

# Yb:CALYO-based femtosecond chirped pulse regenerative amplifier for temporally resolved pump-probe spectroscopy

Alexander Rudenkov<sup>1</sup>, Viktor Kisel<sup>1</sup>, Anatol Yasukevich<sup>1</sup>, Karine Hovhannesian<sup>2</sup>, Ashot Petrosyan<sup>2</sup>, Nikolai Kuleshov<sup>1</sup>

<sup>1</sup>Center for Optical Materials and Technologies,  
Belarusian National Technical University,  
Nezavisimosty Ave., 65, Minsk 220013, Belarus

<sup>2</sup>Institute for Physical Research, National Academy of Sciences of Armenia,  
0203, Ashtarak-2, Armenia

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## Abstract

Diode-pumped femtosecond chirped pulse regenerative amplifiers based on Yb<sup>3+</sup>-materials are of practical importance for wide range of scientific, industrial and biomedical applications. The aim of this work was to study the amplification of broadband chirped femtosecond pulses in regenerative amplifier based on Yb<sup>3+</sup>:CaYAlO<sub>4</sub> crystal.

Such systems use femtosecond mode-locked lasers as seed pulse sources and amplify nJ-seed pulses to sub-mJ energy range. Most chirped pulse regenerative amplifier systems described in the literature use seed lasers with typical pulse spectral width at the level of 10–15 nm full width at half maximum (FWHM) that limit the seed pulse duration of about 90 fs and amplified pulse duration at the level of 200 fs due to strong influence of gain narrowing effect on the amplified pulse parameters. Yb<sup>3+</sup>-doped crystals with wide and smooth gain bandwidth as an active medium of chirped femtosecond pulse regenerative amplification systems allow to reduce negative contribution of gain narrowing effect and lead to shortening of amplified pulses. In this research we study the chirped pulse regenerative amplification of broad-band femtosecond pulses (60 nm spectral width FWHM) in the Yb<sup>3+</sup>:CaYAlO<sub>4</sub>-based chirped pulse regenerative amplifier. Substantial reduction of the amplified pulse duration down to 120 fs (19.4 nm spectral width FWHM) with average power of 3 W at 200 kHz pulse repetition frequency was demonstrated without any gain narrowing compensation technique.

The results of experimental investigation of broad-band seeded Yb<sup>3+</sup>:CaYAlO<sub>4</sub>-based chirped pulse regenerative amplifier are reported for the first time to our knowledge. 120 fs-pulses (19.4 nm FWHM) with average output power of 3 W were demonstrated without any gain narrowing compensation technique. Despite the significant reduction of amplified pulse duration the task of improvement group velocity dispersion balance (including high orders of group velocity dispersion) remains relevant.

**Keywords:** broad-band chirped pulses, chirped pulse amplifier, regenerative amplifier, spectral broadening,

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### Адрес для переписки:

Руденков А.С.  
Центр оптических материалов и технологий, Белорусский  
национальный технический университет,  
пр-т Независимости, 65, г. Минск 220013, Беларусь  
e-mail: a.rudenkov@bntu.by

---

### Address for correspondence:

Alexander Rudenkov  
Center for Optical Materials and Technologies, Belarusian National  
Technical University,  
Nezavisimosty Ave., 65, Minsk 220013, Belarus  
e-mail: a.rudenkov@bntu.by

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УДК 621.373.826

# Регенеративный усилитель чирпированных фемтосекундных импульсов на основе кристалла Yb:CALYO для спектроскопии возбуждения-зондирования с высоким временным разрешением

Руденков А.С.<sup>1</sup>, Кисель В.Э.<sup>1</sup>, Ясюкевич А.С.<sup>1</sup>, Ованесьян К.Л.<sup>2</sup>,  
Петросян А.Г.<sup>2</sup>, Кулешов Н.В.<sup>1</sup>

<sup>1</sup>Центр оптических материалов и технологий,  
Белорусский национальный технический университет,  
пр-т Независимости, 65, г. Минск 220013, Беларусь

<sup>2</sup>Институт физических исследований Национальной академии наук Армении,  
0203, Аштарак-2, Армения

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Регенеративные усилители чирпированных фемтосекундных импульсов на основе материалов с ионами Yb<sup>3+</sup> с диодной накачкой нашли широкое применение в различных отраслях науки, производства и медицины. Целью данной работы являлось исследование режима регенеративного усиления широкополосных чирпированных фемтосекундных импульсов в усилителе на основе кристалла Yb<sup>3+</sup>:CaYAlO<sub>4</sub>.

Используя в качестве задающего генератора лазер с пассивной синхронизацией мод, данные системы усиливают импульсы наноджоулевого диапазона энергий до суб-миллиджоулевого уровня благодаря методике усиления чирпированных импульсов. Большинство описанных в литературе систем усиления используют задающие генераторы, обеспечивающие фемтосекундные импульсы со спектральной полушириной в диапазоне 10–15 нм, что ограничивает минимальную длительность задающих импульсов на уровне 90 фс. В процессе регенеративного усиления длительность усиленных импульсов увеличивается до значений около 200 фс, что связано с сильным негативным влиянием эффекта сужения спектра импульса под воздействием полосы усиления активной среды усилителя. Применение кристаллов, имеющих широкие и гладкие полосы усиления в качестве активных сред систем усиления чирпированных фемтосекундных импульсов широкого спектрального диапазона, позволяет снизить негативный вклад эффекта сужения спектра импульса и приводит к сокращению длительности усиленных импульсов.

В работе впервые представлены результаты исследования режима регенеративного усиления широкополосных чирпированных фемтосекундных импульсов в усилителе на основе кристалла Yb<sup>3+</sup>:CaYAlO<sub>4</sub>. Получены импульсы длительностью 120 фс (спектральная полуширина 19,4 нм) со средней выходной мощностью системы усиления 3 Вт без применения методик компенсации эффекта сужения спектра усиливаемого импульса.

**Ключевые слова:** чирпированные импульсы широкого спектрального диапазона, усилитель чирпированных импульсов, регенеративный усилитель, расширение спектра импульса, эффект сужения спектра импульса.

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## Introduction

Diode-pumped femtosecond laser sources with pulse repetition frequencies (PRF) of hundreds of kilohertz and pulse energies of tens microjoules are of practical importance for high temporal and spectral resolution measurements, precision micromachining, optical memory and biomedicine [1]. Extremely high average output powers up to the kilowatt level could be obtained by means of direct amplification of high repetition-rate oscillators. 1.1 kW of average power at 20 MHz repetition rate with 615 fs pulses were obtained using the Innoslab Yb:YAG concept [2]. 830 W trains of 640 fs pulses at 78 MHz were demonstrated by employing large-mode area Yb-doped fiber amplifiers [3]. Pulses at lower repetition frequencies, up to a few megahertz, with substantially higher energy and peak power are preferred for many applications. These pulse trains can be generated conveniently with solid-state diode-pumped regenerative amplifiers (RAs). Up to now, the highest average power RA systems are presented by thin-disk concept RAs. For example, up to 160 W average output power at 800 kHz PRF with 750 fs pulse duration were achieved in [4]. Up to 100 W average power at 400 kHz PRF with 800 fs pulse duration were reached in [5] employing Yb:YAG thin-disk active element. Despite thin-disk based regenerative amplifier systems demonstrate high average power, it should be noted that pulse duration of such systems is not less than 700 fs. 295 fs pulses were demonstrated for thin disk RA system that applied nonlinear pulse amplification regime but with substantially reduced output power (36 W) [6]. Another approach to implementation of RA systems is based on bulk regenerative amplifiers. The highest output power reported so far for bulk RAs is 42 W at 500 kHz pulse repetition frequency obtained in RA based on the active medium with high thermo-optical properties – Yb:Lu<sub>2</sub>O<sub>3</sub> [7]. Relatively long amplified pulses of about 780 fs pulse duration obtained due to narrow gain bandwidth of Yb:Lu<sub>2</sub>O<sub>3</sub>. Substantially reduced pulse duration of 217 fs with relatively high output power of 28 W demonstrated at 500 kHz in RA based on crystal with wide gain bandwidth – Yb:CALGO [8]. But the usage of active medium with wide gain bandwidth is not a sufficient condition for obtaining short amplified pulse duration due to the strong gain narrowing effect [9] that reduce amplified pulse spectral width and increase minimal transform-limited pulse duration. Several methods for overcoming the negative contribution of gain

narrowing effect have been proposed [10, 11]. The output power of about 21 W at 200 kHz PRF with 200 fs pulse duration is obtained with Yb:KGW dual crystal system [10]. Femtosecond laser pulses with duration as short as 97 fs with output power of 1.2 W at 50 kHz PRF were obtained with the Yb:CALGO RA system which demonstrates the possibility of sub-100 fs pulses amplification [11].

Despite the availability of femtosecond lasers providing wide spectral width pulses [12–14] based on Yb seed lasers a large number of RA systems described in the literature have narrower pulse spectral width not over than 15 nm. And this also limits the amplified pulse spectral width and compressed pulse duration.

The aim of this work was to study the amplification of broadband chirped femtosecond pulses in regenerative amplifier based on Yb<sup>3+</sup>:CaYAlO<sub>4</sub> crystal.

## Crystal growth

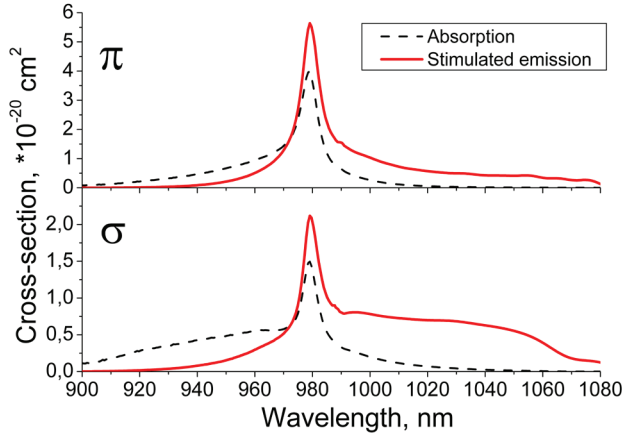
Single crystals of Yb<sup>3+</sup>:CaYAlO<sub>4</sub> (tetragonal structure, space group D<sup>17</sup><sub>4h</sub>-I4/mmm) doped with Yb<sup>3+</sup> (1.4 at. % and 3.5 at. %) were grown from stoichiometric melts by Czochralski method [15]. Yb<sup>3+</sup> ions occupy Y<sup>3+</sup> sites of the lattice with 9-fold coordination. Crystallization was carried out under an enclosed argon atmosphere using iridium crucibles (50 × 50 × 30 mm<sup>3</sup>) and seed crystals oriented along [110]. The pulling and rotation rates were 1.5–2.5 mm/h and 15–25 rev/min respectively. Transparent crystals 15 mm in diameter and 20–30 mm long were obtained for the present studies.

## Spectroscopy

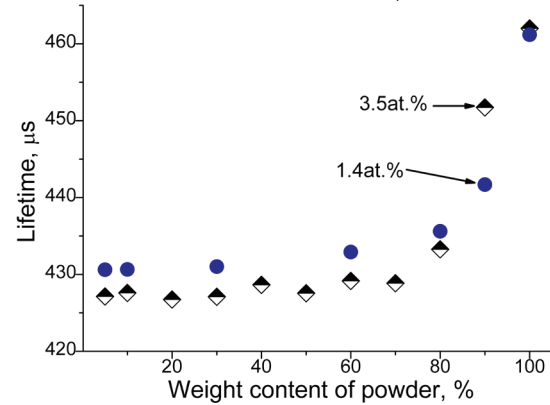
Polarized absorption spectra of Yb<sup>3+</sup> (1.4 at. %): CaYAlO<sub>4</sub> (correspondent ytterbium volume concentration is 1.82 · 10<sup>20</sup> cm<sup>-3</sup>) at room temperature were measured by a Varian CARY 5000 spectrophotometer. The absorption cross-section spectra for different polarizations are shown in Figure 1.

Strong absorption is found for  $\pi$ -polarized light. A peak absorption cross-section at 979 nm was measured to be about 4 · 10<sup>-20</sup> cm<sup>2</sup>, with comparatively wide bandwidth of about 9.5 nm (FWHM). For  $\sigma$ -polarization maximal absorption cross section near 979 nm was 1.5 · 10<sup>-20</sup> cm<sup>2</sup>. Radiative lifetime was estimated by using crystalline powder immersed in glycerine suspension in order to eliminate radiation trapping effect caused by significant overlap of the absorption and emission bands [16, 17]. Measured lifetime for different weight content

of  $\text{Yb}^{3+}$ (1.4 at. %): $\text{CaYAlO}_4$  and  $\text{Yb}^{3+}$ (3.5 at. %): $\text{CaYAlO}_4$  crystalline powder in glycerine suspension are shown in Figure 2. Measured kinetics of luminescence decay for  $\text{Yb}^{3+}$ : $\text{CaYAlO}_4$  with 1.4 at. % and 3.5 at. % concentrations are shown in Figure 3.

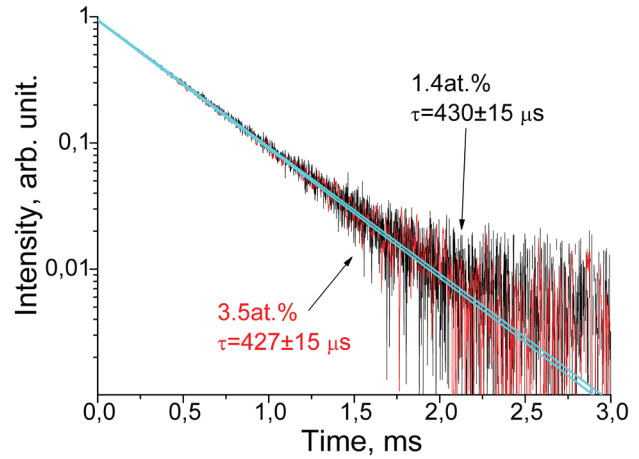


**Figure 1** – Absorption and stimulated emission cross-section spectra of the  $\text{Yb}^{3+}$ : $\text{CaYAlO}_4$



**Figure 2** – Measured lifetime for different weight content of  $\text{Yb}^{3+}$ : $\text{CaYAlO}_4$  crystalline powder in glycerine suspension

The radiative lifetime of the  $^2F_{5/2}$  manifold of  $\text{Yb}^{3+}$ -ions in  $\text{CaYAlO}_4$  was estimated to be  $(430 \pm 15) \mu\text{s}$ . The stimulated emission cross sections (Figure 1) were calculated by use of integral reciprocity method [18].

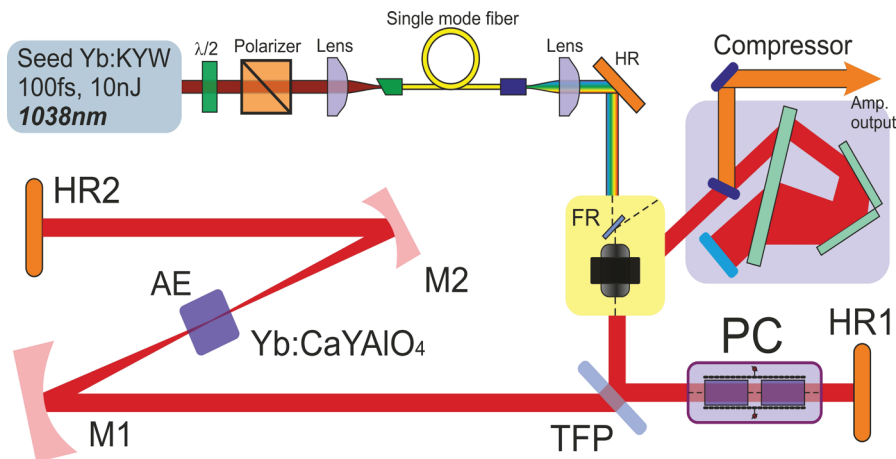


**Figure 3** – Kinetics of luminescence decay  $^2F_{5/2}$  manifold of  $\text{Yb}^{3+}$ -ions in  $\text{CaYAlO}_4$

The  $\text{Yb}^{3+}$ : $\text{CaYAlO}_4$  crystal exhibits broad stimulated emission cross-section spectra in the range 990–1080 nm for both polarizations. The stimulated emission cross-section for  $\sigma$ -polarization demonstrates higher value of about  $\approx 0.7 \cdot 10^{-20} \text{ cm}^2$  at 1030–1040 nm in comparison with  $\approx 0.5 \cdot 10^{-20} \text{ cm}^2$  for  $\pi$ -polarization. Obtained spectroscopic properties of the  $\text{Yb}^{3+}$ : $\text{CaYAlO}_4$  crystal demonstrate good agreement with described in literature data [19].

## Experimental setup

The conceptual scheme of the system layout is shown in Figure 4.



**Figure 4** – Experimental setup of broad-band seeded  $\text{Yb}^{3+}$ : $\text{CaYAlO}_4$  chirped pulse RA



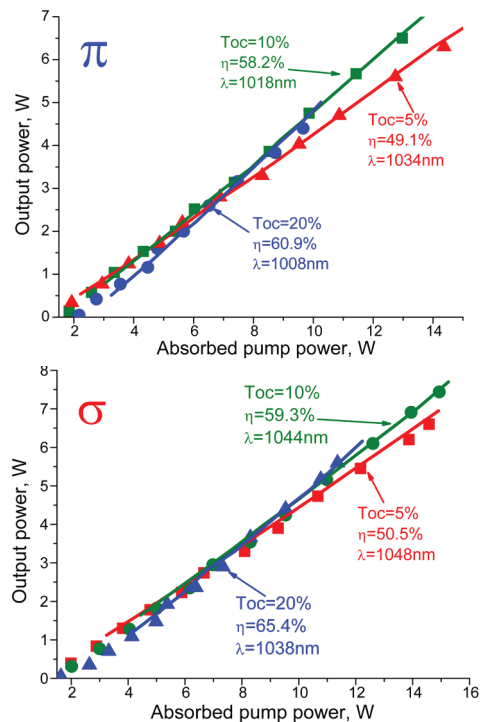
As a seed source laser diode-pumped Yb:KYW oscillator was used which provided 100 fs pulse train with 70 MHz PRF and 10 nJ single pulse energy. The seed pulse spectrum was about 12.5 nm wide (FWHM) and centered at 1038 nm. A 10-m-long single mode Ø9/125 µm telecom fibre was used for pulse spectral broadening and temporal stretching ( $t_{\text{pulse}} \approx 7.5$  ps). After passing through a Faraday isolator, the seed pulse was injected into the RA. The isolator was employed to protect the seeder from high-intensity back reflections and, at the same time, for separating the amplified output pulse from the seed oscillator. The RA setup chosen for this experiment is quite common, employing a 40-mm-long double-BaB<sub>2</sub>O<sub>4</sub> Pockels cell for pulse injection and ejection. Pulse repetition frequency (PRF) was chosen to be 200 kHz to prevent damage of the optical elements. «Off-axes» pump layout was used for longitudinal pumping of the active element [20–22]. Main advantage of such a pump scheme is that all the cavity mirrors have highly reflecting coating at (900–1100) nm. Maximum pump power was 25 W. 2mm-long a-cut Yb<sup>3+</sup>(3.5 at. %):CaYAlO<sub>4</sub> crystal was used as a gain medium. The last unit of the amplifier system is compressor based on transmission diffraction grating with 1000 grooves per millimetre.

### Continuous wave laser experiment

At the beginning we have tested the gain crystal under CW lasing. Laser cavity had the same geometry as a RA cavity without Pockels cell. One of the HR flat mirrors was replaced by the output coupler (OC) with different transmittances. Dependencies of the laser output power on the absorbed pump power for  $\pi$ - and  $\sigma$ -polarized output and different OCs are shown in Figure 5. Absorbed pump power was real time measured during the laser action.

The maximum CW output power of 7.4 W at absorbed pump power of 14.9 W with slope efficiency as high as 59.3 % was obtained with 10 % OC

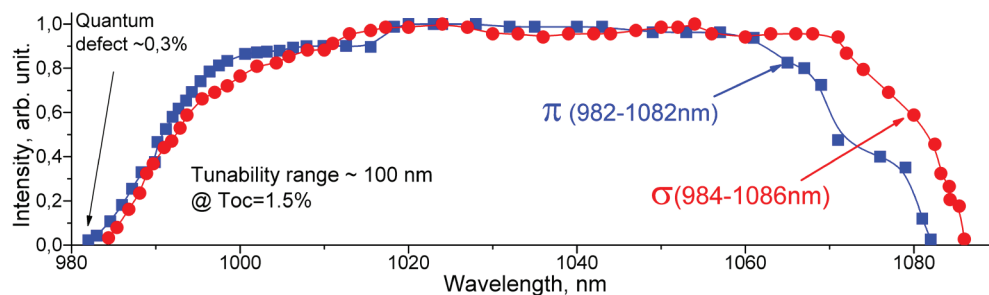
transmittance and  $\sigma$ -polarized output. The output powers of 6.6 W and 5.6 W and slope efficiencies of 50.5 % and 65.4 % were demonstrated with 5 % and 20 % OC transmittances, respectively. For  $\pi$ -polarization output powers of 6.3 W, 6.5 W, 4.4 W with slope efficiencies of 49.1 %, 58.2 %, 60.9 % were obtained with 5 %, 10 % and 20 % OC transmittances.



**Figure 5** – Dependencies of output power of Yb<sup>3+</sup>:CALYO CW laser on absorbed pump power for different OC transmittances and polarizations

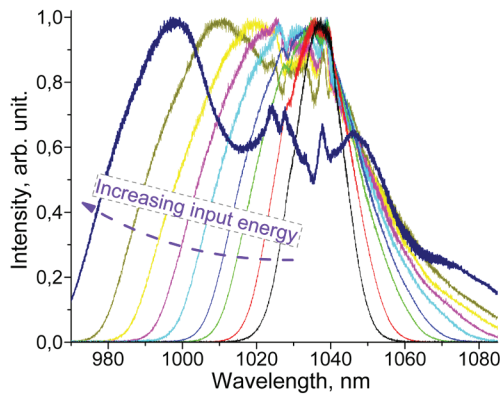
The tunability curves of the Yb<sup>3+</sup>(3.5 at. %):CaYAlO<sub>4</sub> crystal were measured with 1.5 % OC transmission during CW experiments. Central wavelength of the Yb<sup>3+</sup>:CaYAlO<sub>4</sub> laser was tuned in the range of about 100 nm from 982 to 1082 nm for  $\pi$ -polarisation and from 984 to 1086 nm for  $\sigma$ -polarisation (Figure 6).

The results of CW laser experiments show high prospects of Yb<sup>3+</sup>:CaYAlO<sub>4</sub> crystal as an active medium of laser systems operating in a wide spectral range with high average output power.

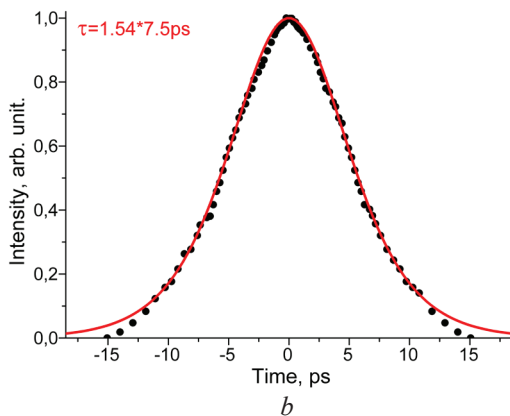
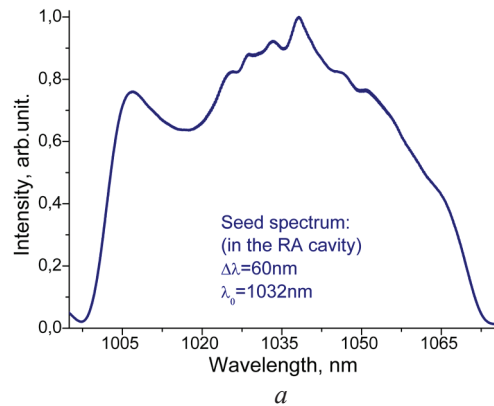


**Figure 6** – Tunability curves of the CW Yb<sup>3+</sup>:CALYO laser

Before the chirped pulse RA experiment seed pulses spectral broadening in fiber was investigated. High peak power of femtosecond pulses provides efficient spectral broadening of seed pulses due to the influence of nonlinear effects such as Raman scattering and self-phase modulation. Seed pulse spectral shape after the fiber in dependence of the incident pulse energy ( $E_{\text{inc}} = 0.15\text{--}7\text{ nJ}$ ) are shown in Figure 7. Intracavity seed pulse spectrum and autocorrelation trace for maximum incident pulse energy are shown in Figure 8.



**Figure 7** – Pulse spectra at the output of the fiber for different incident energy

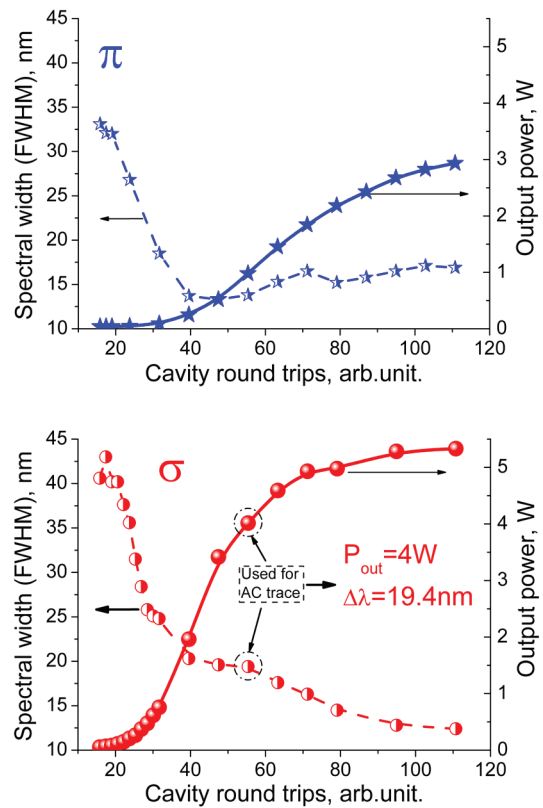


**Figure 8** – Intracavity pulse spectrum (a) and auto-correlation trace of RA seed pulses (b)

Seed pulses have spectral width of about 60 nm FWHM and about 7.5 ps duration while the incident pulse spectral width was about 12.5 nm. Fiber coupling efficiency was about 65 %. Seed pulse energy at the output of the fiber was about 4.5 nJ.

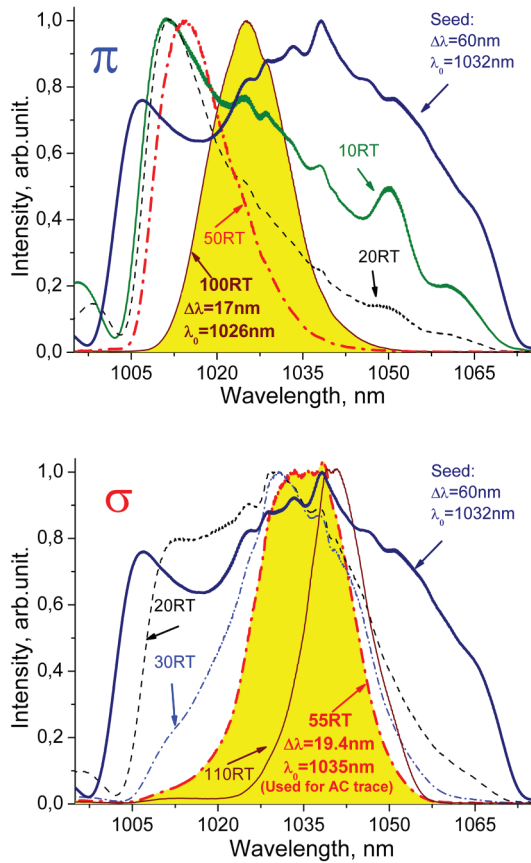
### Chirped pulse regenerative amplification experiment

During the RA experiment we measured the output pulse train parameters (spectral width and output power) for  $\pi$ - and  $\sigma$ -polarized light in the gain medium at 200 kHz PRF (Figure 9).



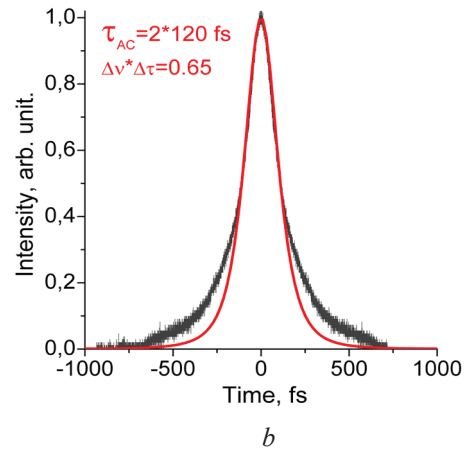
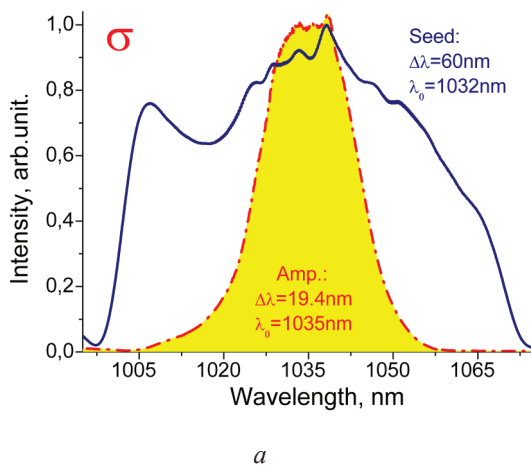
**Figure 9** – Dependencies of spectral width and average output power on the cavity roundtrips of Yb<sup>3+</sup>:CALYO chirped pulse RA

The maximum uncompressed average output power of 5.3 W (2.9 W) was obtained for  $\sigma$ - ( $\pi$ -) polarized light after 110 round trips (RT) of the pulse through the amplifier cavity, while the pulse spectral width (FWHM) decreased to 12.4 nm and 16.9 nm for  $\sigma$ - and  $\pi$ -polarization states, respectively, that demonstrated strong gain narrowing effect. The amplified pulse spectrum evolution versus RT number through the amplifier for  $\pi$ - and  $\sigma$ -polarizations are shown in Figure 10.



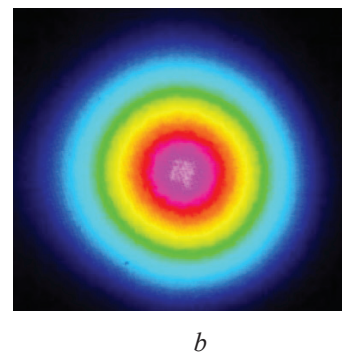
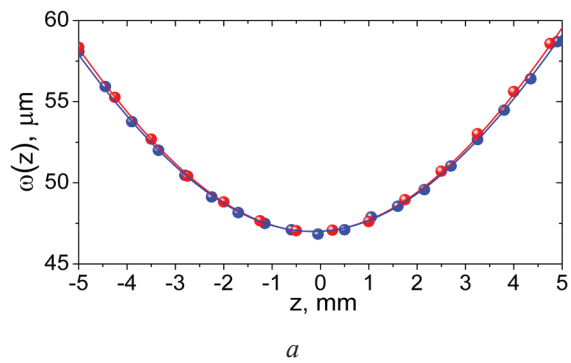
**Figure 10** – Evolution of the output pulse spectrum of Yb<sup>3+</sup>:CALYO chirped pulse RA

The data presented in the Figure 9 indicate that  $\sigma$ -polarization is more suitable for amplification of broad-band ultrashort pulses. Wide amplified spectrum of 19.4 nm (FWHM) was demonstrated after 55 RT with output power of about 4 W (almost 80 % of maximum value) (Figure 11). Corresponding autocorrelation (AC) trace of compressed pulses with average output power of 3 W is also shown in Figure 11. Measured pulse duration was about 120 fs assuming Lorentzian pulse shape (the best fit with AC trace).



**Figure 11** – Seed and amplified pulse spectra (a) and autocorrelation trace of compressed  $\sigma$ -polarized output pulse after 55 round trips (b)

The output beam had good quality with an  $M^2$ -factor of about 1.15. Measured output beam caustic and profile for Yb<sup>3+</sup>:CALYO,  $\sigma$ -polarized broad-band seeded chirped pulse RA are shown in Figure 12.



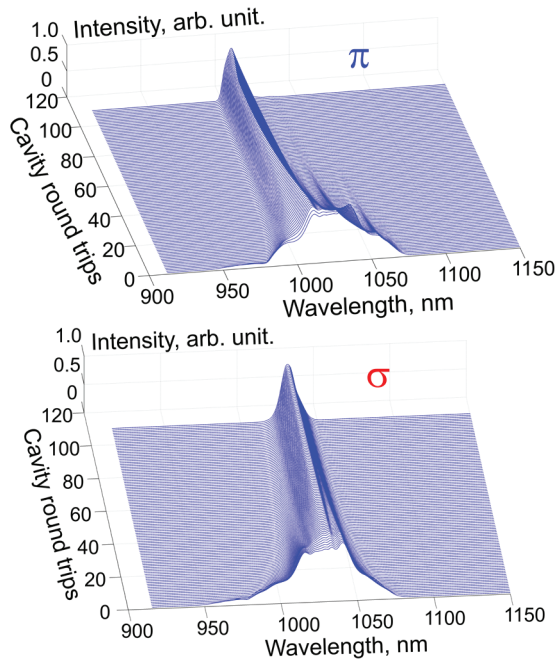
**Figure 12** – Output beam caustic (a) and profile (b)

The profile of the beam remains Gaussian up to the highest output powers.

### Mathematical modelling

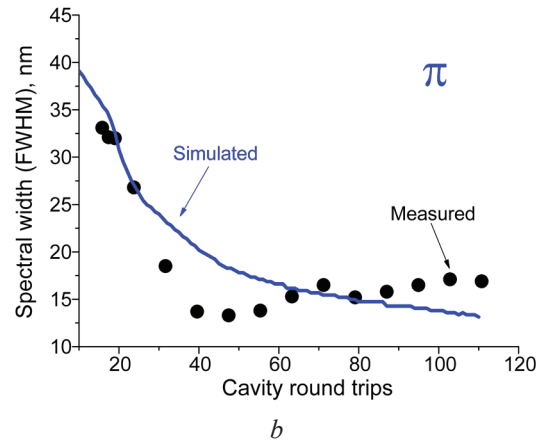
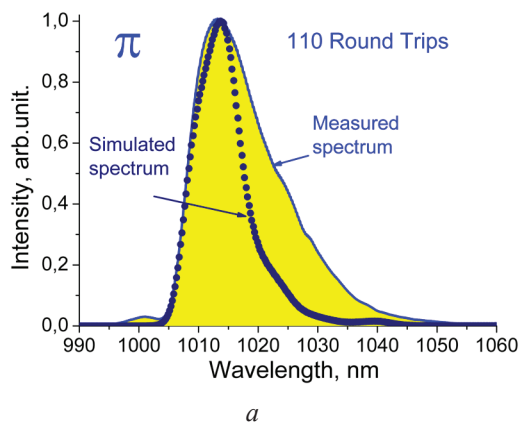
To estimate the amplified pulse spectra limited mostly by gain crystal characteristics and gain narrowing effect mathematical simulation was made

with constant level of intracavity losses ( $\approx 5\%$ ) for wide spectral range covering active crystal gain bandwidth. Simulation was based on the split-step Fourier method [23]. Gain curve was calculated by means of the absorption (ABS) and stimulated emission (SE) cross-section spectra under a certain population of the upper laser manifold of  $\text{Yb}^{3+}$  ions which corresponds to our experimental conditions. Simulation results are well agreed with experimental results. Amplified pulse spectrum evolution during the amplification in the  $\text{Yb}^{3+}:\text{CALYO}$  based chirped pulse RA is shown in Figure 13.



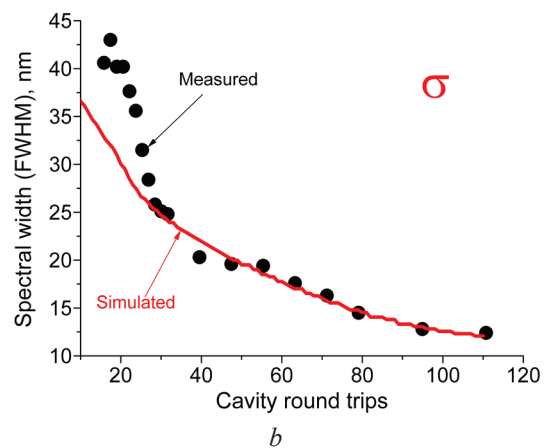
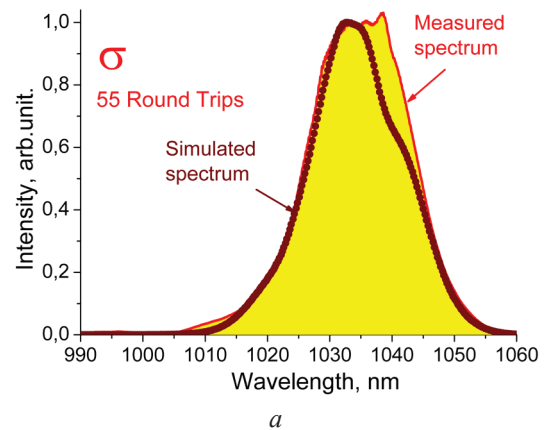
**Figure 13** – Evolution of the output pulse spectrum of  $\text{Yb}^{3+}:\text{CALYO}$  chirped pulse RA

Simulated and measured pulse spectra after 110 round trips and dependency of pulse spectral width on the number of cavity round trips for  $\text{Yb}^{3+}:\text{CALYO}$ ,  $\pi$ -polarized chirped pulse RA are shown in Figure 14.



**Figure 14** – Simulated and measured pulse spectra (a) and dependency of pulse spectral width on cavity roundtrip (b) for  $\text{Yb}^{3+}:\text{CALYO}$ ,  $\pi$  chirped pulse RA

Simulated and measured pulse spectra after 55 round trips and dependency of pulse spectral width on cavity round trips for  $\text{Yb}^{3+}:\text{CALYO}$ ,  $\sigma$ -polarized chirped pulse RA are shown in Figure 15.



**Figure 15** – Simulated and measured pulse spectra (a) and dependency of pulse spectral width on cavity roundtrip (b) for  $\text{Yb}^{3+}:\text{CALYO}$ ,  $\sigma$  chirped pulse RA

It is evident that simulation results are satisfactory agreed with experimental data, especially



for  $\sigma$ -polarisation. Thus, the usage of wide-band femtosecond seed pulses for Yb<sup>3+</sup>:CALYO chirped pulse RA makes it possible to shorten the duration of amplified pulses despite the negative contribution of gain narrowing effect.

## Conclusion

The results of experimental investigation of broad-band seeded Yb<sup>3+</sup>:CaYAlO<sub>4</sub>-based chirped pulse regenerative amplifier are reported for the first time to our knowledge. 120 fs-pulses (19.4 nm FWHM) with average output power of 3 W were demonstrated without any gain narrowing compensation technique. Despite the significant reduction of amplified pulse duration the task of improvement group velocity dispersion balance (including high orders of group velocity dispersion) remains relevant.

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