Optimization of high peak power few-cycle optical parametric chirped pulse amplifier systems

Thesis points of the PhD dissertation

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Contents

1 Introduction

Optics, the behaviour of light as it propagates through matter, has been studied since the era of Euclid. Fundamental laws, such as rectilinear propagation, reflection, refraction, dispersion could be established simply by using the Sun as a light source. The average intensity of sunlight on the Earth's surface is roughly $0.14\,\mathrm{W\,cm^{-2}}$, at which level materials provide linear response. Compared to this, the first operational laser, demonstrated in 1960 [1], could reach an intensity level of a few-$\mathrm{MW\,cm^{-2}}$. Laser light, having this level of intensity, can induce nonlinear material response during propagation. Consequently, the first nonlinear phenomenon, second-harmonic generation (SHG) [2], was observed already in 1961, a year after the invention of the laser.

During nonlinear processes, such as second-harmonic generation, there is no real energy level involved, which is denoted by the word parametric [3]. Due to the virtual energy levels, these processes are instantaneous on the femtosecond to nanosecond time scale, where such processes are studied and utilized.

The possibility of light amplification by utilizing nonlinear material response was first successfully demonstrated by Wang and Racette in 1965 [4]. This technique is called optical parametric amplification (OPA). During OPA, there is an instantaneous energy transfer between the pump and signal waves. The mechanism of the process is that a pump photon splits into two lower energy photons, a signal and an idler. Therefore, the number of signal photons increases while the energy difference of the pump and signal photons is taken away by the idler photon.

Within a few years, as a result of the rapid progress in laser development, the intensity reached a level where the refractive index of the materials became intensity dependent. The intensity dependent refractive index causes self-phase-modulation (SPM) and self-focusing in the temporal and spatial domains, and ultimately beam break-up as a consequence of these two effects. All these nonlinear effects prevented the further scaling of pulse intensity for almost a decade, until the concept of chirped pulse amplification (CPA) was proposed [5]. In convectional CPA systems the amplifier medium is a laser active material, for example Ti:sapphire. It was soon realized that instead of laser materials, parametric crystals can be also utilized in CPA systems. This is called optical parametric chirped pulse amplification (OPCPA).

OPCPA has many beneficial properties over Ti:sapphire based CPA systems. For example, OPCPA gain bandwidth supports the amplification of sub-10 fs pulses, while gain narrowing in Ti:sapphire CPA, in the absence of special techniques, limits pulse duration to the 40 fs to 50 fs range. Parametric super-fluorescence (PSF) in case of OPCPA is confined to the temporal window of the pump pulse, while in Ti:sapphire the amplified spontaneous emission (ASE) is determined by the upper state lifetime of the laser material. Therefore, if the pump and signal durations are matched, OPCPA provides better contrast than CPA. Probably, the biggest advantage of OPCPA over conventional CPA is that during OPA the energy difference of pump and signal photons is taken away by the idler photon, while in laser amplification this energy difference is dissipated as heat. This allows OPCPA operation at much higher average power than conventional CPA.

On the other hand, OPCPA requires more demanding conditions than CPA. Due to the instantaneous nature of OPA, pump and signal pulses have to be precisely synchronised. Furthermore, the properties of the amplified signal are highly sensitive on the pump pulse quality, which challenges pump technology. Additionally to these, the pump-to-signal conversion efficiency in OPCPA is usually 10 % to 25 % [6], while in Ti:sapphire based CPA systems, 50 % can be reached with minor efforts [7]. This is one of the main reasons why most of the PW-class systems are based on CPA and not OPCPA.

One main driving force behind OPCPA development is that the technique can directly deliver pulses as short as a few oscillation cycle of the electric field, together with high peak and average power. Such pulses are widely used in many scientific applications, which aim to explore ultrafast physical processes. For example, few-cycle, mJ-level pulses can be used to generate protons and ions from thin foils and accelerate them to few MeV energies [8]. Recently, 1 fs, MeV electron beam was demonstrated which was driven by 3.4 fs multi-mJ pulses [9]. Sub-10 fs pulses are readily used in time-resolved spectroscopy as well [10], as they can provide high temporal resolution. Beside all these applications, isolated attosecond pulse generation [11] is probably one of the greatest motivator of the development of high peak power, carrier-envelop-phase (CEP) stabilised few-cycle sources.

Another beneficial property of OPCPA is that the gain spectrum can be tuned over the whole transparency range of the nonlinear crystals, while in laser materials the tuning range is restricted to their emission spectrum. This is particularly useful when the goal is the generation of ultrashort pulses in the mid-IR spectral range. It was recognized that many strong field physical and spectroscopic experiments benefit from long wavelength driving fields, which induced a fast evolution in the territory of mid-IR OPCPA systems. By the utilization of this wavelength range the cutoff photon energy during high-harmonic-generation (HHG) can be extended to the keV range and allows the generation of the shortest attosecond pulses [12]. Electron acceleration to MeV energies using mid-IR pulses was also recently demonstrated [13]. The rotational and vibration transitions of many molecules are located in the mid-IR spectral range, consequently these sources have huge importance in trace gas detection, examination of biomedical samples and breath analysis [14].

2 Objectives

All the aforementioned scientific applications call for high peak and average power, ultrashort pulses in the visible, near-IR and mid-IR spectral ranges. Due to the previously described advantageous properties, OPCPA is probably the most suitable amplification technique for this, therefore OPCPA development was the main motivation of my work.

2.1 First objective

The OPCPA concept has many advantages compared to conventional, laser amplifier based CPA systems. Yet, PW-scale systems are rather based on conventional CPA, mainly due to the two-fold increase in conversion efficiency provided by Ti:sapphire.

Therefore, my first aim was to examine the idea of a special OPA arrangement which could potentially increase conversion efficiency.

2.2 Second objective

The production of pulses as short as a single oscillation cycle under the field envelope is mainly motivated by isolated attosecond pulse generation. Such short pulses can be generated by using post-compression techniques, however, currently they are limited to a few-mJ energy level. On the other hand, few-tens-mJ, three-cycle pulses are directly accessible form OPCPA systems. Therefore, the other way of reaching single-cycle duration is to find a method to increase the gain bandwidth of OPCPA.

Consequently, my second aim was to examine the properties of a few broadband OPCPA configurations which could potentially broaden the gain spectrum.

2.3 Third objective

The first development phase of the ELI-ALPS Single Cycle Laser (SYLOS 1) was the first 1 kHz repetition rate TW-class OPCPA system, delivering CEP-stabilized, three-cycle pulses [15]. During the second development phase, called SYLOS 2, the aim was to shorten the pulse duration close to 2-cycle, while keeping the peak power at the same level.

Therefore, my third aim was to determine the optimal OPCPA configuration during the upgrade of the Single-Cycle Laser (SYLOS 2) laser in ELI-ALPS.

2.4 Fourth objective

In the past few years, OPCPA systems operating in the mid-IR have rapidly proliferated due to the recognition that many strong field physical experiments can benefit from the long wavelength driving pulses. CEP-stabilized, mid-IR pulses are generated as the difference frequency of the pump and signal pulses. There are two options to increase the energy of the mid-IR pulses: the first is to amplify the idler pulse after DFG; the second is to amplify the signal prior DFG. During my work, I named these two scenarios as ”idler scheme” and ”signal scheme”, respectively. According to the mid-IR systems reported so far, the two schemes are utilized approximately equally, without reasoning about the chosen amplification scheme. In laboratory conditions it is not straightforward, and in some cases it is even impossible, to switch from one scheme to the other under the same experimental conditions.

Therefore, my aim was to optimize a mid-IR OPCPA in case of both schemes and provide an answer to the previous question.

3 Methods and tools

OPCPA systems can be developed based on empirical observations and simple analytical calculations, however the expenses of research and development can be reduced by first performing an extensive numerical examination. During simulations many parameters, such as crystal thickness, seed pulse duration and beam diameter, can be continuously varied without additional costs and time which is needed for the rearrangement of an experimental setup.

Numerical codes which take into account every feature of OPCPA are very complex [16]. Furthermore, the computational requirements are exponentially increasing in case of large beam sizes and longer than 10 ps pulse durations. However, even with 2D models, which take into account the temporal shape of the pulse and the interaction distance (t, z) [17] or those which neglect some features, for example dispersion [18], can provide useful information. The effectiveness of 4D models (x, y, t, z) during the design of OPCPA systems operating in the few picosecond temporal range, was proven by many authors [19]. In addition, these numerical codes can provide insight into the spatiotemporal shape of the amplified signal pulse, which is possible, but not straightforward to measure.

The tool during my work was a 4D numerical code for the modelling of OPCPA. It was developed by Andrianov et al. and utilizes a special algorithm for highly chirped pulses [20]. Due to this algorithm the computational requirements are highly reduced, thus enabling the numerical simulation of OPCPA systems operating in the $\geq$ 10 ps range without approximations. In this work the 4D numerical simulation results of broadband OPCPAs operating in the 100 ps and 1 ns range are performed. To the best of my knowledge, in this range full featured simulations are not presented in the literature so far.

During my work I was using a server equipped with 128GB of RAM memory and an Intel Xeon E5-1650 CPU. In order to control the OPCPA code I developed a Python script, which enabled to do a series of automatized execution with varied input parameters. I also developed numerical algorithms in Python which was used to simulate linear pulse propagation between individual OPCPA stages. These algorithms take into account nonparaxial spatial phase shift of spherical mirrors and free-space propagation. This way whole OPCPA systems could be numerically simulated in the most realistic way.

4 New scientific results

T1 I have numerically examined the properties of the cascaded-extraction optical parametric amplifier (CE-OPA) design and revealed that CE-OPA increases conversion efficiency by at least 10%, without deteriorating the spatiotemporal shape of the amplified signal[T1].

T2 I have shown that double-BBO configuration introduces spatiotemporal couplings if the two following two conditions are satisfied [T2]:

1. The lateral pump displacement in noncollinear geometry is comparable to the size of the interacting beams.
2. The intensity gain is high, thus the pump guides the signal pulse during amplification.

T3 I have modelled and determined the optimal configuration of an OPCPA system to provide 2.2-cycle, TW class pulses [T2].

T4 I have numerically optimized the performance of two CEP-stabilized mid-IR systems, which are different in the order of amplification (OPCPA) and frequency conversion (DFG) stages and revealed that the overall conversion efficiency is slightly better and the peak power of the idler is higher if DFG is placed before OPCPA, while CEP-stability is somewhat better and the compressed pulses are shorter if OPCPA is done prior DFG [T3]. Due to the small differences, the applicable scheme could be decided by an overall cost-benefit analysis.

T5 I revealed, to my knowledge for the first time, that the interplay of pump depletion and the shortening of stretched pulses during amplification results in gain narrowing and due to this lower peak power can be achieved at the output of the OPCPA system [T3].

Own publications related to the thesis

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[CO2] K. Osvay, A. Börzsönyi, H. Cao, E. Cormier, J. Csontos, P. Jójárt, M. Kalashnikov, B. Kiss, R. Lopez-Martens, **Sz. Toth**, and Z. Varallyay, "Development status and operation experiences of the few cycle high average power lasers of ELI-ALPS (Conference Presentation)," in *Short-pulse High-energy Lasers and Ultrafast Optical Technologies*, International Society for Optics and Photonics, vol. 11034, SPIE, 2019. DOI: 10.1117/12.2523057.

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