### VILNIUS UNIVERSITY INSTITUTE OF PHYSICS

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## INFRARED FEW-CYCLE PULSE OPTICAL PARAMETRIC AMPLIFIER

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VILNIAUS UNIVERSITETAS FIZIKOS INSTITUTAS

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# KELIŲ OPTINIŲ CIKLŲ TRUKMĖS IMPULSŲ PARAMETRINIS STIPRINIMAS INFRARAUDONOJOJE SRITYJE

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#### **Introduction**

Since the first demonstration of the laser in 1960, lasers have become a common tool for engineering and scientific applications. Optical pulses with ultra-short duration (<100 fs) and broad wavelength tunability are required for many modern studies of lightmatter interaction. Shortest pulse laser systems are based on Ti:sapphire oscillators with Kerr lens mode-locking and are now commercially available with sub-6 fs duration and energies about few nano-Joules. However, wavelength tunability of Kerr lens mode locked Ti:sapphire lasers is limited to a narrow range around the fundamental or around the second harmonic wavelength. Through the extreme temporal confinement of the light, peak power of tens  $GW/cm<sup>2</sup>$  can be obtained directly from such laser systems. Further intensity scaling is limited by optical damage and moderate nonlinear effects. To overcome these limits, an amplification scheme based on chirped pulse amplification. (CPA) (see Fig. 1) was proposed in 1985: a temporally stretched low-energy pulse is amplified in a laser medium and subsequently recompressed [1].



**Fig. 1.** Principle of chirped pulse amplification proposed by D. Strickland and G. Mourou [1]. A low energy initial short pulse is temporally stretched before amplification and recompressed after amplification to avoid high peak powers in the amplifier system.

One of the main drawbacks of this technique is that the population inversion in a laser medium leads to nonradiative processes and raise considerable thermal effects in the laser crystal. Another technique for ultra-short laser pulse amplification is optical parametric amplification (OPA) in nonlinear crystal. In contrast to amplification in laser media with population inversion, parametric amplification originates from three-wave mixing process inside crystal and does not involve energy storage in a material thereby reducing thermal load to a minimum. Several unique properties, like high gain per pass,

broad amplification bandwidth and high contrast ratio give optical parametric amplifiers a favorable edge over conventional laser amplifiers based on media with population inversion. The so called optical parametrical chirped pulse amplification (OPCPA) technique was proposed and demonstrated in 1992, which merged OPA and CPA techniques advantages. In 1997 I.N. Ross et al proposed the use of OPCPA system to amplify laser pulses to petawatt peak power levels [2]. This doctoral thesis is dedicated for high peak power few-cycle pulse generation and amplification in novel OPCPA systems.

#### **Main objective** is to:

Generate and amplify few-cycle pulses in the infrared region.

#### **Main tasks**:

- To investigate possibility of few-cycle pulses amplification at 800 nm in multi-beam-pumped OPCPA;
- To explore optimal beam configuration in multiple-beam-pumped OPCPA;
- To generate and amplify high peak power carrier-envelope-phase-stable pulses;
- To compress and characterize infrared pulses.

#### **Innovations of this work:**

In this work we experimentally demonstrated for the first time a noncollinear broadband multi-beam-pumped OPA @ 800 nm, based on type I BBO crystal. Amplified spectrum supports sub-12 fs duration, which is mainly limited by experimental condition (spatial chirp in bulk glass stretcher and used Ti:sapphire oscillator spectrum). Also the possibility to decrease parametric diffraction efficiency by increasing intersecting angles of pumps was demonstrated. The performed numerical calculations reveal that distribution of pump beams in  $\theta - \phi$ -plane allows to manipulate and broaden gain spectrum of non-collinear OPA.

New concept of hybrid type II OPA-OPCPA/filamentation system for generation of high energy carrier-envelope-phase (CEP)-stable few-cycle pulses  $@ 1.5 \mu m$  is proposed and demonstrated. The highest energy 12.5 mJ @ 1.5 µm before compression was generated in a four-stage OPCPA system. Also, the highest energy 4 mJ  $\omega$  1.5  $\mu$ m was measured after filamentation in argon gas with 5 bars pressure and highest energy 1.5 mJ @ 1.5 µm was measured in self-compression regime.

#### **Propositions to defend**:

- 1. Multi-beam  $\theta \phi$ -plane-pumped OPCPA, based on type I BBO crystal (pump 532 nm), enables efficient amplification of broadband few-cycle pulse.
- 2.  $\theta \phi$ -plane-pumped non-collinear OPA scheme enables to manipulate and broaden OPA gain spectrum.
- 3. Hybrid type II OPA-OPCPA/filamentation system enables generation of high energy (up to tens of mJ) carrier-envelope-phase-stable few optical cycle tunable pulses.
- 4. Type II (*eoo*) phase matching in KTP reduces amplitude-to-carrierenvelope-phase noise conversion due to pump/idler-to-signal cross phase modulation in OPA stages.

#### **Approbation**:

The results presented in this thesis are published in 10 international scientific papers: 6 of them (3, 5, 9, 12 and 13 in the publications list) are published in ISI-rated journals, 1 (10<sup>th</sup> in the publication list) in a book chapter and 4 (2, 7, 8 and 11 in the publication list) in other peer-reviewed articles. Also the results were presented in 18 (1- 9 and 13-21 in the conference list) international and 3 (10-12 in the conference list) national conferences.

**Author's publication list** (publications linked to this thesis are marked with the asterisk):

- 1. **S. Ališauskas**, R. Butkus, A. Piskarskas, K. Regelskis, V. Smilgevičius, *Modulation of spatial spectrum in tunable two crystal optical parametric generator*, Opt. Commun. **280**, 463-467 (2007).
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- 12\*. O. D. Mücke, D. Sidorov, P. Dombi, A. Pugžlys, **S. Ališauskas**, V. Smilgevičius, N. Forget, J. Pocius, L. Giniūnas, R. Danielius, and A. Baltuška, *10-mJ Optically Synchronized CEP-Stable Chirped Parametric Amplifier at 1.5 µm*, Opt. and Spect. **108**, 456-462 (2010).
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1. O. D. Mücke, D. Sidorov, P. Dombi, A. Pugžlys, A. Baltuška, S. Ališauskas, J. Pocius, L. Giniūnas, and R. Danielius, *Multimillijoule Optically Synchronized and Carrier-Envelope-Phase-Stable Chirped Parametric Amplification at 1.5 µm*, postdeadline talk MG1, *Advanced Solid-State Photonics*, Nara-Ken New Public Hall, Nara, Japan, January 27-30, 2008.

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- 3. O. D. Mücke, D. Sidorov, P. Dombi, A. Pugžlys, A. Baltuška, S. Ališauskas, J. Pocius, L. Giniūnas, and R. Danielius, *Multimillijoule Optically Synchronized and Carrier-Envelope-Phase-Stable Chirped Parametric Amplification at 1.5 µm*, talk WED4a.4, *16th International Conference on Ultrafast Phenomena*, Stresa (Lago Maggiore), Italy, June 9-13, 2008.
- 4. O. D. Mücke, D. Sidorov, P. Dombi, A. Pugžlys, S. Ališauskas, N. Forget, J. Pocius, L. Giniūnas, R. Danielius, and A. Baltuška, *10-mJ Optically Synchronized CEP-Stable Chirped Parametric Amplifier at 1.5 µm*, invited talk Sa20B(E)-1, *XII International Conference on Quantum Optics and Quantum Information (ICQO 2008)*, Vilnius, Lithuania, September 20-23, 2008.
- 5. O. D. Mücke, A. Pugžlys, P. Dombi, A. Baltuška, S. Ališauskas, N. Forget, J. Pocius, L. Giniūnas, and R. Danielius, *10-mJ Few-Cycle Chirped Pulse Parametric Amplification at 1.5 µm*, invited talk WR1, 21st IEEE/LEOS Annual *Meeting*, Newport Beach, CA, November 9-13, 2008.
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- 7. O. D. Mücke, A. J. Verhoef, A. Pugžlys, A. Baltuška, S. Ališauskas, V. Smilgevičius, J. Pocius, L. Giniūnas, R. Danielius, and N. Forget, *10-mJ Infrared Phase-Stable Parametric Amplification Based on a Femtosecond Yb-MOPA*, talk WD5, *Advanced Solid-State Photonics*, Denver, CO, February 1-4, 2009.
- 8. O. D. Mücke, A. J. Verhoef, A. Pugžlys, A. Baltuška, S. Ališauskas, V. Smilgevičius, J. Pocius, L. Giniūnas, R. Danielius, and N. Forget, *Infrared Multimillijoule Single-Filament Supercontinuum Generation*, talk JWD6, *CLEO*, Baltimore, Maryland, May 31-June 5, 2009.
- 9. O. D. Mücke, A. J. Verhoef, A. Fernandez, L. Zhu, A. Pugžlys, A. Baltuška, S. Ališauskas, V. Smilgevičius, J. Pocius, L. Giniūnas, R. Danielius, C.-H. Liu, K.- H. Liao, and A. Galvanauskas, *12-mJ IR OPCPA Based on Picosecond Nd Pump and Femtosecond Yb Seed Technologies*, invited talk Th3, *Middle Infrared Coherent Sources (MICS'2009)*, Trouville France, June 8-12, 2009.
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- 11. S. Ališauskas, V. Smilgevičius, A. Piskarskas, O. D. Mücke, A. J. Verhoef, A. Pugžlys, A. Baltuška, J. Pocius, L. Giniūnas, R. Danielius, and N. Forget, *12,5 mJ faziškai moduliuotų impulsų su stabilia nešančiojo dažnio faze parametrinis šviesos stiprintuvas 1,5 µm srityje*, *Lietuvos Nacionalinė Fizokos konferencija*, Vilnius, birželio 8-12, 2009.
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- 18. O. D. Mücke, S. Ališauskas, A. J. Verhoef, A. Pugžlys, V. Smilgevičius, J. Pocius, L. Giniūnas, R. Danielius, and A. Baltuška, *Efficient 4-Fold Self-Compression of 1.5-mJ Infrared Pulses to 19.8 fs*, talk, *Conference on Ultrafast and Nonlinear Optics UFNO'2009*, Burgas, Bulgaria, September 14-18, 2009.
- 19. S. Ališauskas, R. Butkus, V. Pyragaitė, V. Smilgevičius, A. Stabinis and A. Piskarskas, *Optocal Parametric Amplifier of Chirped Pulses Pumped by Two and Tree Beams*, poster, *Conference on Ultrafast and Nonlinear Optics UFNO'2009*, Burgas, Bulgaria, September 14-18, 2009.
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#### **Author contributions:**

The author contributed in all experiments and experimental measurements, preparation of experimental methodics and partly in numerical calculations, processing of experimental results, their interpretations and publishing.

#### **Contribution of coauthor's**:

**V. Smilgevičius<sup>1</sup>** formulated tasks for experiments, contributed in preparation of experimental methodics, interpretation of experimental results and publishing;

**A. Piskarskas<sup>1</sup>** initiated multi-beam pumped OPCPA experiment, contributed in interpretation of experimental results;

**R. Butkus<sup>1</sup>** jointly performed all multi-beam pumped OPCPA experiments, contributed in interpretation of experimental results and publishing;

**V. Pyragaite<sup>1</sup> and A. Stabinis<sup>1</sup>** performed theoretical analysis of parametrical diffraction effects in type I BBO crystal by multiple-beam pumping;

**A. Baltuška<sup>2</sup>** initiated 1.5 µm CEP-stable pulse generation and OPA/OPCPA experiments, performed stretcher-compressor calculations, contributed in interpretation of experimental results and publishing;

**O. D. Mücke<sup>2</sup>** jointly performed all 1.5 µm CEP-stable pulse generation and OPA/OPCPA experiments, contributed in interpretation of experimental results and publishing;

A. Pugžlys<sup>2</sup> partly performed 1.5 µm CEP-stable pulse generation and OPA/OPCPA experiments, contributed in interpretation of experimental results and publishing;

**D. Sidorov<sup>2</sup>** partly performed Yb:KGW oscillator and Yd:YAG regenerative amplifier optical synchronization;

A. J. Verhoef<sup>2</sup> partly performed 1.5 µm filametation experiments and contributed in interpretation of experimental results.

**J. Pocius<sup>3</sup> , L. Giniūnas<sup>3</sup> ir R. Danielius<sup>3</sup>** designed DPSS Yb:KGW MOPA.

**N. Forget<sup>4</sup>** designed AOPDF for higher order dispersion compensation of 1.5 µm pulses.

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#### **Thesis summary**

The thesis consists of 5 sections: introduction, two chapters, main results and conclusions, and a list of references.

In the **Introduction,** the main techniques for generation and amplification of ultra-short pulses are outlined, motivation for high peak power OPCPA development is revealed and main objective of this thesis, main tasks, innovations of this work, propositions to defend and approbation of results are presented.

In **Chapter 2,** experimental investigations of ultra-broadband two- or three-beampumped OPCPA at 800 nm are described. The two-stage OPCPA system is based on type I BBO (β-barium borate) crystals (pump  $\omega$  532 nm), where the 1<sup>st</sup> OPA stage (OPA I) was pumped by one beam and the  $2<sup>nd</sup>$  OPA stage (OPA II) was pumped by two or three pump beams derived from independent Nd:YAG laser amplifiers (see Fig. 2).



**Fig. 2.** Experimental setup. BS1–4 are beam-splitters; PCF, photonic crystal fiber; FI, Faraday isolator; SHG, second-harmonic generator; DL, delay line; BD, beam dumpers.

The femtosecond Ti:sapphire oscillator was used as a seed source for OPA and for a picosecond pump laser. The spectrum of Ti:sapphire oscillator extended from

670 nm to well beyond 900 nm. About one third of the femtosecond radiation was split off and coupled into ~70 cm long photonic crystal fiber (PCF) which had zero group velocity dispersion at 750 nm and was used as a frequency-shifter to produce 1064 nm radiation sufficient for seeding the Nd:YAG regenerative amplifier (RA) and ensuring optical synchronization between seed and pump lasers [3]. A weak seed pulse was amplified in a flashlamp-pumped regenerative amplifier RGA60 (Continuum Inc.) to ~9 mJ 30 ps pulses at a repetition rate of 30 Hz. The Faraday isolator (FI) was placed at the output of RA to prevent optical damage by a back-reflected beam. The beam splitter BS2 reflected half of the beam towards frequency doubling crystal and only up to 650  $\mu$ J of 532 nm radiation was used to minimize the parametric superfluorescence. The remaining transmitted part was split into three parts and sent to parallel Nd:YAG power amplifier's (PA's). Each PA contained a flashlamp pumped Nd:YAG rod (6x80 mm) and operated at the same repetition rate of 30 Hz. After double-passing and frequency doubling the amplified pulse energies at 532 nm were up to  $\sim$ 2 mJ, were collimated by three demagnifying telescopes and reflected towards the OPA II stage.

The other part of Ti:sapphire radiation was stretched to  $\sim$ 20 ps after doublepassing a 5 cm long bulk SF57 glass. The Pockels cell was used to extract a pulse that will be amplified from the whole pulse train and the temporal overlap of seed and pump pulses was accomplished by an optical delay line. In the OPA I stage, a 4 mm long BBO crystal (cut at  $\theta = 24$  deg) was used. The seed was focused by concave silver mirrors and the pump was focused by a lens (1 m focal length). The internal angle  $\alpha$  between pump and seed was about 2.3 degrees and could be varied to find the optimal spectral profile of the amplified signal. In the second OPA stage, a 5 mm long BBO crystal cut at the same angle was used, but interaction geometry was more complex.

The broadband phase matching can be achieved in a non-collinear interaction geometry [4]. The broadest phase matching and amplification gain is achievable for a signal-idler angle  $\Omega$  such that the signal group velocity is equal to the idler group velocity projected to the signal direction:

$$
v_{gi}\cos(\Omega) = v_{gs} \tag{1}
$$

Eq. (1) can be fulfilled in principal  $\theta$  or  $\phi$  crystal planes, but only up to two pump beams could be used to amplify the same broadband signal in those cases [5, 6]. In this thesis, it is demonstrated that distribution of pump beams in  $\theta - \phi$ -plane allows to manipulate and broaden OPA gain spectrum in non-collinear geometry. Theoretical calculation of  $\theta - \phi$ -plane-pumped OPA based on type I BBO (pump 532 nm) gain spectra and phase matching curves are depicted in Fig. 3. The pump intensity of 10 GW/cm<sup>2</sup> is limited by the crystal damage threshold. The gain bandwidth in  $\theta - \phi$ plane-pumped OPA increases in comparison to Gale configuration (black curves in Fig. 3). Modulation in gain spectra increases with decreasing noncollinearity angle  $\alpha$  or increasing angle  $\phi$ .



**Fig. 3.** Phase-matching curves and gain spectra of type I BBO based multiple beam-pumped OPCPA ( $\lambda_p = 532$  nm,  $I_p = 10$  GW/cm2,  $L = 4$  mm). The pump beams are arranged in  $\theta - \phi$ . plane at: (a)  $\phi = 0$  deg and  $\phi = 0.5$  deg, (b)  $\phi = 0.5$  deg and  $\phi = 1$  deg, (c)  $\phi = 0$  deg and  $\phi = 1$  deg.

The seed spectrum and amplified signal spectrum behind the OPA stages are depicted in Fig. 4a. The higher-frequency part of the seed spectrum was cut already in the pulse stretcher but the bandwidth of the remaining spectrum could be supported by the used BBO crystal effortlessly. The Fig. 4b summarizes spectral characteristics of the

second OPA stage. Apparently, if pumped by one of the three beams, spectra of amplified signal are similar but not identical and this indicates that phase-matching conditions were not fulfilled equally for all pump beams (see Fig. 3). The bandwidths of signal spectrum when pumped by separate pump beams separately were 87, 95, and 96 nm @ FWHM, respectively. If pumped by all three pump beams, the resultant bandwidth was 92 nm @ FWHM with transform-limited pulse duration of 11.4 fs.



**Fig. 4.** Spectral properties of two-stage OPA: (a) seed spectrum (green) and amplified signal spectrum behind the first (black) and second (red) OPA stage and (b) spectra of signal amplified separately by one pump beam and by all three beams in the second stage

In the first OPA stage, the seed was amplified from  $\sim$  1 nJ to 30  $\mu$ J and higher gain resulted in increased levels of parametrically generated superfluorescence which was effectively amplified in the second OPA stage altogether with the signal. The results of signal energetic characteristics in the second BBO crystal are depicted in Fig. 5. The total pump energy in front of the second OPA stage was 8.3 mJ with each of pump beams having roughly the same energy. If pumped by only one of the beams, the signal energy increased to 0.23-0.31 mJ and if pumped by any combination of two beams, the signal energy varied from 0.46 to 0.54 mJ. Finally, if pumped by all three beams, the signal was amplified up to 0.72 mJ and this corresponds to an efficiency of close to 9 %. However, if pumped separately by each of three beams and adding the energies of amplified signals, the sum energy would be 0.8 mJ (see Fig. 5). A 10% discrepancy in efficiency may be linked to the creation of additional beams under conditions of threebeam pumping. And the relatively low OPA efficiency in general is attributed to poor temporal pulse contrast and non-uniform spatial modes form different laser amplifiers.



**Fig. 5.** Dependence of amplified signal energy versus pump energy at the output of the second OPA stage on different number of pumping beams

The additionally generated beams in the second OPA pumped by all three beams possess a rather complex distribution so the interplay of interacting waves was studied in detail using only two pump beams for the sake of simplicity. Although it resembles the effect of self-diffraction [7]. The representative distribution of interacting beams in the far field is depicted in Fig. 6. Two pump beams were crossed at an angle (internal)  $\Delta \phi$ of 0.8 degrees in type I BBO crystal and spatial distribution of all prominent beams in the visible was registered. Besides two pump beams, signal beam and their newly appeared counterparts there can be seen many other beams which are parasitically generated second harmonic of signal and sum-frequencies of signal-idler spectral wings that are different from pump wavelength.

All additional waves could carry away up to 10% of total energy in this case. Obviously, high losses would seriously reduce advantages of such pumping method. However, it can be controlled by adjusting the angle between pump beams. At different pump intersecting angles we measured the energy of pump beams at which additional beams are not visible yet. Higher pump energy corresponds to lower losses introduced by additionally created beams at a given angle between pump beams. The measured

variation of this energy on angle between two pump beams is depicted in Fig. 7. By increasing the angle between pump beams the energy at which additional beams are not visible yet increases linearly, although at larger angles the efficiency of OPA decreases due to reduced interaction length and it may also be more difficult to maintain the bandwidth. We found an optimum external angle of about 1.5-2 degrees in our setup as a good compromise between efficiencies of creation of additional beams and signal amplification.



**Fig. 6.** Two-beam (P1 and P2) pumped OPA output in the far-field at  $\Delta \phi = 0.8$  deg. SFG, sumfrequency generation, SH, second harmonic.



**Fig. 7.** A variation of the energy of pump beams at which additional beams are not visible yet with external angle between the two pump beams

In **Chapter 3** the concept and realization of hybrid system based on type II KTP OPCPA and filamentation is described. For many applications in attosecond science, in particular for the generation of isolated attosecond XUV/soft-X-ray pulses, extremely short pulses comprising only one or two light oscillations underneath the field envelope are required. The well-established standard technology for the generation of few-cycle driver pulses based on Ti:sapphire amplifier systems is spectral broadening of mJ-level femtosecond pulses in noble gases. The energy throughput of gas-phase broadening schemes, such as the hollow-core fiber compressor [8] and filamentation [9], is limited to 4-5 mJ at 0.8 µm due to ionization losses. In this thesis, a CEP-stable 1.5 µm OPCPA system is demonstrated with pulse energies up to 12.5 mJ after four OPCPA stages based on a fusion of a DPSS femtosecond Yb:KGW MOPA and picosecond Nd:YAG solidstate technology. By employing more narrowband type II phase matching, one can optimize the spectral brightness of the seed at the expense of the seed energy, achieve a more uniform saturation across the pulse spectrum, and minimize energy backconversion into the pump. Furthermore, self-compresion of CEP-stable 2.2 mJ, 74.4 fs, 1.57 µm input pulses is demonstrated down to 19.8 fs duration in a single filament in argon gas at 5 bar pressure with 1.5 mJ output energy. We foresee that our TW-peak power sub-4 cycle pulse will open the door to exciting new experiments in attosecond high-field science in the near future.

In our IR OPCPA scheme (see Fig. 8), both Yb:KGW and Nd:YAG regenerative amplifiers (RA) are simultaneously directly seeded from a single Yb:KGW master oscillator. The repetition rate of the Yb:KGW diode-pumped solid state MOPA (Pharos, Light Conversion, Ltd.) was set at  $10 \text{ kHz}$  as the  $500^{\text{th}}$  harmonic of the flash-lamppumped Nd:YAG amplifier (Ekspla Ltd.) operating at 20 Hz. The 1030 nm output from the femtosecond Yb:KGW MOPA is first split into two parts by means of a variable beam splitter: part one is used for implementing the first OPA stage, part two is used for pumping the second OPA stage. In the first OPA stage, the 1030 nm pulses are first frequency-doubled in a 1 mm thick type I BBO crystal. Typically 8.5 µJ of 515 nm pulses are again split by a variable beam splitter into two parts: 1.3 µJ are focused onto a 10 mm thick sapphire plate to generate a white-light continuum in a single filament. The with-light continuum is recollimated and used to seed the first OPA stage. The whitelight seed pulses and the 515 nm pump (second harmonic of Yb:KGW MOPA) pulses

are combined collinearly to avoid idler angular dispersion. In particular, selecting the ~785 nm wavelength for amplification, this configuration produces CEP-stable idler pulses [10] at 1.5 µm that we use as a seed in the second OPA stage.

Following the pioneering work of Kraemer et al. [11, 12], we employ Type II KTP crystals (1030/1064 nm pump,  $\sim$ 1.5 µm signal,  $\sim$ 3.5 µm idler) for the subsequent OPA stages 2-4 because these crystals (unlike borate crystals) are transparent for the mid-IR idler wavelength and exhibit a relatively broad bandwidth around 1.5 µm. The CEP-stable idler pulses from the first OPA stage are recollimated and focused onto a 6 mm thick type II KTP crystal (cut at  $\theta = 45.5$  deg,  $\varphi = 0$  deg). The pump beam is focused onto the same KTP crystal under an external walk-off compensation angle of 2.2 deg with respect to the seed beam which was amplified up to 10  $\mu$ J.



**Fig. 8.** Scheme of the OPCPA power-amplification stages 3 and 4: G, reflection grating; CM, curved mirror; PM, plane mirror; TM/BM, top/bottom mirror; AOPDF, acousto-optic programmable dispersive filter; RA, regenerative amplifier; PA, double-pass post amplifier; f, f3, f4, f5, lens focal lengths; T1, T2, telescopes; A, aperture; W1/W2, input/output windows; BP, beam profiler.

In the subsequent booster-amplification stages 3 and 4 shown in Fig. 8, the 1.5- $\mu$ m signal pulses are amplified from the 10  $\mu$ J level to energies >10 mJ before recompression. The CEP-stable 1.5 µm pulses from the front-end are temporally stretched to ~40 ps using a grating-based stretcher and an IR DAZZLER in order to optimize energy extraction from the 60-ps long Nd:YAG pump pulses. To guarantee a homogeneous pump profile free of hot spots, we relay-image the 10-mm-diameter crystal rod in the Nd:YAG power amplifier onto the 10-mm-thick KTP crystals in stages 3 and 4. We obtain a pump diameter of 2 mm for stage 3 and 3.1 mm for stage 4. Relayimaging is achieved with three lenses with focal lengths of  $f = 75$  cm,  $f = 10$  cm, and  $f4 = 35$  cm. The 1.5 um (seed) pulses are focused onto the third-stage KTP crystal and imaged onto the fourth-stage KTP crystal with telescope T1. With this pumping geometry and 90 mJ pump pulses, we have achieved up to 12.5 mJ signal pulses centered at 1.57  $\mu$ m and pump-signal conversion efficiency of  $\sim$ 14% in the final OPCPA stage. To avoid damage to the gold-coated gratings in the OPCPA compressor, we expand the  $1/e<sup>2</sup>$  beam diameter of the fourth-stage output by a factor of 5 to 9.5 mm by means of a Galilean beam expander T2.



**Fig. 9.** Spectral properties of the power-amplification OPCPA stages: (a) measured spectrum of the third stage seed (black doted line) and amplified signal spectra after stages 3 (red line) and 4 (green line), (b) the same data in logarithmic scale, (c) recalculated idler spectrum, the blue line indicates the idler transmission thru 1 cm of KTP. The amount of superfluorescence is immeasurable in absence of the white-light seed in OPA stage 1.

The spectra of the seed and amplified signal pulses of the power-amplification stages are shown in Fig. 9. As idler absorption increases above 3.45 µm in KTP, we can achieve higher output powers when tuning the signal center wavelength above 1.55  $\mu$ m. The SHG-FROG characterization data of 3.5 mJ 1.57  $\mu$ m pulses with 62 nm bandwidth from the 20 Hz four-stage IR OPCPA (see Fig. 10) indicate a 74.4 fs @ FWHM pulse duration, close to the transform-limit of 72.6 fs.



**Fig. 10.** SHG-FROG characterization of the 20 Hz output from the four-stage IR OPCPA: (a) measured and (b) retrieved FROG traces, (c) measured spectrum (back curve), retrieved spectral intensity (blue) and phase (red), (d) retrieved temporal intensity (blue) and phase (red) profiles exhibiting a 74.4 fs FWHM pulse duration. The transform-limited intensity profile (dashed) corresponds to a 72.6 fs duration.



**Fig. 11.** SHG-FROG characterization of 2.1 mJ filamentation output pulses for argon at 4 bar: (a) Measured and (b) retrieved FROG traces. (c) Measured spectrum (black curve), retrieved spectral intensity (blue) and phase (red). (d) Retrieved temporal intensity (blue) and phase (red) profiles. The TL intensity profile (dashed) corresponds to a 14.6 fs duration.



**Fig. 12.** Self-compression of 1.5 mJ pulses in argon at 5 bar: (a) measured and (b) retrieved FROG traces, (c) measured spectrum (black curve), retrieved spectral intensity (blue) and phase (red). (d) Retrieved temporal intensity (blue) and phase (red) profiles indicating a 19.8 fs pulse duration. The transform-limited intensity profile (dashed) corresponds to a 15.9 fs duration.

In the filamentation regime without plasma-induced pulse self-recompression, we generated up to 4 mJ 600 nm-wide IR supercontinum supporting 8 fs pulse durations which less than two optical cycles at 1.5  $\mu$ m. SHG-FROG data of 2.1 mJ spectrally broadened pulses are displayed in Fig. 11 and reveal a rather complex spectro-temporal structure. The observed strong nonlinear phase leads to a temporal break-up into two peaks of 20 fs and 15 fs separated by 60 fs. Since a clean single-filament spatial profile was observed simultaneously, we conclude that the temporal splitting helps to keep the pulse intensity below the break-up threshold of a single filament.

By lowering the input pulse energy and tuning the gas pressure in the cell, we achieved the regime of pulse self-compression. In the experiment, CEP-stable 2.2 mJ 74.4 fs 1.57 µm input pulses are compressed in a single filament in argon down to a 19.8 fs duration (see Fig. 12). This represents a temporal compression of the input pulses by a factor of ~4. The output energy was 1.5 mJ, corresponding to the energy throughput of 66%. In addition, the spectral phase was shown to be remarkably reproducible on a daily basis which holds potential for further recompression using fixed-dispersion chirped mirrors.

#### **Conclusions**

- $\theta \phi$ -plane-pumped (pump 532 nm) OPA based on type I BBO enable amplify few-cycle broadband pulses at 800 nm.
- Separate two-beam-pumped OPA gain spectra can bee the same for "plus"  $\phi$ and "minus"  $\phi$  pump-planes. In special case, for nonsymmetrical  $\theta - \phi$ plane-pumped geometry, OPA extend gain spectra.
- Parametrical diffraction can be controlled by adjusting the angle between pump beams. By increasing the angle between pump beams (0.5-3 deg) the threshold increases linearly, although at larger angles the efficiency of OPA decreases due to reduced interaction length. Optimum external angle of about 1.5-2 degrees in our setup is a good compromise between efficiencies of creation of additional beams and signal amplification.
- Hybrid type II KTP OPCPA (at 1.5 µm)/filamentation approach enable to generate few-cycle high peak power infrared pulses.
- A more narrowband type II (*oo-e*) phase matching in KTP can optimize the spectral brightness of the seed at the expense of the seed energy, and reduce amplitude-to-CEP noise conversion due pump/idler-to-signal cross phase modulation in OPA stages.
- The filamentation regime involving plasma-induced self-compression is possible for sub-100 fs pulses @ 1.5 µm.

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#### **Santrauka**

### Kelių optinių ciklų trukmės impulsų parametrinis stiprinimas infraraudonoje srityje

Disertacijoje nagrinėjama tematika susijusi su kelių optinių ciklų trukmės impulsų formavimu bei parametriniu stiprinimu infraraudonoje srityje. Šioje disertacijoje buvo tirtos dvi moduliuotosios fazės ("čirpuotų") impulsų parametrinio stiprinimo sistemos, stiprinančios ypač plataus spektro impulsus 800 nm bei 1,5 µm srityse.

Disertaciją sudaro 5 dalys: įvadas, du parametrinio stiprinimo sistemas aprašantys skyriai, pagrindiniai rezultatai ir išvados bei cituojamos literatūros sąrašas.

**Įvade** yra aprašomi pagrindiniai ultratrumpų impulsų formavimo bei stiprinimo metodai. Trumpai pristatoma darbo motyvacija, darbo tikslas bei užduotys. Pristatomas darbo naujumas, ginamieji teiginiai bei rezultatų aprobacija.

**Antrajame skyriuje** pristatomi keliais pluoštais kaupinamo plataus spektro signalo parametrinio stiprinimo rezultatai. Darbe pirmą kartą pademonstruotas plataus spektro impulsų 800 nm srityje parametrinis stiprinimas kaupinant dviem ir trimis nepriklausomų lazerinių šaltinių pluoštais (kaupinimų išdėstymas  $\theta - \phi$ -plokštumoje). Kaupinant trimis pluoštais išdėstytais  $\theta - \phi$ -plokštumoje I-tipo BBO parametriniame stiprintuve buvo sustiprinti 92 nm spektro pločio impulsai. Sustiprinti impulsai palaiko sub-12 fs trukmes, t.y. <5 optinių ciklų. Kaupinamame keliais pluoštais parametriniame stiprintuve stebimas naujų erdvinių komponentų atsiradimas, kuriuos sąlygoja parametrinė pakopinė difrakcija (pakopiniai  $\chi^{(2)}$  procesai), o tai mažina bendrą sistemos energetinio keitimo efektyvumą.. Darbo metu pademonstruota galimybė mažinti parametrinės difrakcijos sąlygotus nuostolius parametriniame stiprintuve derinant kampus tarp kaupinimo pluoštų. Taip pat teoriškai numatyta galimybė praplėsti parametrinio stiprinimo kontūrą kaupinimo pluoštus išdėstant  $\theta - \phi$ -plokštumoje.

**Trečiajame skyriuje** pristatomas alternatyvus būdas formuoti didelės energijos (kelių dešimčių milidžaulių) kelių optinių ciklų trukmės stabilios fazės impulsus 1,5 µm srityje. Būdas paremtas sąlyginai siauro spektro 1,5 µm srityje stiprinimu II-tipo KTP kristale (kaupinama ~1 µm spinduliuote) bei spektro plėtra inertinėse dujose po stiprinimo. Keturių pakopų parametriniame stiprintuve pasiekta iki šiol didžiausia 12,5 mJ impulso energija 1,5 µm srityje prieš kompresiją. Impulsą pavyko suspausti iki artimai spektriškai ribotos trukmės – 74,4 fs (spektriškai ribotas impulsas atitrinka 72,6 fs). Maksimali impulso energija po dviejų atspindžio gardelių neigiamos dispersijos kompresoriaus siekė 6 mJ. Norint pasiekti kelių optinių ciklų trukmes, sustiprinto impulso spektras buvo plečiamas pavienėje gijoje argono dujose. Iki 4 mJ išvadinės energijos gijos formavimosi metu išplitusio spektro impulsas atitinka 8 fs (1,6 optinio ciklo 1,5 µm spinduliuotei) spektriškai ribotą impulsą. Taip pat rastos sąlygos kuomet impulsas patiria iki 4 kartų savispūdą gijos formavimosi metu. Savispūdos režime iki 1,5 mJ išvadinės energijos impulsai susispaudė iki 19,8 fs (<5 optinių ciklų). Suformuoti impulsai yra tinkami pavienių atosekundinės trukmės impulsų generacijai aukštesnės eilės harmonikų generacijos metu.

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