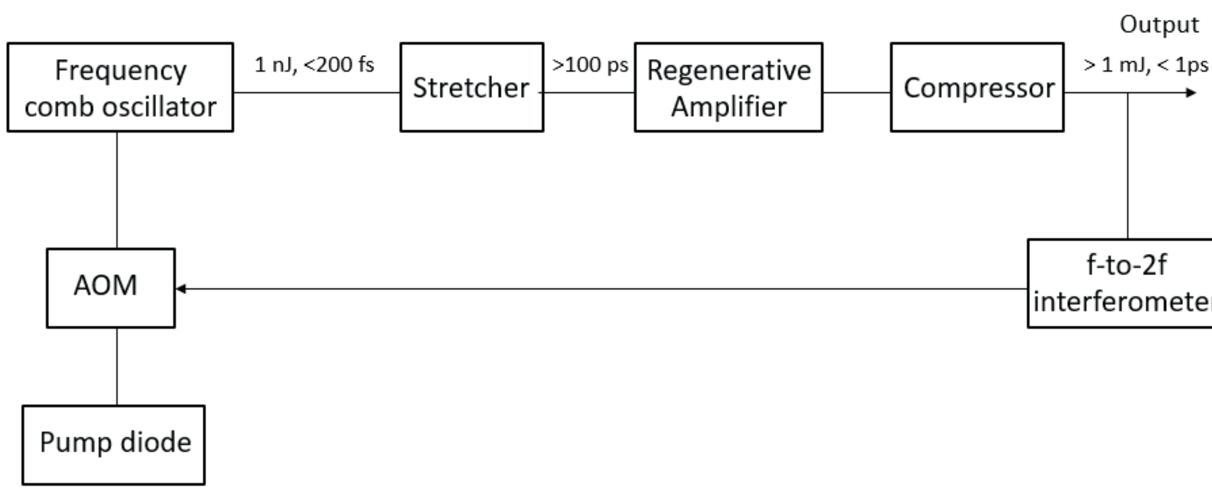

Figure 4: Block diagram of sub-ps scale, multi-mJ, kHz-rate, 2 µm Thulium doped laser system.

5. CONCLUSION

Highly efficient amplification of frequency comb femtosecond oscillators in the high-pulse energies regime should allow future applications to Lidar-type measurements in space. The desired performance for the amplifier have not yet been demonstrated in the literature, particularly in terms of wall-plug efficiency. We have proposed an innovative approach to design this amplifier based on a Thulium-doped regenerative amplifier. We showed that the YLF, and in the background the YAP, appear as the two possible candidates to achieve the required performances. The YAP can amplify ultra-short pulses (<400 fs) but the maximum accessible energy seems to be limited to 1 mJ. Based on our simulations, we demonstrated that the YLF should achieve multi-mJ energy with a good wall-plug efficiency, but to amplify pulses of less than one picosecond duration with this crystal it will be necessary to pre-compensate the spectral gain narrowing. Independently of the concept of amplification, we also brought a solution to control the relative phase of the carrier in the pulses, necessary for frequency comb spectroscopy. Finally, we discussed the overall system concept. After having demonstrated the feasibility of such an architecture and having obtained the required performances, ALPhANOV and the CNES will work on the integration and spatialization of this system.

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