



Multi-millijoule class, high repetition rate, Yb:CALYO regenerative amplifier with sub-130 fs pulses

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Abstract: We demonstrate a high-energy, 1 kilohertz, Yb-based, femtosecond regenerative amplifier in a chirped pulse amplification (CPA) architecture by using a single disordered Yb:CALYO crystal, providing 125 fs pulses of 2.3 mJ energy per pulse at a central wavelength of 1039 nm. The amplified compressed pulses, with a spectral bandwidth of 13.6 nm, represent the shortest ultrafast pulse duration reported to date for any multi-millijoule class, Yb-crystalline classical CPA system without additional spectral broadening techniques. We have demonstrated an increase in the gain bandwidth proportionally to the ratio of the excited to total Yb³⁺ ion densities. A net wider spectrum of the amplified pulses is the result of the interplay between the increased gain bandwidth and the gain narrowing. Finally, our broadest amplified spectrum of 16.6 nm, corresponding to a 96 fs transform limited pulse, can be expanded further to support sub-100 fs pulse durations and 1–10 mJ energies at 1 kHz.

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1. Introduction

In the last decade, Yb-based laser amplifiers have established themselves as a promising technology for the generation of intense ultrafast femtosecond pulses, using compact and robust diode laser pumping. The transitions between the ²F_{5/2}–²F_{7/2} manifold energy levels govern the distinctive optical properties of the Yb-doped crystalline materials, which include broad spectral bandwidths of luminescence and absorption lines, as well as allow for straightforward pumping with well-engineered, fiber-coupled InGaAs lasers with scalable high average power [1]. Since the beginning of the Yb laser era, one of the grand challenges in the development of femtosecond Yb-based amplifiers is achieving multi-millijoule level pulse energies combined with pulse durations of sub-100 femtoseconds, or potentially as short as 10s of femtoseconds, at a kHz repetition rate. Historically, Yb:YAG laser amplifiers have been a great option for applications where high energy and intermediate power levels are required. However, their pulse durations are limited to 500–1000 fs due to gain narrowing and the relatively narrow bandwidth of the emission spectrum of roughly 9 nm at 20°C [2]. To overcome the limitations of the gain narrowing effect and the limited emission band of the Yb-crystalline materials, a variety of techniques have been combined with Yb-CPA lasers to expand the spectral bandwidth of the amplified pulses. These techniques include: non-linear broadening of the spectrum by self-phase modulation (SPM)

[3–5], dual crystal amplification with different crystal orientations to merge their gain bandwidth [6,7], seed-pulse spectral shaping [5,8], etc. Some of them enable the generation of pulses with tens of femtosecond pulse duration at relatively high energy, however, they also increase the complexity of the system by requiring additional optical setups, and most importantly, do not address the central issue of how to achieve the shortest pulse durations at mJ-levels of pulse energy for a single-crystal Yb CPA in a classical architecture [9], currently well developed and of wide industrial use for femtosecond Ti:sapphire amplifiers. Yb-materials with the broadest emission lines are favored in this classical CPA architecture where the pulses from a femtosecond oscillator are supplied straight to an optical stretcher, then amplified in a regenerative amplifier operating below the threshold of nonlinear effects, and optically compressed at the end. Although there are Yb crystals with broader bandwidths of emission [10,11], such as Yb-doped calcium gadolinium aluminates (Yb:CaGdAlO₄ or Yb:GALGO), potassium gadolinium tungstates (Yb:KGd(WO₄)₂ or Yb:KGW) [12,13], potassium yttrium tungstates (Yb:KY(WO₄)₂ or Yb:KYW) [13,14], calcium fluorides (Yb:CaF₂) [15], and rare earth sesquioxides (Yb:(Sc_x,Lu_y,Y_z)₂O₃), obtaining amplified pulses with energy greater than 1 mJ and pulse durations shorter than 160 fs is still a main thrust of scientific and technological research. Among many Yb crystals, Yb:KGW and Yb:KYW provide great amplification properties. However, their performance is limited by the relatively narrow amplification bandwidth of less than 18 nm [16]. Ytterbium-doped tetragonal rare-earth calcium aluminates, Yb:CaLnAlO₄ (Ln=Gd, Y), i.e. disordered Yb:CALGO and Yb:CaYAlO₄, also known as Yb:CALYO or Yb:CYA [17], which have ultra-broad emission spectra of 60 to 80 nm and relatively good thermo-optical properties [1,18], are highly promising for ultrashort pulse amplification. While Yb:CALGO crystal has been widely adopted for amplification of short femtosecond pulses, to date, the most advanced systems feature spectral shaping of the seed pulse [5,8], however, no significant development has been disclosed where a straightforward CPA architecture has achieved the desired scaling. On the other hand, the relatively new Yb:CALYO is unexplored. In the last decade, there have been no reports of its use in classical CPAs without any additional spectral broadening techniques, and the shortest amplified pulses were limited to about 190 fs with 11.5 μJ of energy at 200 kHz [19]. The prospect of developing a Yb-based simple CPA laser featuring multi-millijoule pulses, substantially shorter than 150 fs has recently been opened up by preliminary work demonstrating 135 fs output pulses with 1 mJ energy using a single crystal Yb:CALYO regenerative amplifier [20]. Here, we present femtosecond amplified pulses in a compact single-crystal regenerative amplifier based on disordered Yb:CALYO with an energy greater than 2.9 mJ after the regenerative amplifier and a broad compressible spectrum of >13.6 nm (FWHM), centered at 1039 nm. The laser pulses were compressed to 125 fs with 2.3 mJ energy per pulse at 1 kHz, which is the shortest pulse duration reported to date for any millijoule class Yb-based laser amplifier in classical CPA architecture, i.e. without using any additional techniques or optical schemes for spectral broadening and post-compression.

2. Yb:CALYO single-crystal regenerative amplifier architecture

We have designed and demonstrated a classical chirped pulse amplification laser [9] based on a regenerative amplifier with single-crystal Yb:CALYO architecture, pumped by a single fiber-coupled laser diode, as illustrated schematically in Fig. 1. All control and synchronization electronics and laser diode power supplies have been developed in collaboration with IBPhotonics Ltd. The custom-designed seed Yb-based fiber laser has 2.7 nJ energy per pulses at 52 MHz and a spectral bandwidth of more than 45 nm (FWHM), centered at 1035 nm. Seed pulses with 0.5 nJ energy are stretched using a Martinez transmission diffraction grating geometry and are injected into the Yb:CALYO regenerative amplifier. The diffraction gratings of the stretcher and the Treacy compressor are transmission type with matching groove densities of 1700 lines/mm. The cavity of the regenerative amplifier is based on a single active element (z-type design) with an overall length of 1546 mm. A fiber-coupled diode stabilized for lasing at 981 nm is used as a

pump, focused at a $\sim 350\ \mu\text{m}$ diameter spot using an objective. The beam propagation quality factor of the laser diode is $M^2_{x,y} = 20$, and the fiber core diameter is $105\ \mu\text{m}$. The pump pulses are $500\ \mu\text{s}$ long, operating at a $1\ \text{kHz}$ repetition rate.

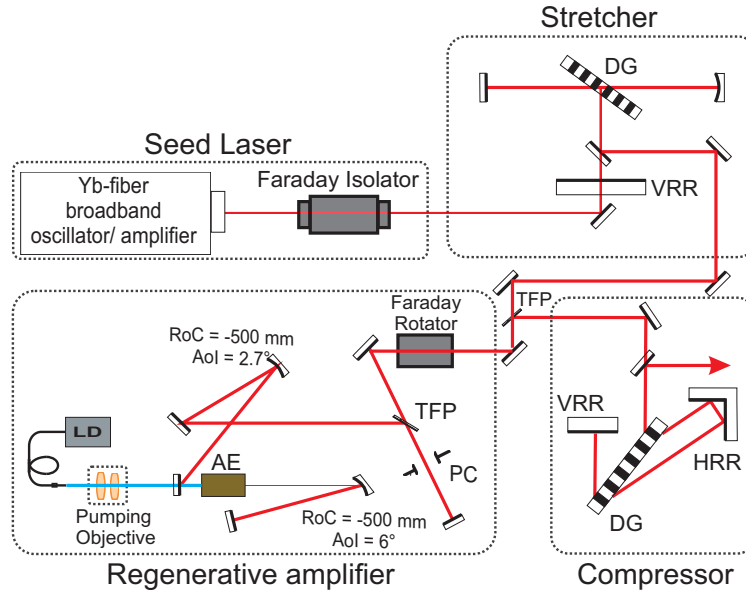


Fig. 1. Schematic of the compact CPA, consisting of a seed laser module, a transmission grating stretcher, a regenerative amplifier, and a transmission grating compressor. DG – transmission diffraction grating, VRR – vertical roof reflector, HRR – horizontal roof reflector, LD – fiber-coupled laser diode pump, AE – active element, PC – Pockels cell, TFP – thin film polarizer. The cavity is z-type with two identical concave mirrors with radius of curvature 500 mm, the longer arm (with the PC) is 719 mm, the distance between the two concave mirrors is 542 mm, and the shorter arm is 285 mm. The Yb:CALYO crystal is placed before the waist between the concave mirrors to best match the cavity mode with the pump waist.

Amplification using the ordinary σ polarization of an 8 mm and 5 mm thick a-cut Yb:CALYO crystals, doped with 2% of Yb^{3+} ions, has been studied. This resonator design is selected to provide great mechanical stability and ease of scaling of the resonator mode diameter. The overall cavity length has a round trip of 10.4 ns. A Pockels cell with a 25 mm long BBO crystal is used to select the amplified pulses out of the cavity.

3. Experimental results

3.1. Laser pulse spectrum broadening

The experiments described in this section reveal how the spectral width of the pulse, and hence its duration, depends on the ratio of excited Yb^{3+} -ions density to all those within the laser application region, and how it is affected by the gain narrowing effect in Yb-disordered crystals. According to the theoretical prediction for Yb:CALYO the gain spectral bandwidth should grow as the ratio of excited Yb-ion density to total Yb-ion density increases (RETID) [19]. We investigate the amplification in two crystals of different lengths that are interchanged in a regenerative amplifier cavity under identical conditions to explore this phenomenon. As a result, the RETID for the 5 mm crystal is 1.27 times greater than for the 8 mm crystal. In both cases we the regenerative amplifier with pulses that are first stretched to 750 ps with a spectrum of 25 nm (FWHM). We

calculate stretcher group delay dispersion (GDD) of $14 \times 10^6 \text{ fs}^2$. The crystals are pumped by a fiber-coupled diode with 65 W peak power, which corresponds to 32.5 mJ pump energy. For the 8 mm thick Yb:CALYO crystal, 80% of the pump power is absorbed in the active medium and we achieved more than 2 mJ output energy before compression. In this setup, the seed pulses need 81 roundtrips to reach saturation. The obtained spectral bandwidth of the amplified compressed pulses is 11.5 nm (FWHM) with 1.5 mJ energy, shown in (Fig. 2(A)), while the measured autocorrelation trace of 226 fs indicates a pulse duration of 147 fs, assuming a sech^2 temporal pulse shape (Fig. 2(C)).

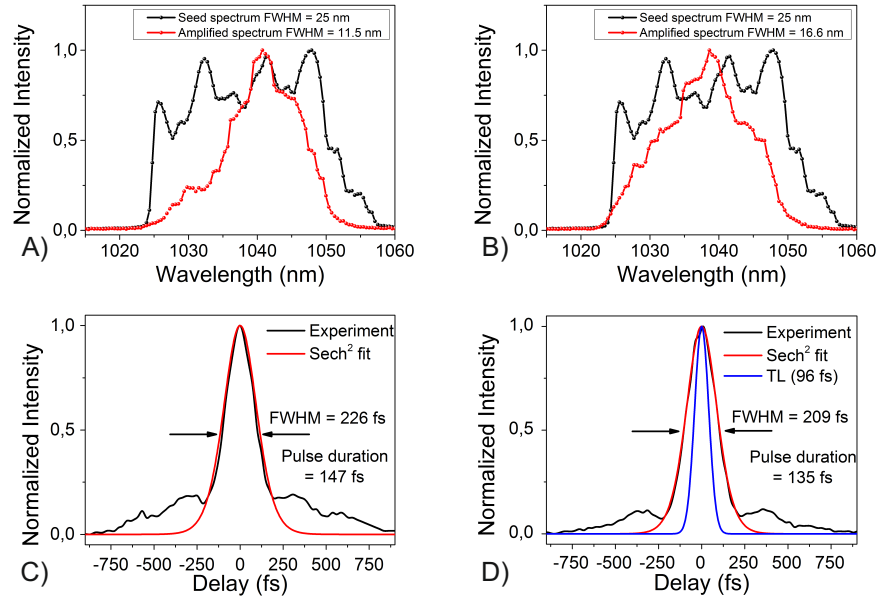


Fig. 2. Spectrum and autocorrelation measurements of compressed 1039 nm laser pulses, demonstrating a spectrum widening effect, from 11.5 nm to 16.6 nm, and pulse shortening effect from 226 fs to 209 fs autocorrelation traces, corresponding to 147 fs and 135 fs sech^2 pulses, when shorter active medium is used. A) and C) Measurements using 8 mm, and B) and D) for 5 mm long Yb:CALYO.

On the other hand, the 5 mm thick Yb:CALYO crystal absorbed 63% of the pump power. Here, the seed pulses require 100 roundtrips to achieve saturation, and the output energy decreases to 1.4 mJ before compression due to the lower number of excited Yb-ions in the active media. The amplified spectrum is broadened to 16.6 nm FWHM (Fig. 2(B)). This bandwidth increase (by 5 nm) is due to the fact that the RETID grows 1.27 times that of the 8 mm crystals. The measured autocorrelation traces of 209 fs and 226 fs (FWHM) for 5 mm and 8 mm crystals, respectively, show a decrease of the pulse duration, which corresponds to the observed spectral broadening. An estimate value of the pulse duration for the shorter pulse is 135 fs, assuming sech^2 pulse shape (Fig. 2(D)). The output energy in this case is lower (1 mJ) after compression due to the lower pump energy absorbed. The 16.6 nm spectrum allows compression to 96 fs transform-limited pulse durations. In our case the longer pulses are due to the uncompensated higher orders of dispersion and the additional nonlinear phase, B-integral [21], for the 5 mm crystal ~ 3 rad.

3.2. Optimized highest laser energy and shortest pulse duration

To achieve multi-mJ pulse energy, we select an 8 mm thick Yb:CALYO crystal in the regenerative amplifier described on Fig. 1. The crystal was pumped by a fiber-coupled laser diode with 100

W peak power, providing up to 50 mJ pump energy. The seed pulse was stretched up to 850 ps (calculated GDD = 16×10^6 fs²) to reduce the B-integral of the system in order to avoid nonlinear effects. The spectrum of the seed was cut to 20 nm to avoid clipping from the limited aperture of components in the stretcher. The seed amplification is evaluated for different pump energies starting at 25 mJ which corresponds to ~ 1 mJ energy of the output pulse. The pronounced output pulse spectrum broadening as a function of pump energy is shown in Fig. 3(A). The compressed pulses' duration and energy were measured simultaneously for the same pump levels (Fig. 3(B)). We observe saturation in the broadening of the spectrum as well as pulse shortening for pump energies above 42 mJ (Fig. 3(A),(B)). The width of the spectrum of the amplified pulse changes from 10.8 nm to 13.6 nm and the significant shortening from 152 fs down to 125 fs was observed as the pump increased from 25.3 mJ to 45 mJ. The output energy changes respectively from 1.46 mJ to 2.9 mJ for uncompressed pulses and from 1.17 mJ to 2.32 mJ for compressed pulses. The amplified beam has an excellent spatial profile close to a near-perfect Gaussian TEM₀₀ mode with measured M² factors of M²_x = 1.08 and M²_y = 1.16 after the compressor (Fig. 4).

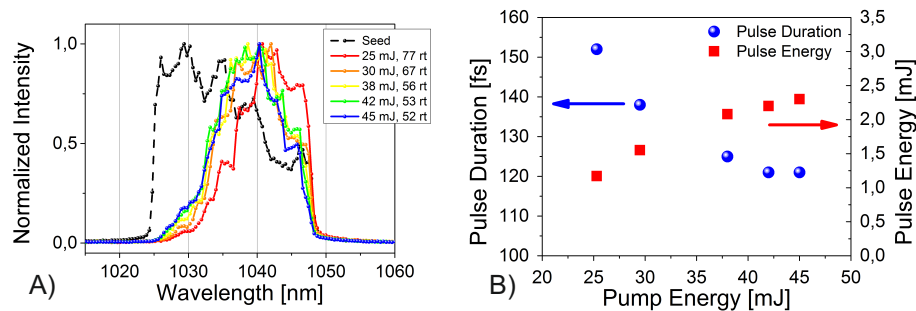


Fig. 3. A) Broadening of the spectrum of the compressed pulses and roundtrips (rt) in RA cavity for different pump energy. B) Increase of the compressed pulse energy correlates favorably with shorter pulse durations.

FROG is used to characterize the pulses. The compression is improved using feedback from the FROG traces and the retrieved phase and pulse durations. The FROG characteristics of the shortest pulse duration of 125 fs with the highest pulse energy of 2.3 mJ at 1 kHz repetition rate are shown in Fig. 5. The inset in Fig. 5(C) shows the marginal autocorrelation trace from the FROG measurement. It can be seen that the higher orders of dispersion are much better compensated, compared to the autocorrelations in Fig. 2. This result demonstrates the shortest ultrafast pulse duration generated to date directly from any multi-millijoule class, single-crystal, Yb-based regenerative amplifier without using any complex post-compression techniques, nonlinear spectral widening techniques or seed spectral shaping techniques. Additional shortening of the pulse duration is also feasible by redesigning the system to reduce clipping of the 45 nm broad bandwidth seed by geometrical and optical apertures.

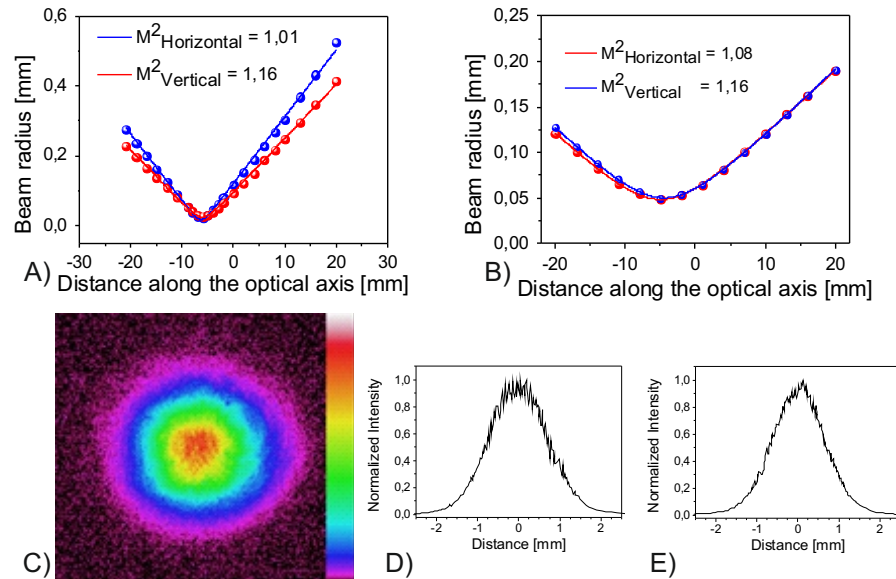


Fig. 4. M^2 measurements of the amplified 1039 nm laser beam A) directly after the amplifier, and B) after the transmission grating compressor, demonstrating C) an excellent near-perfect Gaussian beam profile, with the beam profiles D) horizontal and E) vertical.

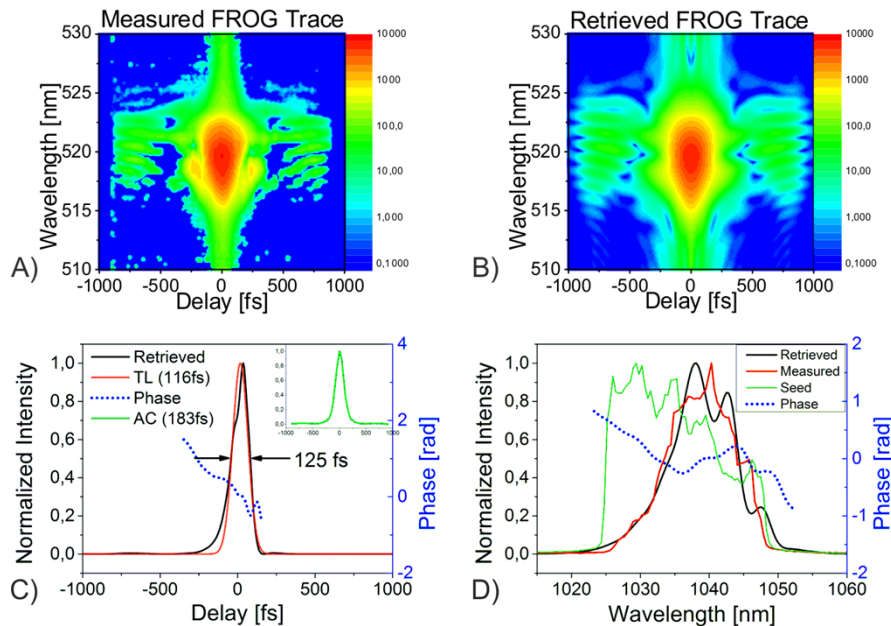


Fig. 5. Phase retrieval FROG measurement: A) experimental data and reconstruction B) of the shortest 125 fs compressed laser pulses, in logarithmic scale. C) Retrieved pulse intensity (black) with some residual high-order temporal phase (blue dotted line), and corresponding Fourier transform limited pulse duration of 116 fs (red), autocorrelation trace with 183 fs duration as inset (green). D) Measured spectrum (red), retrieved spectrum (black), and spectral phase (blue dotted line) of the compressed pulse, compared to the seed spectrum (green). The FROG error for this measurement was 0,002.

4. Conclusion

In summary, we have presented a comprehensive study of the spectrum widening of amplified pulses with an increase in pump energy in Yb-disordered crystals (Yb:CALYO). First, it was found that by increasing the pump, while the ratio of the excited-to-total Yb³⁺-ion densities increases, the gain bandwidth also increases. It dominates the gain-narrowing, resulting in a net gain broadening of the spectrum of the amplified pulses. When the ratio is increased by 27%, the spectrum broadens from 11.6 nm to 16.6 nm. Second, this gain bandwidth broadening reaches saturation at high enough pump energies, setting a limitation in further shortening of the amplified pulse. Based on the aforementioned factors, we have developed and demonstrated an ultrafast multi-millijoule class Yb:CALYO regenerative amplifier with a 125 fs pulse duration, 2.3 mJ of energy at a 1 kHz repetition rate. It is the shortest femtosecond pulse with a multi-mJ energy reported for any Yb-based straightforward CPA system [9] i.e., without additional spectral broadening techniques. This rapid progress in the development of broadband Yb-based amplifiers in the past few years establishes Yb:CALYO as one of the most promising Yb-doped materials for a new class multi-mJ high power femtosecond amplifiers with greater than 20 nm compressible spectra [20,22]. Finally, since in disordered Yb³⁺ crystals the bandwidth of the gain cross-section broadens for higher population inversion, the shortest generated pulses are favorably produced at the highest output laser energies. This allows for a robust and up-front scaling to much shorter sub-100 fs pulses and higher 1-10 mJ energies in the near future, ideal for strong field experiments and fully coherent EUV and X-ray generation based on extreme high harmonic upconversion schemes [23,24].

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Disclosures. LSP: IBPhotonics Ltd. (F)

Data availability. Data underlying the results presented in this paper are available in Ref. [25] and Ref. [26].

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