New materials for laser amplifiers:
Diode-pumped ytterbium amplifiers

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Abstract: The introduction of new laser amplifier materials has been one of the main drivers of this technology since its invention, almost 50 years ago. In the last decade, ytterbium-doped materials have become the focus of particular attention, due to their ability to be diode-pumped. This combination allows a wall-plug efficiency several orders of magnitude greater than other sources such as flashlamps. Diode-pumped ytterbium-based laser systems have thus become the preferred option for the generation of high energy, high repetition rate pulses.

The work done in this Thesis is part of the research done in this area at the Laboratory of Intense Laser of the Group of Lasers and Plasmas of IST. In this document we describe the implementation of a first regenerative amplifier capable of producing mJ pulses in near infrared, based on Yb:glass pumped by a diode stack. This amplifier was characterized and its limits of operation were determined.

Simultaneously, a numerical code was developed and tested in order to simulate in a physically realistic fashion any generic amplifier of this kind, which was then applied to four different hosts (glass, CaF$_2$, KYW and YAG). The agreement between the experimental results and the code demonstrates its reliability. This tool was used to study a multi-pass amplifier setup designed to generate pulses in the 100 mJ range and a repetition rate a hundred times greater than the currently available at the LI2, whose preliminary implementation is described.

1. Introduction

Although as early as 1962 certain crystals and glassy materials doped with ytterbium ions (Yb$^{3+}$) were identified as gain media, only three decades later these materials started being introduced in practical laser systems. This was due largely to the beginning of the industrial development of laser diodes [Koechner], in the end of the ‘80s. Before the development of laser diodes, optical pumping of these materials was done by flashlamps. These have a broad emission spectrum, making lasers based on the Yb$^{3+}$ ion very inefficient due to its simple electronic structure, since most of the emitted light is not absorbed. Indeed, when the Yb$^{3+}$ ion is isolated, it has two levels of energy, which are divided into sub-levels when the ion is inserted in a crystal or a glass [Chénais 02, Raybaut 03].

The diodes, in contrast to the lamps, have a smaller emission spectrum, which allows virtually all the emitted radiation to be absorbed by materials with intense absorption peaks at certain wavelengths. This feature, added to the fact that diodes can emit high power radiation with electric/optical efficiency up to 85% (higher than any other pumping devices), and have a long life, makes these pumping sources more appropriate to be used with ytterbium-based lasers.

The "rediscovery" of ytterbium-doped materials and their growing application in laser devices was not by chance though, since they also exhibit significant advantages when compared to other gain media – in particular, neodymium-doped hosts [Chénais 02, Raybaut 03]:

- As with other lanthanides, the simple electronic structure of the ytterbium ion prevents parasitic effects involving other levels of energy.
- Yb-doped materials have a large storage capacity and long fluorescence times, compared to other materials, therefore allowing long pump pulses.
- They allow higher repetition rates (e.g. compared to Nd:glass), since they have lower thermal losses due to their small quantum defect (the difference between the energies of a pumping photon and lasing photon).
- Large saturation fluence, meaning that for generating a given output energy a smaller area is required than with competing materials. This also makes cooling and refrigeration easier, allowing higher repetition rates. [Nees]

The main motivation for this work is the development of diode-pumped Yb amplifier technology at the Laboratory for Intense Lasers, which is a major theme of IST’s participation in large scale european research projects (such as LASERLAB-EUROPE). Another motivation is the need for an alternative, reliable mJ-level laser source for seeding the large aperture Nd:glass amplifiers, competitive with the...
current Ti:sapphire regenerative amplifier. And finally, the preliminary study and subsequent assembly of a diode-pumped, 100 mJ-level amplifier at high repetition rate (~Hz), available for either laser-plasma experiments or as a pumping source for an optical parametric amplifier.

The objective of this work is therefore to install and characterise a diode-pumped, ytterbium-based regenerative amplifier, capable of consistently delivering pulses with energies around the milijoules level.

It was also necessary to develop a numerical code capable of realistically simulating the performance of any Yb amplifiers, and to be robust enough to allow the simulation of regenerative and multipass amplifiers. The code was specifically meant to be applied to a group of four ytterbium-doped materials (Yb:glass, Yb:CaF$_2$, Yb:KYW and Yb:YAG), in order to determine which one is the most suited for amplification at 1053 nm (the operating wavelength of L2’s Nd:glass amplifiers), for a regenerative and a multipass configuration.

Another objective was the experimental characterization of the single pass gain as a function of the seed diameter for a series of candidate materials for the 100 mJ multipass amplifier (three crystals of Yb:CaF$_2$ of different sizes and one of Yb:glass).

2. Experimental setup

In the Yb:glass regenerative amplifier (Figure 1), as the name suggests, the material used is glass (fluoride phosphate) doped with ytterbium. The medium has a length of 6.5 ± 0.05 mm and a Yb$^{3+}$ density of 6×10$^{20}$ per cm$^3$. The surfaces have an anti-reflective coating. The cavity of the amplifier (between mirrors M1 and M3) includes a polarizer, a Pockels cell, a quarter-wave-plate ($\lambda/4$), providing the injection and extraction of pulses from the cavity in the same direction. This assembly of the cavity also requires the installation of another set of elements to its input/output in order to carry out the extraction of the pulses. The extraction of the pulses can be made through a Pockels cell and a polarizer.

Pumping of the ytterbium amplifier is made by a stack of 25 diode bars, model JOLD-4000-QAXH-25A of JENOPTIK Laserdiode GmbH, which has an electro-optical efficiency of 45.6% at maximum power. This stack, at a temperature of 25 °C, emits infrared radiation at a central wavelength of 931.7 nm and a spectral bandwidth of 1.3 nm. The power depends directly on the emission current that is applied to the stack and may vary from about 120 W to 4000 W, with the current varying from 20 A to 199 A. The dependence of power with current is almost linear, being given approximately by the ratio of 21.73 W/A. The stack is also equipped with a set of micro-lenses, which significantly reduce the
divergence of the light emitted by it. A maximum repetition rate of 1 Hz can be achieved, due to the
inability to make more than one "shot" per second of the device that controls the power and time to
pump the diodes (Laser Diode Driver’s Töpfer & Hein). Beyond this, the Laser Diode Driver imposes a
maximum time of 3 ms of pumping.

3. Numerical Code
Given the great diversity of Yb-doped materials, with several features and a wide variety of
parameters relevant to their operation in a laser amplifier, it became necessary to develop a numerical
code capable of simulating the behaviour of a generic Yb-based amplifier. Thus, without great cost and
little time, we are able to model and analyze the performance of several Yb-doped gain media. Similarly,
the numerical code will allow a study of what materials are best suited to our objectives.
Nowadays the development of simulation codes for pump and amplifications processes is common. There
are several numerical codes that allow determining some amplification parameters or properties like gain
narrowing and gain shifting in a broad-band Yb$^{3+}$-doped material during regenerative amplification
processes [Raybaut 05], thermal effects on diode-pumped materials (temperature map on the material
surface) [Altmann, Didierjean] and the influence of Yb$^{3+}$ fluorescence lifetime on the laser threshold,
Yb$^{3+}$ doping on the laser threshold and on the laser efficiency for diode-pumped Yb-doped planar
waveguide laser [Petit].

The code was built with the objective of simulating any type of Yb amplifier (regenerative and
multipass). This code allows determining the single pass gain, the maximum extracted energy, the number
of passes for maximum energy, and also the signal spectrum at any pass.

The code was designed in MatLab, and all its operation is based on discrete variables and
numerical calculations. The simulation of the processes of pump and amplification is based on the
numerical calculation of differential system of equations [Chénais 02]:

\[
\frac{dN_{\text{ex}}(z,t)}{dt} = \left[\sigma_e(\lambda)N_{\text{ground}}(z,t) - \sigma_a(\lambda)N_{\text{ex}}(z,t)\right] \frac{\lambda}{hc} I(z,t) - \frac{N_{\text{ex}}(z,t)}{\tau_{\text{fluor}}} \\
\frac{dI(z,t)}{dz} = \left[-\sigma_a(\lambda)N_{\text{ground}}(z,t) + \sigma_e(\lambda)N_{\text{ex}}(z,t)\right] I(z,t)
\]

(S1.1)

where $I$ is pump or pulse intensities for different a wavelength $\lambda$, at a $z$ crystal length and at time $t$, $c$ is
light velocity, $h$ is Planck constant, $\sigma_a$ is absorption cross-section $\sigma_e$ is emission cross-section, $N_{\text{ex}}$, $N_{\text{ground}}$
are the densities of Yb ions on excited and ground levels respectively.
The operating parameters can be essentially summarized in four groups:
- the type of crystal (absorption cross-section, emission cross-section, the ion concentration, length, fluorescence time)
- the pumping conditions (power, duration, wavelength, diameter)
- the input signal characteristics (bandwidth, wavelength, diameter, energy, duration)
- the cavity properties (losses, spectral performance).

The simulation program is basically composed of three parts: the first is defined the absorption and emission cross-sections of the material to be studied; the second part is the pump routine; and, finally, the last part of the code is the amplification routine. Figure 2 shows a schematic of the relative interconnection of these routines and parameters.

The absorption and emission cross-sections play a central role in the pump and amplification simulations. These are present in all differential equations needed for those two processes. The introduction of cross-sections has to account for their dependence on the wavelength. Only in this fashion one can obtain a more realistic simulation of the amplification, by being able to observe the evolution of pulse spectra after each pass through the active medium, and thus numerically study the effects of gain narrowing and gain shifting.

4. Yb:glass amplifier characterization

a) Output energy vs. repetition rate

One of the first tests done with the Yb:glass regenerative amplifier is related to its performance at different repetition rates. During these tests the output energy of twenty pulses was measured. The amplifier made $184 \pm 2$ passes through the active medium. The first ten pulses were obtained at the repetition rate of 1 Hz and the remaining ten pulses at the rate of 0.5 Hz. The pump power in all these measurements was kept constant at $1624 \pm 37$ W.

Figure 3 shows the pulse energy for the both measurements. From this, we see that at both repetition rates it is possible to amplify pulses up to milijoule energies in a stable manner. The mean energy acquired at 1 Hz was $1.54 \pm 0.17$ mJ (11.1%), while at 0.5 Hz it was $2.59 \pm 0.25$ mJ (9.7%). The increase in average energy of 69% indicates that the single pass gain is higher at the lower repetition rate. Another observation from these results is that the stability at 1 Hz was lower, since the energy spread was relatively larger. The explanation for different single pass gain and for stability is thermal load. At 0.5 Hz the deposition of heat in the glassy material is slower, allowing for more cooling of the medium and consequently a greater homogenization of the temperature. Thus, the expansion created in the material (thermal lens [Chénais 02, Chénais 06, Wemans]), where it is pumped, has a smaller amplitude at 0.5 Hz.

Despite the evidence that it is possible to obtain pulses with energies above 1 mJ at 1 Hz, the following measurement were performed at 0.5 Hz, to ensure a better energy stability.

b) Output energy vs. pump power

The next study consisted in evaluating the evolution of the output pulse energy as a function of pump power. These results were obtained by varying the current applied to the diodes from 75 A to 93 A. The pump time was 3 ms and the number of passes through the gain medium was $184 \pm 2$. The graph in Figure 4 shows the output pulse energy increasing exponentially with pump power. From this, we can conclude that the maximum storage of energy (or saturation) by the Yb:glass was not achieved by the pump powers used, and that we are clearly working in the unsaturated gain regime.
In this experiment it was also observed that for a pump power of 1662.9 ± 37.6 W (corresponding to a 93 A current applied to the diodes) we begin to damage the components in the cavity. However, we were still able to measure energies above 5 mJ.

![Figure 4: Output pulse energy as a function of pump power](image1)

**b) Amplifier tunability – seed spectrum**

In order to study the amplifier tuning capability, two methods were implemented. In the first, the objective was to evaluate the response of the amplifier for seeds with different central wavelengths, \( \lambda_i \), without neglecting the fact that we want mJ-level energies. The stretcher is tuned for hard spectral filtering to vary the \( \lambda_i \) injected and the oscillator was centred at 1053 nm. For this study, the pulses were forced to cross the active medium around 186 ± 2 passes and pump power was equivalent to a 90 A current. The results obtained are presented in Figures 5 and 6. The seed spectrum before to make any adjustment in the stretcher is indicated in red.

![Figure 5: Output energy as a function of seed pulse central wavelength.](image2)

![Figure 6: Tunability for different seeds. Seed spectra (left) and corresponding output pulse spectra (right), matched by colour.](image3)

For all seeds injected during this experimental procedure, pulses with energies of several milijoules were obtained. In general, the output pulse energy decreases with the increase of \( \lambda_i \). This is due mainly to the influence of the cross-sections of emission, \( \sigma_e \), and absorption, \( \sigma_a \), of the gain medium.
These measurements also formed the basis to test the numerical code. We considered three of the seeds injected in the amplifier (corresponding to colours magenta, red and blue in Fig. 6), and then determined numerically the output spectrum. The numerical code was fed with real data of the three seeds (a fit was made to each spectrum), the pump and the gain medium. Also, for each case, we adjusted the number of passes, the spectral filtering at the stretcher and the spectral response of the cavity.

The comparison of experimental and numerical results for the three signals can be observed in Figure 7. In all the graphs presented in this figure the numerical spectra show pronounced cuts at a certain wavelength. These are due to the action of the stretcher as a filter. Experimentally, these cuts also happen in the spectrum. It can be easily detected by the high asymmetry of the spectra after amplification (for spectra resulting from signal $\lambda_i$ equal to 1050.3 nm and 1056.9 nm).

Figure 7: Comparison of numerical (red) and experimental (blue) results. Seed spectra on the left, output pulse spectra on the right. These results are ordered from top to bottom for seed central wavelength at 1050.3 nm, at 1053.7 nm and at 1056.9 nm.

The numerical simulation results presented in Figure 7 are, in general, consistent with the experimental results.

c) Amplifier tunability – cavity tuning

In a second method, we studied the wavelength tuning of the amplifier when seeding it with a constant seed spectrum, but changing the spectral response of the cavity. This was achieved by varying the angle of the intra-cavity polarizer. The results are shown in Figure 8, using a pump power of 1502.4 ± 36.0 W (equivalent to 80 A current), for 260 ± 2 passes through the gain medium and for a seed of $\lambda_i = 1056.3$ nm with a bandwidth $\Delta\lambda_i = 11.1$ nm (the black curve in the figure).

We can see that the tuning of output pulses is limited for a wavelength $\lambda < \lambda_i$. It is very difficult to obtain amplified pulse spectra with $\lambda_o$ higher than the spectrum shown in blue ($\lambda_o = 1051.8$ nm).

From the results, it was discovered that by using either methods for tuning the output pulses on the amplifier it was possible to tune amplifier from 1037 nm to 1052 nm.
Figure 8: Tunable spectral window for the study of tunability for different cavity spectral responses. Seed pulse spectrum is shown in black, other colours correspond to the output pulse spectra.

5. Numerical code results - tunability for different materials and amplification at 1053 nm

In this section we present some results of the simulation for the tunability of different Yb materials (Yb:glass, Yb:CaF
_2_, Yb:KYW and Yb:YAG) and amplification at 1053 nm.

All simulations were performed for the same conditions of pump and amplification, so that we can compare the behaviour and performance of the amplifier for the several materials. These tests were carried out in order to find the material which is more appropriate for hybrid ytterbium-neodymium based amplification at the L₁. 

In all cases, it was considered that the pump beam has a power of 2000 W, a diameter of 2 mm and a duration of 3 ms; the material has a length of 7 mm and a density of 6 x 10^{20} ions per cm^3; the injected seed has an energy of 10 pJ, a diameter of 1.8 mm and a bandwidth of 12 nm; and the spectral bandwidth of the cavity was 20 nm. The results for the tunability for the Yb:glass and the Yb:KYW and amplification at 1053 nm are given in Figure 9 and Tables 1 and 2.

Figure 9: Numerical results for amplifier tunability with different gain media. Output pulse spectra for Yb:glass (top) and Yb:KYW (bottom).
Table 1: Numerical results for the study of tunability and amplification at 1053 nm for Yb:glass. \( \lambda_i \) is the seed central wavelength; \( \lambda_c \) is the centre of cavity; the \( \lambda_o \), \( \Delta\lambda_o \) and \( E_0 \) are the output pulses central wavelength, bandwidth and maximum energy. The first four rows correspond to spectra shown in Figure 9 (top).

<table>
<thead>
<tr>
<th>( \lambda_i ) (nm)</th>
<th>( \lambda_c ) (nm)</th>
<th>( \lambda_o ) (nm)</th>
<th>( \Delta\lambda_o ) (nm)</th>
<th>( E_0 ) (mJ)</th>
</tr>
</thead>
<tbody>
<tr>
<td>1030</td>
<td>1040</td>
<td>1031.9</td>
<td>4.20</td>
<td>3.209</td>
</tr>
<tr>
<td>1040</td>
<td>1045</td>
<td>1037.6</td>
<td>5.31</td>
<td>4.435</td>
</tr>
<tr>
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<td>1050</td>
<td>1045.1</td>
<td>6.27</td>
<td>5.218</td>
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<tr>
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<td>1055</td>
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<tr>
<td>1064</td>
<td>1055</td>
<td>1052.7</td>
<td>8.96</td>
<td>0.002</td>
</tr>
</tbody>
</table>

Table 2: Numerical results for the study of tunability and amplification at 1053 nm for Yb:KYW. \( \lambda_i \) is the seed central wavelength; \( \lambda_c \) is the centre of cavity; the \( \lambda_o \), \( \Delta\lambda_o \) and \( E_0 \) are the output pulses central wavelength, bandwidth and maximum energy. The first four table rows correspond to spectra shown in Figure 9 (bottom).

<table>
<thead>
<tr>
<th>( \lambda_i ) (nm)</th>
<th>( \lambda_c ) (nm)</th>
<th>( \lambda_o ) (nm)</th>
<th>( \Delta\lambda_o ) (nm)</th>
<th>( E_0 ) (mJ)</th>
</tr>
</thead>
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<tr>
<td>1030</td>
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<tr>
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<tr>
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<td>1055</td>
<td>1045.9</td>
<td>3.41</td>
<td>12.729</td>
</tr>
</tbody>
</table>

From the graphs of Figure 9, we can see that both Yb:glass and Yb:KYW show tuning of the amplified pulse spectrum from 1030 nm to 1060 nm.

Looking in closer detail, it is difficult for either of these materials to generate pulses with spectra centred at 1053 nm and with energies of \(~\text{mJ}\). However, we can conclude that the material more suited to operate with Nd:glass is Yb:glass, while Yb:KYW is ideally suited for use in a multipass amplifier, for obtaining energies of \(~\text{100 mJ}\) at a slightly smaller wavelength. This application is investigated in the next section.

6. Code results - amplification to 100 mJ

Given the results described in the previous section, we decided to simulate the operation of a multipass amplifier based on Yb:KYW, with eight passes. For these simulations we considered: the crystal has a density of \( 8 \times 10^{20} \) ions per \( \text{cm}^3 \); the pump has an output of 4000 W, a diameter of 4 mm and a duration of 3 ms; the seed has an energy of 1 mJ, a diameter of 4 mm and a bandwidth of 5 nm; and finally the bandwidth of the cavity is 20 nm. Given the fixed geometry chosen, we investigated the energy of the output pulses as a function of the crystal length and central wavelength of the seed (see Figure 10). The cavity operating wavelength for each seed is equal to the central wavelength of the seed.

From the graph of Figure 10, we can see that it is possible to numerically obtain a 100 mJ-level amplifier at 1030 nm.

![Figure 10: Yb:KYW multipass amplifier simulation. Output pulse energy (J) as a function of crystal length and seed central wavelength.](image)

We can also see that there is an optimum crystal length of 3.2 cm, where the gain of the amplifier is at its maximum. This is because, as the length increases, the number of ytterbium ions increases, causing a greater amount of absorbed pump, and thus benefiting the beginning spontaneous emission.
However, from 3.2 cm onwards the pump is going to become insufficient to maintain a high density of ions in the excited state. This leads to increased seed absorption during its passage by the crystal, and consequently to lower pulse energies at the end of the eighth pass.

7. Single pass gain for multipass amplifier

The actual assembly of the 100 mJ multipass amplifier is outside the scope of this work; however, we were able to measure experimentally the single pass gain for four Yb-doped materials (three CaF$_2$ crystals with 2.1 cm, 1.37 cm and 0.4 cm of length and one Yb:glass crystal with 0.65 cm of length), as a function of the seed diameter. We could then ascertain the best suited material for a multipass amplifier.

For this experiment we used the nJ-level, chirped and stretched pulses coming from the oscillator and stretcher. These pulses were passed through the Yb-doped medium and then directed to a photodiode. The current applied to the diodes during the experiment was 160 A (maximum current supplied by Laser Diode Driver), corresponding to power of 3158.9 ± 46.2 W.

For each material we calculated the single pass gain for seed diameters between 1-4 nm. For each diameter we considered two situations (see Figure 11): absence (red) and presence (blue) of thermal lensing. The results are shown in Figure 11. The blue circles represent single pass gain considering the existence of thermal lens, while the red circles do not include that effect.

As expected, the single pass gain is higher when we do not take into account the effect of thermal lensing. Additionally, for both crystals the single pass gain decreases in general with the increase of seed diameter. This fact is more visible for longer crystals, since the phenomenon of absorption becomes more significant.

8. Conclusions

The main objective of this experimental work – to install and characterise the diode-pumped ytterbium regenerative amplifier, capable of consistently generating pulses with energies around millijoules – was achieved successfully for an Yb:glass medium. With this material it was possible to:
- measure energies >1 mJ consistently, at 1 Hz and at 0.5 Hz repetition rate for a pump power and duration of 1624 ± 37 W and 3 ms, respectively. It was also shown that at a 0.5 Hz rate the amplifier operated in a more stable manner, having also a greater single pass gain;
- determine the evolution of pulse energy as a function of pumping power. It was observed that this evolution was exponential, showing that single pass gain and population inversion increased with the pump power (unsaturated regime).
- determine the performance limits of the installed amplifier. A maximum energy of 5 mJ was generated, and tunability between 1037–1052 nm was demonstrated.

Concerning the numerical objectives:
- we have developed a new numerical code and demonstrated its reliability by benchmarking it against the obtained experimental spectra for the same conditions of amplification;
- we studied the amplifier tunability for different Yb-doped materials towards practical amplification at 1053 nm. These studies confirmed that the best material for a regenerative amplifier in the L$^2$I is the Yb:glass. It was also concluded that Yb:KYW is the most promising material for a possible multipass amplifier.
- the simulation of a Yb:KYW multipass amplifier was carried out. It was shown numerically that it could reach energies higher than 100 mJ, in 8 passes, at 1030 nm.

As a preparation for the multipass amplifier, the single pass gain was also measured experimentally as a function of the seed diameter for two Yb:CaF$_2$ crystals. It was observed that the single pass gains decreases as the seed diameter increases.

In conclusion, this work has been useful in providing valuable information for future technological options related to the development of diode-pumped ytterbium-based amplification at the L$^2$I, thanks to
its comprehensive evaluation of some of the most popular Yb-based amplifier materials near the 1053 nm region. These results are inestimable for eventually coupling a new mJ-level amplifier to the existing high-aperture, high-energy Nd:glass rod amplifiers.

Additionally, the possibility of developing a new, high repetition rate source of 100 mJ pulses was also investigated, with a positive outlook. Such a pulse source will have a significant impact in the research of this laboratory, for either laser-plasma experiments or as an advanced pump source for optical parametric amplification experiments.

I would like to thank João Wemans, whose assistance, availability expertise were essential for the success of this work.

References:


