High pulse energy multiwatt Yb:CaAlGdO₄ and Yb:CaF₂ regenerative amplifiers

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Abstract: We investigated and compared Yb:CaAlGdO₄ and Yb:CaF₂ regenerative amplifiers at repetition rates 5-10 kHz, a frequency range interesting for industrial applications requiring relatively high pulse energy. Both materials allow for pulse energies close to 1 mJ with sub-400-fs pulses. The two laser materials offer comparable performance in the pump power range investigated. The same regenerative amplifiers can be run up to 500 kHz for much faster material processing, with maximum output power of up to 9.4 W.

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1. Introduction

Femtosecond laser pulses with energy at the millijoule level have enabled many scientific and industrial applications over the last two decades [1]. Initially, the high-energy femtosecond technology was confined to the laboratory owing to the complex and expensive Ti:sapphire amplifier setups, requiring high-power green pump lasers to achieve such pulse energies [2]. A major step forward toward practical applications was the development of the first diode-pumped laser systems based on Ytterbium-doped materials. These proved to be very promising and cost-effective for the generation of high energy and ultrashort laser pulses in the 1-µm wavelength range [3,4]. Thanks to the great advances in laser diode technology and the availability of new Yb-doped materials with both good thermo-optical properties and broad gain bandwidth, remarkable achievements in directly diode-pumped femtosecond amplifier systems at 1 µm have been reported in the last few years. Among the most promising laser media, Yb:CaAlGdO₄ (Yb:CALGO) has a very high thermal conductivity allowing high-power diode pumping, as well as the flattest and broadest emission spectrum of all Yb-doped crystals. Recently, it has shown great potential for the amplification of femtosecond laser pulses at room temperature with demonstrated sub-100 fs pulse with energy up to 24 μJ at 50 kHz repetition rate [5]. The highest output power reported to date for a Yb:CALGO based regenerative amplifier is 28 W operating at 500 kHz, with 217-fs pulses [6]. Up to 3 mJ (uncompressed, 2.2-nm FWHM bandwidth) of pulse energy at 1 kHz repetition rate has also been demonstrated in a double crystal setup [7].

Another attractive laser medium is ytterbium-doped alkaline-earth fluoride Yb:CaF₂ [8]. It can be easily grown since fabrication technology for CaF₂ is very well developed for production of large commercial optics. Furthermore, Yb:CaF₂ has a broad emission bandwidth, a much better thermal conductivity than glass and a long fluorescence time of 2.4 ms, which is attractive for very high energy ultrafast amplifiers. Indeed, Yb:CaF₂ has shown great potential for the amplification of femtosecond laser pulses at room temperature with demonstrated pulse energies up to 0.63 mJ at 500 Hz repetition rate and 178 fs pulse duration [9] or up to 16.6 J at much lower repetition rate for extremely high-energy ultrafast laser facilities [10]. Multi-mJ, sub-ps pulses at 1 kHz repetition rate have been reported, but only with cryogenic cooling of the active medium [11].

Despite the fact that Yb:CaF₂ exhibits much poorer thermo-optical parameters than Yb:CALGO (the thermal fracture parameter is ~ 5 times smaller), very high power was demonstrated with special setups such as the thin-disk laser, where as much as 250 W output power was reported [12]. The more relevant spectroscopic and thermal parameters of both materials at room temperature are summarized in Table 1.

Yb-doped tungstates such as Yb:KYW were also successfully employed in regenerative amplifiers, although the emission bandwidth of these crystals is narrower and supports longer pulses: for example, 480-fs long pulses with 1 mJ energy and multi-kHz repetition rate has been reported [13].

In this work, we report the first sub-400 fs Yb:CALGO and Yb:CaF₂ regenerative amplifiers capable of providing pulse energies exceeding 1 mJ at 5-kHz repetition rate, and close to 1 mJ up to 10 kHz, at room temperature.
Table 1. Spectroscopic and Thermal Properties of Yb:CALGO and Yb:CaF$_2$ at Room Temperatures

<table>
<thead>
<tr>
<th>Properties [Unit]</th>
<th>Yb:CALGO [14–16]</th>
<th>Yb:CaF$_2$ [14, 17, 18]</th>
</tr>
</thead>
<tbody>
<tr>
<td>Crystal family, symmetry</td>
<td>Aluminates, tetragonal uniaxial</td>
<td>Alkaline-earth fluorides, isotropic cubic</td>
</tr>
<tr>
<td>Pump wavelength $\lambda_p$ [nm]</td>
<td>979</td>
<td>979.8</td>
</tr>
<tr>
<td>Absorption bandwidth (FWHM) [nm]</td>
<td>&gt;5</td>
<td>&gt;8</td>
</tr>
<tr>
<td>Absorption cross section at $\lambda_p$ [10$^{-20}$cm$^2$]</td>
<td>1 $\parallel$ a, 2.7 $\parallel$ c</td>
<td>0.54</td>
</tr>
<tr>
<td>Emission cross section [10$^{-20}$cm$^2$]</td>
<td>0.75 $\parallel$ a, 0.25 $\parallel$ c @ 1040 nm</td>
<td>0.17 @ 1049 nm</td>
</tr>
<tr>
<td>Emission bandwidth [nm]</td>
<td>~80</td>
<td>~70</td>
</tr>
<tr>
<td>Fluorescence lifetime [μs]</td>
<td>420</td>
<td>2400</td>
</tr>
<tr>
<td>Non-linear index [10$^{-20}$m$^2$W$^{-1}$]</td>
<td>9</td>
<td>2.5</td>
</tr>
<tr>
<td>Thermal conductivity [W m$^{-2}$K$^{-1}$]</td>
<td>6.9 $\parallel$ a, 6.3 $\parallel$ c (2% doped)</td>
<td>6 (2.6% doped)</td>
</tr>
<tr>
<td>Thermo-optical coefficient [10$^{-6}$K$^{-1}$]</td>
<td>13</td>
<td>$-11.3$</td>
</tr>
<tr>
<td>Power resistance parameter [10$^8$ W$^{-2}$ m$^{-2}$ K$^{-1}$]</td>
<td>0.35 (2% doped)</td>
<td>0.07 (5% doped)</td>
</tr>
</tbody>
</table>

2. Setup and experimental results with Yb:CALGO

The experimental setup is shown in Fig. 1. It consists of a diode-pumped Yb:CALGO oscillator, a Yb:CALGO regenerative amplifier, and a stretcher-compressor assembly based on transmission gratings.

![Experimental setup](image)

The available Yb:CALGO oscillator provided approximately 650 mW average output power in a 63-MHz repetition rate train of 95-fs long pulses. The corresponding spectrum was 12-nm wide (FWHM) and centered around 1050 nm. The pulse was stretched to ~400 ps by means of a transmission grating with 1700 grooves/mm in a folded configuration. After passing through the Faraday isolator, the oscillator beam was injected in the amplifier. The amplifier active medium was a water-cooled 4-mm long, 2% doped Yb:CALGO crystal, antireflection-coated for both pump and laser wavelength. The crystal was oriented to absorb $\pi$-polarization and to emit $\sigma$-polarization in order to maximize the efficiency. The fiber-coupled pump laser diode provided as much as 65 W at ~980 nm out of a 200-μm fiber, and
the absorbed pump power was ~84%. The pump and laser beam diameters in the crystal were 400 μm and 340 μm, respectively. This provided the best performance with the available pump power. A 40-mm long double-BBO Pockels cell was used to switch the pulses in and out of the regenerative amplifier. The beam radius in the Pockels cell was expanded to ~0.6 mm in order to reduce self-phase modulation (SPM). After the amplifier the pulses were sent to the compressor, again consisting of a high efficiency transmission grating with 1700 grooves/mm in a folded configuration. The total transmission was 70%. At the maximum pump power we achieved up to 17 W continuous wave power when using an optimum output coupler with transmissivity T = 15% (Fig. 2).

![Fig. 2. Yb:CALGO output power versus incident pump power in cw regime.](image)

In the pulsed regime we performed experiments at different repetition rates ranging from 5 kHz to 500 kHz. In Fig. 3 the output energy is plotted against pump power for different repetition rates. With 40 round trips and 65 W of pump power, we obtained a maximum energy of 1.62 mJ before compression. The pulse was then compressed to 380 fs (see Fig. 4(a)) with an output energy of 1.13 mJ, corresponding to ~3 GW peak power. The pulse spectrum was centered at 1043 nm and the FWHM bandwidth was 5.9 nm. The corresponding time-bandwidth product was 0.62. This relatively large time-bandwidth product, about 2 times the Fourier limit, can be mainly attributed to higher order dispersion terms, nevertheless the spectral width suggests that there is room for substantial improvement in terms of pulse shortening.

Indeed, the B-integral was mostly accounted for by SPM occurring in the BBO cell, and in this experiment with Yb:CALGO it was calculated to be ~0.4π rad, hence it was weakly...
affecting the spectrum. The number of roundtrips with pulse amplitudes > 50% of the final pulse was 5.

At the highest repetition rate of 500 kHz, the average output power was 9.4 W before compression (corresponding to 6.6 W after compression); the compressed pulse duration was 370 fs with an optical spectrum of 5.6 nm (FWHM). The time bandwidth product of 0.57 was comparable to that of the much higher-intensity pulses at 5 kHz, ruling out SPM as the limiting factor for further pulse compression.

The result of the beam quality measurement is shown in Fig. 4(b). The beam was nearly TEM$_{00}$ up to full pump power, with $M^2 = 1.3$ in both axes.

![Fig. 4. a) Autocorrelation and spectrum of the pulses with 1.02 mJ energy at 5 kHz. b) Beam quality measurement and beam profile.](image)

3. Experimental results with Yb:CaF$_2$

The experimental setup was the same as shown in Fig. 1. We simply replaced the Yb:CALGO laser crystal in the regenerative amplifier with a 4-mm long, 2.7% doped Yb:CaF$_2$ crystal, antireflection-coated for both pump and laser wavelength. The crystal was housed in a water-cooled mount for efficient heat removal.

The pump and laser beam diameters in the crystal were kept at 400 μm and 330 μm, respectively. The absorbed pump power in this case was ~52%. We obtained a maximum continuous wave power of 14 W when using an output coupler with optimum transmission of $T = 10\%$ (smaller than in the previous setup, owing to the smaller gain of Yb:CaF$_2$), see Fig. 5.

The slope efficiency with respect to the absorbed pump power was as high as 66% with the Yb:CaF$_2$, higher than for Yb:CALGO crystal (~46%), which had been slightly damaged by previous experiments, adding some intracavity losses.

![Fig. 5. Yb:CaF$_2$ output power versus incident pump power in cw regime.](image)
Figure 6 shows the extracted pulse energy as function of incident pump power for different repetition rates. With 130 round trips and 65 W of pump power, we obtained a maximum energy of 1.46 mJ before compression. The pulse was then compressed to 324 fs (see Fig. 7(a)) with an output energy of 1.02 mJ, corresponding to ~3 GW peak power. The optical spectrum was centered at 1045 nm with a FWHM bandwidth of 7.5 nm. The corresponding time bandwidth product was 0.69. A higher number of round-trips was required due to the smaller gain with respect to Yb:CALGO. Please note that the number of roundtrips with pulse amplitudes > 50% of the final pulse was 10 in this case, increasing the B-integral to ~0.8π rad. Although this value is higher than for Yb:CALGO (~0.4π rad), the pulse spectrum is not significantly affected yet. Therefore, the same considerations on the time-bandwidth product and the possibility to improve pulse shortening also apply for Yb:CaF₂.

At 500 kHz, the average output power was 6.1 W (corresponding to 8.5 W before compression) with a pulse width of 313 fs, and a FWHM optical spectrum of 6.8 nm. The time-bandwidth product is calculated to be 0.58, comparable to that of much higher intensity pulses at 5 kHz.

The result of the beam quality measurement is shown in Fig. 7(b). The beam was nearly TEM₀₀ up to full pump power, with $M^2 = 1.1$ in both axes. The more pronounced beam asymmetry with respect to Yb:CALGO is thereby due to a larger difference in thermal focal lengths in orthogonal directions. However, please note that it will be straightforward to optimize the cavity design for circular output beam.
4. Conclusions

In conclusion we have demonstrated - to the best of our knowledge - the first single-crystal, Ytterbium-doped regenerative amplifiers delivering pulse energies >1 mJ @ 5 kHz and room temperature with pulses shorter than 400 fs.

Taking advantage of the good thermo-mechanical properties of Yb:CALGO and, especially in case of CaF\(_2\) through a careful thermal management and doping level selection, the extracted energy has the potential to be significantly improved with further pump power scaling. An immediate upscaling route with two laser heads can be considered instead of stretching the limit of the present design into unsafe operation regime by mere doubling of the pump lasers. Shorter pulses are also potentially achievable by optimizing the compressor and stretcher design to better compensate higher order dispersion and to further minimize nonlinear effects.

These laser sources are extremely interesting for industrial applications where high pulse energies in combination with a relatively high repetition rate allow for considerably reducing the manufacturing throughput time. Furthermore, these laser sources are extremely flexible in providing repetition rates up to 500 kHz (and even up to 1 MHz) with average power ~10 W for applications requiring extremely fast processing times with lower energy femtosecond pulses.