High Average Power Near-Infrared Few-Cycle Lasers

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Ultra-short laser pulses with only a few optical cycles duration have gained increasing importance during the recent decade and are currently employed in many laboratories worldwide. In addition, modern laser technology nowadays can provide few-cycle pulses at very high average power which advances established studies and opens exciting novel research opportunities. In this paper, the two complementary approaches for providing few-cycle pulses at high average power, namely optical parametric amplification and nonlinear pulse compression, are reviewed and compared. In addition, their limitations and future scaling potential are discussed. Furthermore, selected applications particularly taking advantage of the high average power and high repetition rate are presented.

1. Introduction

Ultra-short laser pulses with few-cycle durations have first become available in 1985 from dye laser oscillators. Only a few years later a true revolution in ultra-fast science began: Ti:Sa oscillators, which nowadays provide the shortest pulses with only 5 fs duration (one optical cycle lasts 2.7 fs at 800 nm wavelength) directly from the oscillator,[2] quickly replaced the ultra-fast dye lasers. Ti:Sa based amplifiers could deliver high-peak-power ~ 20 to 30 fs pulses with a few tens of millijoules of pulse energy at ~1 kHz repetition rate,[3] and spectral broadening in gas-filled capillaries followed by subsequent compression with chirped mirrors could shorten them back to sub-two-cycle durations.[3] These lasers have been the workhorse for ultra-fast science for more than two decades and enabled seminal achievements such as observing and controlling electron dynamics in atoms, molecules, ions and solids,[6,7] high harmonic generation,[8,9] laser particle acceleration[10] and ultra-fast material ablation.[11] In addition, stabilization of the carrier envelope phase (CEP) enabled precise control of the electrical field of the few-cycle pulse in such a way that field-sensitive ultra-fast effects could be studied. The combination of few-cycle laser pulses and CEP control particularly enables the generation of isolated attosecond pulses which themselves opened a completely new research field called attoscience.[12]

With the exploration of innovative solid-state laser concepts femtosecond laser with much higher average powers as compared to Ti:Sa amplifiers have become available. Ytterbium-based Fiber,[13,14] Slab,[15] and Thin-disk lasers[16] can nowadays provide ultrashort pulses with average powers up to ~1 kW – more than two orders higher than that of typical Ti:Sa lasers.[17] This increase in output power is possible due to the lower quantum defect of Yb (typically <10 %) compared to Ti:Sa (> 30 %) and the improved heat dissipation within the laser medium. Fiber, thin-disk and slab provide a much larger surface-to-volume ratio compared to a typical rod geometry and thus allow for efficient cooling. Moreover the waveguide nature of a fiber makes the propagating light insensitive to thermo-optical distortions, a thin-disk causes purely longitudinal heat flow along the propagation direction of the laser beam and therefore minimizes transversal gradients, while a zig-zag beam path through a slab can cancel out most of the thermo-optical wave-front distortions. Thus, thermo-optical distortions are minimized in all three cases by the geometry of the gain medium. In addition, pumping of Yb-doped laser materials is directly possible with high average power laser diodes.

The concept of coherent combination of multiple lasers or amplifiers allows for further average power and pulse energy scaling simply by increasing the number of amplifiers.[18–22] These modern femtosecond lasers have come of age and are described in a greater detail in the following articles.[13–16,20,21,23–29] However, due to the limited gain bandwidth of Yb, the pulse duration of these systems are typically limited to ~1 ps for Yb:YAG down to ~100 fs for Yb:glass lasers.

Obtaining few-cycle pulses from these modern femtosecond lasers thus requires efficient pulse shortening techniques. To do so, either the pulses can be used to pump an ultra-broadband optical parametric amplifier (OPA) to transfer their energy to few-cycle pulses via optical parametric amplification[30] or nonlinear compression based on spectral broadening and subsequent compression can be exploited.[31] In this paper both methods will be presented, discussed and compared.
Note that this paper is focusing particularly on high average power few-cycle lasers in the near-infrared spectral region (0.8 μm to 1 μm central wavelength). High average power few-cycle lasers in the short wave infrared and middle-infrared (>2 μm wavelength) will be presented in a second review article following the present one.

The manuscript is organized as follows: Section 2 is devoted to optical parametric amplification. After a brief introduction the important aspects to achieve large gain bandwidths, to control the spectral phase, to obtain a broadband signal and pump signal synchronization will be discussed. This is followed by an overview of state-of-the-art high average power optical parametric amplifiers. Particular emphasis is put on the generation of sub-2-cycle pulses and the control of their carrier-envelope phase. Furthermore, limiting thermal effects and potential mitigation strategies are outlined. Section 3 is devoted to nonlinear compression to few-cycle durations. After introducing the basic concept, design criteria will be evaluated and limiting effects as well as their mitigation will be discussed. Finally, the state-of-the-art will be presented and future scaling possibilities are outlined. Section 4 presents a number of selected applications that have been enabled by these novel high average power few-cycle lasers.

Finally, a summary and an outlook on future developments and applications will be given.

2. Optical Parametric Amplification of Few-Cycle Pulses

Optical parametric amplifiers (OPAs) have attracted huge interest due to their special properties. In contrast to conventional laser amplifiers the gain is not related to ‘real’ transitions within the laser medium. Thus, the spectral region and bandwidth of amplification can not only be freely designed by phase matching, but there is also no energy stored in the OPA crystal. As such, OPAs represent an ideal choice for amplification of the broad bandwidth of few-cycle pulses to high average powers. The concept of optical parametric chirped pulse amplification (OPCPA) additionally allows for an efficient energy transfer from pump to signal since optical parametric amplification is an instantaneous nonlinear effect. The amplification bandwidth of the OPA system depends mainly on the phase-matching conditions. It has been shown that the performance of OPCPA additionally allows for an efficient energy transfer from pump to signal since optical parametric amplification (OPCPA) scheme ensures efficient energy transfer from pump to signal even when the process is non-linear.

2.1. General Layout of Few-Cycle Optical Parametric Amplifiers

Optical parametric amplifier systems usually consist of the following main parts as displayed in Figure 1. A broadband seed is generated and amplified in the optical parametric amplifier (OPA), consisting of a nonlinear crystal, which is pumped by a synchronized pump laser. In some cases, the broadband seed is directly generated by a fraction of the pump beam. The broadband signal is usually chirped by a stretcher prior to amplification to match the pump pulse duration and recompressed by a pulse compressor afterwards. This optical parametric chirped pulse amplification (OPCPA) scheme ensures efficient energy transfer from pump to signal even when the process is non-linear.

The amplification bandwidth of the OPA system depends mainly on the phase-matching conditions. It has been shown that the performance of OPCPA is dependent on the phase-matching conditions. It has been shown that the performance of OPCPA is optimized when the phase-matching condition is achieved within the laser medium. Thus, the spectral region and bandwidth of amplification can not only be freely designed by phase matching, but there is also no energy stored in the OPA crystal. As such, OPAs represent an ideal choice for amplification of the broad bandwidth of few-cycle pulses to high average powers.
2.2.1. Gain Bandwidth and Pump Pulse Duration

Broadband phase-matching in OPAs relies on group velocity mismatch in non-collinear wave vector matching within an OPA. Angular dispersion of either the pump or signal wave vector for group-velocity matching of signal and idler is usually termed “magic-angle”. For example, a target signal wavelength of 800 nm and a pump wavelength $\lambda_p = 515$ nm require an internal non-collinear wave vector angle $\alpha = 2.6^\circ$ in the commonly used Beta Barium Borate (BBO) for broadband amplification.

In addition, thin nonlinear crystals can be used to decrease the overall/integrated phase mismatch $\Delta \varphi = \Delta k l$ caused by the wave vector mismatch in OPAs, e.g. at non-perfectly phase-matched wavelengths. To achieve sufficient gain in this case, the pump intensity has to be increased accordingly. The damage threshold of the nonlinear crystal ultimately limits this approach. In a rough assumption, it scales inversely with the square root of the pulse duration for transparent media. Thus, shorter pump pulses can be focused to higher intensities and, therefore, allow using shorter crystals. Due to the reduction of accumulated phase mismatch in non-perfectly phase-matched regions of the spectrum, e.g. the spectral wings, the spectral bandwidth of the OPA is increased. This effect can be clearly seen in Figure 2b) where the parametric gain spectrum is plotted for different BBO crystal lengths. Note that the pump intensity has been set according to the square root scaling law of the damage threshold and the crystal length is adjusted for the same peak parametric gain at each pump intensity. Using 1 ps pump pulses (black curve) allows 100 GW/cm² pump intensity and an only 1 mm long crystal. In contrast, 1 ns pulses only permit 3 GW/cm² (green curve) and thus require a 5.8 mm long crystal, which reduces the gain bandwidth by nearly a factor of two. Consequently, ultra-broadband amplification requires preferably picosecond pump pulses for amplification of few-cycle laser pulses in an optimized non-collinear geometry, which will be presented at the end of the following section.

2.2.2. Bandwidth Enhancement to Support Few-Cycle Pulses

Many different approaches for increasing the bandwidth of parametric amplifiers have been demonstrated so far. They are essential to support few-cycle pulse durations.

Broadband Pump Radiation. Usually, the OPA pump is considered to be quasi-monochromatic. For example a 1 ps long Gaussian pulse at 515 nm wavelength has a relative spectral bandwidth of $\Delta \lambda / \lambda < 0.1\%$. Broadband, pump beams ($\Delta \lambda / \lambda >> 1\%$) can however enhance the amplification bandwidth, since different pump wavelengths can amplify different signal wavelengths in a phase-matched manner. This concept has been demonstrated already with broadband pump lasers,[39,40] but is not well suited for narrowband ps-class high power Yb-pump lasers.

Angularly Dispersed Beams. Angular dispersion of either the pump or signal wave adds an additional degree of freedom to tailor phase matching within an OPA.[41–43] An angularly dispersed pump provides different phase matching angles, while an angularly dispersed signal results in different non-collinear angles for different spectral components of the signal. So far, signal pulse...
Figure 2. a) Visualization of perfect non-collinear type-I phase-matching in birefringent crystals. The solid arrows represent the k-vectors of pump (green), signal (red) and idler (grey). The dashed arrows illustrate the situation for a different signal wavelength, which can be perfectly phase-matched at the same non-collinear angle $\alpha$ in special cases. b) Small-signal parametric gain spectrum for different crystal lengths and pump pulse durations. The calculation assumes a wide rectangular input spectrum and uses BBO in a non-collinear geometry with the following parameters: $\theta = 24.2^\circ$, $\alpha = 2.6^\circ$, $\lambda_p = 515$ nm.

Figure 3. a) Output spectrum of a two-color pumped OPCPA system (blue: linear scale, red: logarithmic scale, black: seed spectrum). b) Fourier-limited intensity profile (blue) and electrical field (red) assuming perfect compression of the output spectrum depicted in a). Reproduced from.\[43\]

Multiple Pump Wavelengths. Multiple pump beams can be applied to a single OPA. Particularly, multiple harmonics of the pump laser can be utilized to enhance the gain bandwidth.\[44\] Recently, the amplification of a signal covering more than one optical octave has been demonstrated by using a multi-color pump scheme.\[45\] The resulting output spectrum is displayed in Figure 3a). The corresponding Fourier-limited pulse duration is as short as 2.5 fs, but has not been achieved so far.

Pulse Synthesis. In order to circumvent the problem of spectral phase control of multi-octave spectra within a single pulse compressor, different spectral regions can also be amplified and compressed in different channels of an OPA system.\[46\] Subsequently, these pulses can be temporally overlapped provided that a precise relative phase control is feasible.\[47\] As such an ultrashort pulse can be composed out of multiple pulses with longer duration. The potential of this concept has been demonstrated for two OPA channels.\[46,48\] The results are illustrated in Figure 4. Figure 4a) displays the spectra delivered by two OPAs optimized for different wavelengths (blue and brown). If both are combined in phase, the green spectrum can be obtained, which spans nearly one optical octave. As a result, a pulse as short as 3.8 fs has been obtained as shown in Figure 4b). In principle the concept is even
suited to generate sub-cycle waveforms\[49\] however at the price of high system complexity.

**Frequency Domain OPA.** Spatially separated amplification can also be realized by placing multiple OPAs side by side within the Fourier-plane e.g. of a grating based zero-dispersion-line as shown in Figure 5.\[50\] Therefore, each OPA amplifies just a part of the whole spectrum and the spectral components are recombined by a second grating. This concept demonstrated two-cycle pulses with a four-crystal arrangement already, but possesses high demands on the stability of the optical setup and introduces additional losses and bandwidth limitations due to the required grating at the output. On the other hand, this scheme is very well suited to ps and very energetic/high average power pump lasers as (i) the signal is stretched to ps duration at the Fourier plane and (ii) the high energy/average power is spread over the total cross-section of the elongated beam mitigating issues related to damage threshold and heat dissipation. Finally, by transversely shaping the pump beam it is possible to directly address the gain at specific signal wavelength thus potentially increasing the amplified bandwidth.

**Phase-Mismatch Engineering.** In contrast to the rather complex approaches presented up to now, octave-spanning amplification can be achieved with a simple single-color pumped non-collinear OCPA system, via phase-mismatch engineering.\[37,51\]

In order to exploit the full potential of the non-collinear arrangement numerical simulations have been performed investigating the spectral gain characteristics of BBO for various non-collinear and phase matching angles. The resulting amplified signal spectrum is found by solving the coupled wave equations for the OPA. A Fourier transform of the spectrum (assuming flat spectral phase) provides the temporal pulse envelope. An effective peak power (peak power per millijoule pulse energy) can be calculated and used as a measure to find the optimum geometry.\[37\] The results of this two-dimensional analysis are presented in Figure 6. The three grey dots mark particular cases, which will be discussed in more detail in the following. The upper grey dot in Figure 6 represents the commonly used configuration with a non-collinear angle \( \alpha = 2.58^\circ \) and a phase matching angle \( \theta = 24.55^\circ \). Here, both the wave vector mismatch \( \Delta k \) and its first derivative are zero (see Figure 7a). The “magic angle” provides phase and group velocity matching simultaneously. It supports 6.5 fs pulse duration, whereas the effective peak power is 146 GW per mJ pulse energy.

However, the highest peak pulse power (207 GW/mJ) is obtained for a different configuration (\( \alpha = 2.16^\circ \) and \( \theta = 23.92^\circ \)). In this case the phase matching curve has three zero points and a small phase mismatch is tolerated in the regions in between as shown in Figure 7b). As a result, the spectral bandwidth is larger. The resulting pulse is shorter (4.1 fs), while the amount of energy in satellite pulses is slightly increased (see Figure 7d).

The shortest pulse is achieved by tolerating even larger phase mismatch as illustrated in Figure 7d). A duration of only 2.9 fs (FWHM) is achieved at \( \alpha = 1.86^\circ \) and \( \theta = 23.46^\circ \). However, the corresponding spectrum (Figure 7c) shows a significant gap in the center, which results in pronounced pre- and post-pulses.

It has to be emphasized that tailoring of phase mismatch is most effective, when using sub-picosecond duration pump pulses. Only with such short pump pulse durations it is possible to achieve sufficiently high gain (due to high pump intensity), in particular, when a certain amount of wave-vector-mismatch \( \Delta k \) is present requiring to use short crystal lengths.

**2.2.3. Understanding and Control of the Optical Parametric Phase**

As presented in the previous sub-section phase-mismatch engineering is a suitable approach for the amplification of few-cycle pulses in OPAs. However, phase mismatch also leads to a phase distortion of the amplified signal. In the spectral domain, the shape of this optical parametric phase (OPP) is proportional to the wavelength-dependent wave vector mismatch \( \Delta k \) and hinders pulse-compression to the Fourier-limit.

The measurement and the compensation of the optical parametric phase (OPP) has been reported in.\[53\] In the experiments only one OPA stage, a 1 mm long BBO crystal, was used. The spectral phase of the amplified pulses was measured with a SPIDER device. Chirped mirrors (second order compensation) and an active phase shaper (higher order compensation) were employed for pulse compression. In a first step, the signal pulses were compressed without being amplified in the OPA (no pump pulses). In a second step, the pump radiation was applied with an intensity of 280 GW/cm\(^2\) to the OPA. In this case the measurement of the spectral phase of the amplified signal pulses revealed the OPP.
Figure 7. a), b), c) Calculated gain spectrum (blue) and wave-vector mismatch versus wavelength (black dashed) for three different non-collinear angles. d) Corresponding normalized pulse profiles.

Figure 8. Measurement (black) and simulation (red) of the spectral intensity and the OPP for a non-collinear angle $\alpha = 1.92^\circ$.

The largest amplification bandwidth has been achieved with a non-collinear angle of $\alpha = 1.92^\circ$, which is in good agreement with the calculations presented in Figure 6. Figure 8 shows the corresponding measured and numerically calculated spectra and phases after amplification. Note that the spectral phase is dominated by third order dispersion (TOD) and group delay dispersion (GDD). However, higher order dispersion contributes as well. Hence, the OPP needs to be compensated in few-cycle OPA systems to achieve perfect pulse compression. The interested reader can find more information, experimental results and an extensive discussion of the OPP in.\textsuperscript{53}

2.2.4. Parasitic Nonlinear Effects and Parametric Fluorescence

Besides optical nonlinear effects several, usually unwanted, nonlinear effects may occur in the nonlinear crystal. This includes self-phase modulation and cross-phase modulation between the interacting waves. Other effects are parasitic second harmonic generation of signal or idler components and difference frequency generation, as well as cascaded nonlinearities. Usually, these effects are weak and negligible. However, if the OPA is operated at high intensities and particular processes are phase matched, they can severely influence the performance of OPAs. Detailed investigations of these effects and strategies how to avoid these effects can be found in the literature.\textsuperscript{54}

Parametric fluorescence is a quantum effect and can be considered as parametrically amplified quantum fluctuations. It is particularly strong if the seed energy of an OPA is low and the small signal gain is very high. For optimum performance and pulse contrast it needs to be minimized in any OPA system.\textsuperscript{55,56}

2.2.5. Ultra-Broadband Signal Generation

The generation of pulses with durations below two optical cycles requires nearly octave spanning spectra. Ti:Sapphire oscillators can be easily employed as seeders for OPAs, but do not necessarily offer such a large bandwidth which overlaps with the gain spectrum of the OPA. Ideally, a flat seed spectrum spanning from 600 nm to >1200 nm would be required (compare Figure 6c)). To overcome these limitations, several interesting spectral broadening schemes of ultrashort pulses have been subject of recent research. One possibility is white-light generation in bulk materials when focusing ~100 nJ to few-µJ pulse energies to high intensities.\textsuperscript{57,58} Laser host materials such as Sapphire, yttrium aluminum garnet (YAG), yttrium vanadate (YVO$_4$), gadolinium vanadate (GdVO$_4$) or potassium-gadolinium tungstate (KGW) are usually used for this purpose.\textsuperscript{59} Typical white-light spectra
generated in YAG with different driving wavelengths are displayed in Figure 9. Although the spectral intensity drops significantly around the driving wavelength, the flat part of the spectrum ranging from ~600 nm up to ~100 nm below the driving wavelength can be used for seeding of few-cycle OPAs.

Alternatively, optical fibers with a solid core can be utilized for spectral broadening. For example, a short piece of single mode fiber in combination with a chirped mirror compressor enabled pulse shortening to only 4 fs duration (1.7 optical cycles). However, this scheme is limited in terms of achievable bandwidth by the material dispersion of bulk silica, which introduces strong temporal pulse broadening. On the other hand photonic crystal fibers (PCF) are well suited for ultra-broad supercontinuum (SC) generation due to their special dispersion properties.

SC generation in a PCF with one zero dispersion wavelength (ZDW) close to the pump wavelength enabled pulse duration of 5.5 fs (2.4 optical cycles). However, the broadening process in such fibers is dominated by soliton dynamics, which cause fluctuations of the incident pulse. Therefore, they can cause variations of the spectral shape and phase from pulse to pulse, which limits the temporal coherence properties and, hence, the achievable pulse duration.

Spectral broadening in an all-normal dispersion PCF (ANDi PCF) represents an interesting alternative. This fiber exhibits only normal dispersion across the whole visible and near-infrared spectral region eliminating noise sensitive soliton dynamics. The process of spectral broadening is dominated by self-phase modulation (SPM) and optical wave breaking only. As a result very smooth and highly-coherent SC spectra can be obtained.

Figure 9. White-light spectra generated in a 4 mm YAG crystal (solid lines) with different driving spectra (dotted lines). All spectra covers more than one optical octave. Reproduced from.

Figure 10. a) un-shaped spectrum (black), shaped spectrum (blue shaded) and spectral phase (red) of the compressed pulses. b) Retrieved pulse shape with (blue shaded) and without shaping the spectrum (black). Spectral broadening and pulse compression in such a fiber has been investigated theoretically and experimentally.

By coupling 6 fs pulses from a Ti:Sapphire oscillator to a 13 mm long piece of ANDi PCF, spectral broadening to more than an optical octave of bandwidth was achieved. Recompression of the spectral fraction spanning from 600 nm to 1200 nm by chirped mirrors, a spectral phase shaper and fused silica wedges created pulses close to their Fourier-limit. The corresponding measured spectrum is shown in Figure 10a (black line). To increase the effective bandwidth of the spectrum even further, the spectral amplitude of the pulses has been shaped. The achieved spectrum (blue shaded) and the measured spectral phase (red) is shown in Figure 10a. Figure 10b shows the corresponding pulse envelope. The pulse FWHM duration is 3.64 fs (blue shaded), which is only slightly above the transform-limit (3.49 fs). These pulses contain only 1.3 optical cycles at the central wavelength of 810 nm. Thus, so far they are the shortest pulses achieved via temporal compression of SC spectra generated in PCF. The pre- and post-pulse contrast is 14 dB and 81% of the total energy is contained within the main pulse. In principle, spectral broadening within the ANDi PCF can even support more than one optical octave of bandwidth and single-cycle pulse durations. In consequence, these pulses are ideally suited as a seed source of an ultra-broadband OPCPA system.

2.2.6. Pump-Signal-Synchronization for Broadband Ti:Sa Oscillators

In OPCPA systems either a broadband seed is generated from a fraction of the pump pulses (as presented in the previous section) or the pump laser is seeded by a fraction of a broadband seed oscillator. In the case of a Ti:Sa oscillator, whose spectrum matches well with the gain curves presented in section 2.2.2, spectral broadening is usually required to achieve sufficient seed energy within the gain bandwidth of Yb-based pump lasers. One possibility consists in the use of an ANDi-PCF as presented in the previous section. The use of soliton self-frequency shift (SSFS) in PCFs to generate pJ level seed energy for high power Yb- or Nd-based amplifiers is another approach. It was first proposed and demonstrated by Teisett et al. and is nowadays widely used. Since it is well known that the dispersion properties of PCFs can be tailored to a large extent by controlling the fiber geometry, one could ask which PCF is best suited for this purpose. Typically the frequency shift is achieved by seeding in the negative dispersion region, above the zero dispersion wavelength (ZDW)
of the employed fiber.[66] However, Ti:Sa oscillators have beam pointing instabilities, which can cause amplitude fluctuations on the percent level when coupling to a small core fiber. Depending on the employed fiber and frequency shifting mechanism, this amplitude noise will translate to the frequency-shifted radiation. Not only will it influence the spectral shape and the frequency-shifted pulse energy, but also results in a timing jitter. A stability analysis of the frequency shifted radiation revealed that the above mentioned case of seeding in the negative dispersion region is very sensitive to input pulse fluctuations.[68] A thorough numerical analysis revealed that fibers, with longer ZDWs provide significantly lower amplitude fluctuations and timing jitter and are therefore beneficial.[68] Please note that propagation through the pump laser system can cause additional timing jitter of the pump pulses with respect to the signal pulses at the point of the OPA. This issue and adequate active stabilization schemes are discussed in detail in section 2.3.7.

2.3. High Average Power OPCPA System

In the following the experimental realization of a high average power OPCPA system is presented following the design guidelines presented in section 2.2. After introduction of the experimental setup, octave-spanning amplification and pulse compression to sub-two-cycles is demonstrated at record-high average power. In addition, the carrier envelope phase stability is discussed. Furthermore, the availability of a high average power fiber based pump laser system allows observing thermal effects in high power OPCPA system for the first time. The origin of these effects as well as possible mitigation strategies are discussed.

2.3.1. Experimental Setup

Figure 11 shows a simplified setup of the fiber laser pumped OPCPA system reported in.[36] A CEP-stabilized 76 MHz Ti:Sapphire oscillator served as a seed for both the OPA stages and a state-of-the-art fiber-chirped-pulse-amplification (FCPA) system that was employed as pump laser (for more details see[66]). One output of this oscillator was directed to the spectral broadening stage (ANDi-PCF, section 2.2.5). After passing through a phase shaper employing a spatial light modulator (SLM) the duration of the seed was adapted by chirped mirrors to match the pump pulse duration. Amplification of the broadband seed was then achieved by employing the frequency-doubled output of the FCPA as pump for a two-stage OPA. The second OPA stage was pumped by the remainder of the pump coming from the first stage. Such a two-stage amplification setup allows for efficient extraction of the pump energy, which is usually hindered by spatial gain narrowing.[69,70] The amplified pulses were finally compressed by a chirped mirror compressor and the phase shaper.

2.3.2. High Average Power Octave Spanning OPCPA

The 700 fs, 120 μJ pump pulses delivered by the fiber-based pump laser were focused to a peak intensity of ~300 GW/cm² in the first and ~200 GW/cm² in the second OPA stage, each consisting of a 2 mm long BBO crystal. Both stages employed the optimized geometry presented in section 2.2.2 (α = 2.16° and θ = 23.92°). The first amplification stage was optimized for a high gain factor of 10⁴ increasing the seed pulse energy (200 pJ) to 12 μJ. The second stage with lower gain allows for high energy conversion resulting in 29 μJ pulse energy at the output. Careful fine tuning of the non-collinear angle, the phase matching angle and the delay between signal and pump pulses in both stages resulted in ultra-broadband amplification. The obtained amplified spectrum is shown in Figure 12a). It possesses a -10 dB bandwidth larger than 500 nm and supports a Fourier-limited pulse duration of only 3.8 fs (1.3 optical cycles).

After amplification the beam was sent through a chirped mirror pulse compressor (6 bounces, -36 fs² each), which removed all the second order dispersion from the pulses. The compressor throughput was measured to be 90 %, resulting in up to 26 μJ of compressed pulse energy.

Compensation of higher order dispersion was performed with a pulse shaper placed in front of the OPA stages.[36] Unfortunately, the SPIDER measurement of the amplified pulses was severely disturbed due to parasitic nonlinear effects,[54] amplified fluorescence[55] and the strongly modulated spectrum. Thus, high-order phase compensation has been performed with the un-amplified seed pulses only.
In order to avoid phase-matched parasitic second harmonic generation and sum-frequency-generation (SFG) of signal and idler at around 800 nm signal wavelength, the non walkoff-compensating geometry has been selected for both OPA stages. Nevertheless, due to the octave-spanning bandwidth, phase matched SHG of 1120 nm signal was observed.

The amount of parametric fluorescence was estimated by blocking small fractions of the seed spectrum in the Fourier plane of the phase shaper. Then the power content in this particular spectral region, which can only originate from fluorescence, was measured. Overall 8% of the output energy were found to be fluorescence. Nevertheless, a sufficient pulse contrast of 30 dB can be expected, since the signal pulses are shortened in time by two orders of magnitude due to the compressor, while the fluorescence is not. In total, 24 μJ of energy have been attributed to the compressed few-cycle pulse.

Compression of the amplified pulses has been optimized for the shortest pulse duration by means of a fused silica wedge pair. The measured interferometric autocorrelation trace of the compressed pulses is displayed in Figure 12(b) (blue dots) together with the calculated autocorrelation trace of the Fourier-limited pulses (black line). Although these measurements suffered from some background signal, it confirmed a pulse duration of only 5.0 fs. This corresponds to sub-two optical cycles at 880 nm mean wavelength. Obviously, the measured pulses (5.0 fs) were significantly longer than the Fourier-limited pulses (3.8 fs). The reason is a non-perfect pulse compression, due to uncompensated high order dispersion originating from the optical parametric phase (OPP). In addition, the autocorrelation trace reveals a significant amount of energy in pre- and post-pulses. Nevertheless, the peak power of the pulses can be estimated to be about 2 GW.

2.3.3. Sub-Two Cycle Pulse Compression

The measurement of the spectral phase imposed by the two OPA stages required a modification of the OPAs. To reduce the saturation of the amplifiers and resulting multiple forth- and back-conversion, the pump intensity has been reduced to 200 GW/cm² in the first and to 74 GW/cm² in the second stage. Additionally, a 1 mm long BBO crystals is used for the second stage. Optimal bandwidth is achieved with a non-collinear angle of α = 1.92°. In this case the compressed signal pulse energy is reduced to 12 μJ. However, the amplified spectrum, as displayed in Figure 13a) (black line), possesses a much smoother shape and is free of strong spectral modulations. This spectrum supports 5.36 fs of Fourier-limited pulse duration and at the same time allows for reliable SPIDER measurements and pulse characterization. The measured phase that is introduced by the amplification process of both stages is shown by the blue line in Figure 13a) and represents mainly the OPP (see section 2.2.3). This phase was iteratively compensated by the phase shaper resulting in the red curve, which has been measured after full OPP-compensation. The compressed pulse is displayed in Figure 13b) (black). Almost perfect compression to 5.39 fs (1.9 optical cycles) is achieved. On the other hand, the pulse without compensated OPP Figure 13b)
red) is significantly longer with a pulse duration of 7.46 fs. This corresponds to 2.6 optical cycles and a loss of 29% in peak power. These experiments clearly demonstrated that compensation of the OPP is crucial for obtaining the shortest and most intense output pulses in few-cycle OPCPA systems.

2.3.4. Carrier-Envelope Phase Stability

For pulse durations of a few optical cycles the carrier-envelope phase (CEP) becomes an important quantity. It is defined as the phase between the peak of the envelope and the peak of the underlying oscillating carrier. Due to the short pulse durations of few-cycle pulses the shape of the electrical field will change dramatically for different values of the CEP. Therefore, it is essential to stabilize the CEP of the presented OPCPA system to have full control over the electrical field and, finally, use it for isolated attosecond pulse generation.[14]

Usually CEP-stabilized laser systems start with a seeder, delivering a CEP-stable pulse train. This can be either an actively CEP-stabilized femtosecond oscillator[71] or passively CEP-stable pulse train generated by difference frequency generation.[72,73] Subsequent amplifiers need to be designed for low CEP drifts. Usually an additional active compensation of those CEP drifts, e.g. caused by vibrations, temperature changes or air flow, is performed to achieve the highest possible CEP stability.[74–78]

It is well-known that the amplification of a signal in an OPA does not affect its carrier-envelope phase stability.[79] Hence, OPA systems can in principle deliver CEP-stable output pulses, provided that they are seeded with a CEP-stable pulse train. However, nonlinearities such as self-phase modulation (SPM), cross-phase modulation (XPM) and the optical parametric phase (OPP) can couple power fluctuations, i.e. amplitude instabilities of either one of the interacting waves (pump, signal and idler), to additional phase fluctuations.[13,77] Another critical aspect in optical parametric chirped-pulse amplifiers (OPCPA), is the timing between pump and the chirped signal pulses in the nonlinear amplification stages. It can lead to power fluctuations as well as a change in spectral and temporal shape of the few-cycle laser pulses. Such timing jitter can originate from the synchronization scheme of signal and pump. In addition, thermal drifts and mechanical vibrations can contribute significantly due to generally different beam paths of pump and signal. In particular, long beam paths (of the order of tens of meters) in the pump laser (e.g. through grating stretchers or regenerative amplifiers) result in temporal drifts of the order of 100 fs easily. Controlling this timing jitter is especially important for sub-ps-pumped OPCPA systems that are employed for octave spanning amplification.

2.3.5. Measuring Carrier Envelope Phase Drifts by Utilization of Parasitic Nonlinearities

A number of methods have been introduced to measure the drifts of the carrier envelope phase, e.g. different interferometric techniques such as f-2f interferometry[77] or techniques relying on the field sensitivity of physical processes like above-threshold ionization (stereo ATI).[76] To measure the CEP drifts usually an octave-spanning spectrum has to be generated. Then the long wavelength part of this spectrum is frequency doubled and interferes with the short wavelength part. The interference pattern contains the information of the CEP drift, which can be extracted via Fourier transform. When this technique is applied to the amplified pulses it is realized by creating white-light in a plate of bulk material (e.g. Sapphire or YAG), frequency doubling in a BBO and interference is obtained by projecting the orthogonally polarized fields onto a common polarization axis with a polarizer.

Due to its broad amplification bandwidth, which becomes available by using the design criteria explained in section 2.2.2, the OPCPA system inherently offers the bandwidth required for f-2f interferometry. Moreover, it is operated in the so-called non-walk off compensated geometry, which leads to broadband parasitic second harmonic generation of the signal at 1120 nm.[34] Consequently, it also offers the frequency doubling part of the f-2f interferometer (see Figure 14a). To measure the CEP drifts the components below 630 nm of the signal needed to be reflected and sent through a polarizer to achieve interference. An error signal of the Ti:Sa oscillator’s phase locking electronic modulated with a 0.1 Hz sinusoidal signal, served as a reference signal. This allowed to compare the results of the parasitic second
Figure 15. a) Drift of the central wavelength of the OPCPA system caused by the movement of the piezo-driven mirror. b) The drifts of the CEP corresponding to the wavelength drift. The PSHG based method (black) agrees well with the standard white-light based f-2f interferometer (red).

harmonic generation (PSHG) based f-2f interferometer with a standard white-light based f-2f interferometer, which has been placed after the compressor of the OPCPA. It can be clearly seen in Figure 14b) that both results agree very well and, therefore, the PSHG method can be used for measurements of the CEP drift without affecting the performance of the OPCPA system in terms of energy and pulse duration.[80]

2.3.6. Influence of Pump-Signal Timing Jitter to CEP Stability

Besides energy fluctuations of the pump or seed pulses themselves, the timing jitter between these two waves can lead to fluctuations of the amplified signal. Due to the instantaneous nature of the parametric amplification any drifts in the arrival time of the respective waves will immediately change the characteristics, in particular, the output energy and the spectral shape. It was found that the latter one not only can change the pulse duration, but also the CEP.

Once a temporal drift between pump and signal occurs the spectral shape, i.e. the central wavelength of the signal, will change. Propagation of the amplified signal in the remainder of the BBO crystal or other dispersive components, e.g. air or material, will shift the CEP by

\[ \Delta \varphi_{\text{CEP}} = -2\pi \left( \frac{dn}{d\lambda} \right)_{\lambda_0} z \]  

according to its central wavelength \( \lambda_0 \), the material length \( z \) and the index of refraction \( n(\lambda) \).[81] According to eq. (1) the value of the CEP after propagation will be different for different central wavelengths. Obviously, a drift of the pump and the signal arrival time will lead to a drifting central wavelength and, therefore, additional CEP jitter.

This phenomenon has been investigated with the OPCPA system presented above. A piezo-driven mirror has been included before the main amplifier fiber, which allows for changing the timing by several tens of femtoseconds. For systematic investigations a triangular voltage was applied to the piezo-driven mirror in order to shift the temporal arrival time of the pump pulse by 45 fs from the point of maximum output energy. Obviously, this lead to a drift of the central wavelength as shown in Figure 15a). Please note that the central wavelength has been calculated from spectra measured with a silicon detector and, therefore, is only qualitative due to the decreased detector response above 950 nm. Nevertheless, a simultaneous measurement of the CEP drifts (Figure 15b)) shows that they follow the drift in central wavelength with the same periodicity.

Even a small drift in central wavelength of only 5 nm (<1% of the mean wavelength) can significantly shift the CEP (0.8 rad). However, it has to be stressed that the actual value of the CEP drift depends on the exact experimental conditions (durations of pump and signal, chirp of the signal, saturation behavior of the amplifier, phase-matching condition) and, therefore, will be different in other systems. The periodicity of the CEP drifts allowed, however, to exclude other sources, such as the already mentioned nonlinear amplitude to phase coupling mechanisms.[80] Consequently, a new source of CEP drifts has been identified in OPCPA systems, which is particularly important for (sub)-ps-pump lasers as typically utilized in few-cycle OPCPA systems.

2.3.7. Active Stabilization of the Timing Jitter and the CEP

The above described experimental findings show the need for an accurate timing between pump and signal pulses in the parametric amplifiers. In section 2.3.1 it was shown that the same oscillator is used for seeding of the pump laser and the OPCPA system. Furthermore, it was shown that the required frequency shifting mechanism to seed the pump laser can be optimized for minimal timing jitter by a proper choice of the nonlinear fiber. However, due to the long beam paths, in particular in the stretcher and compressor of the pump laser, an additional timing jitter of several tens of femtosecond is easily acquired.[82] Thus, a detection of the timing jitter and an active stabilization system needs to be implemented to increase the stability and to reduce CEP drifts. Usually, part of the signal and the pump is split and mixed through a certain nonlinear effect e.g. sum frequency generation[83] to generate a feedback signal to control an optical delay line within the pump laser beam path. Alternatively, balanced cross-correlators[48] can be employed or, the spectrum of an additional small-scale parametric amplifier, seeded with a strongly chirped signal, can be recorded.[84] The precision of these methods has been pushed down to a few-femtoseconds already. Figure 16a) displays the corresponding pump-signal jitter for a typical OPCPA system without active stabilization (blue). When
applying the active stabilization feedback, the jitter is dramatically reduced (green).84

An elegant alternative is inline recording of the spectrum of the actual OPCPA system, which does not require additional nonlinear mixing. The idler radiation, which is usually not employed, is already angularly dispersed. Temporal drifts between pump and signal pulses will lead to a change in the signal spectrum and, via energy-conservation, also in the idler spectrum. This change will appear most rapid in the spectral wings. Thus, a simple arrangement of two photodiodes placed at the edges of the idler spectrum, which is spread out in space, is sufficient to detect pump-signal timing jitter (Figure 16b)). The difference signal of these two photodiodes was used as input for a PID (proportional, integral, differential) controller, which generates an error signal. This was fed back to the piezo-driven mirror (delay) and used for active stabilization. This simple stabilization scheme could reduce the delay drift (out-of-loop) to $\frac{18}{223}\text{fs}$ and was operated without detriment in performance of the OPCPA system. Therefore, such stabilization can be implemented as online monitoring without sacrificing performance.

When applying the presented stabilization scheme to the high power OPCPA system presented in 2.3, a CEP drift as small as 86 mrad (standard deviation, 100 μs integration time) at 150 kHz repetition rate over a time-span of 40 minutes has been achieved as shown in Figure 16c).80

2.4. Thermal Limitations

Optical parametric amplifiers have the reputation to be scalable to high average powers due to the lack of energy storage inherent to the instantaneous nature of the nonlinear interaction of the waves. However, it was never expected that this holds true for unlimited power levels. With the availability of high average power pump sources thermal effects similar to those observed in conventional lasers will be obtained. Such effects have already been observed in optical parametric oscillators (OPOs) and have been assigned to absorption of the resonant wave in the nonlinear crystal.85,86

A detailed experimental study and analysis of thermal effects has been carried out at a few-cycle OPCPA system operated at very high average power and repetition rate88 (see also section 0). For the experiments presented in this section 100 μJ pump pulses at up to 1 MHz repetition rate have been used to pump a 3 mm long BBO (non-collinear angle 2°, phase matching angle 21.8° at a peak intensity of 200 GW/cm²). The octave-spanning signal (see section 2.2.5) serves as seed. The crystal peak temperature is monitored by means of a thermo-camera with careful calibration of the material emissivity. Figure 17 illustrates the measured temperatures versus the pump laser repetition rate at constant pump pulse energy. Similar spectra and pulse energies are achieved for all repetition rates by slight tuning of the phase matching angle. The blue points represent measurements with the full seed spectrum. At a moderate output power of 2.1 W (150 kHz repetition rate) the crystal is already heated to 70°C. At 14 W output power (1 MHz repetition rate) up to 170°C have been measured.

The observed heating of the crystal is caused by absorption of light. The absorption coefficient of BBO is very small at the signal wavelength range ($\alpha$<0.002 cm$^{-1}$ at 1 μm).88 The pump wave, however, experiences much higher absorption (0.01 cm$^{-1}$ at 532 nm). Moreover, the absorption coefficient increases strongly at longer wavelengths from 0.07 cm$^{-1}$ at 2.09 μm to 0.5 cm$^{-1}$ at 2.55 μm.88 Hence, the idler wave spanning from 900 nm up to 3.6 μm in this configuration, is expected to contribute to the heating of the OPA crystal as well.
Idler wavelengths larger than 1950 nm can be avoided by blocking of the seed wavelengths below 700 nm which reduces idler absorption to a minimum. Another series of temperature measurements at different repetition rates has been performed in this configuration, which is represented by the black dots in Figure 17. The temperature difference is reduced by \( \sim 40\% \). In consequence, the remaining part can be attributed to absorption of the pump pulses. Note that only a small amount of amplified fluorescence is measured, when the seed beam is completely blocked. The corresponding temperatures are represented by the red points in Figure 17. Interestingly they are found to be slightly higher compared to the seeded case without idler absorption (black dots). This difference is most likely due to absorption of the infrared fraction of the amplified fluorescence (idler).

In order to quantify the tensile stress \( \sigma \), which is induced by the temperature gradients inside the nonlinear crystal, a numerical simulation based on the finite element method is performed. More details on the simulation and the corresponding parameters can be found in.\[87\] The resulting temperature difference to the ambient temperature is shown in Figure 18a) and confirms the measurements very well. Figure 18b) displays the resulting mechanical tensile stress. Due to the thermal expansion of the interior of the crystal, the largest tensile stress of up to 40 MPa is found at the front and rear surface. It is important to know that such high tensile stress can already induce damage of the crystal\[89\] and thus needs to be avoided.

Please note that, considering only radial heat flow, a larger diameter will reduce the generated heat per volume, hence, reduce the temperature gradient. However the total temperature difference between center and wing of the beam will only depend on the applied heat per volume and the thermal conductivity. Therefore, in this simple consideration, the thermal limitations of OPAs are completely independent of the beam diameter. Consequently, a 1 kHz system will show the same temperature gradient and thermal dephasing as a 1 MHz system with \( \sim 32 \) times smaller beam diameter (assuming the same beam intensities). In reality, there will be a small contribution of longitudinal heat flow towards the front- and back surface of the nonlinear crystals, which slightly favors systems with larger beam diameters (higher pulse energy and lower repetition rate).

2.4.1. Mitigation Strategies

Clearly avoiding absorption in the crystal material is a key strategy to mitigate thermal heating. It is of enormous importance that all interacting waves are far from any absorption of the nonlinear material. From the measurements presented in the previous sub-section it can be concluded that special care should be taken not to overlap the idler spectrum with an infrared absorption band e.g. by restricting the signal wavelength range.\[36\] Furthermore, the maximum photon energy of the pump wave should be kept well below 50% of the UV-band gap energy to avoid two-photon absorption. Additionally, the process of crystal growth could be optimized aiming for a minimum amount of impurities and inclusions to reduce the residual absorption.\[89–91\] Note that cooling of the side surfaces of the nonlinear crystal will reduce the peak temperature, however, not the gradients within the crystal. Thus, the material parameters of the nonlinear crystals play a key role in future power scaling. LBO or YCOB, for example, have a more than three times higher thermal conductivity compared to BBO.\[92,93\] Additional absorption within the coating of the crystals can be reduced by removing the coating.\[87\] The resulting Fresnel reflections at both crystal surfaces could be avoided by employing the nonlinear crystals under Brewster’s angle in this case, at least for a single polarizations axis.

Alternatively, a transparent material of high thermal conductivity could be attached as heat spreader to the front and the rear surface of the nonlinear crystal in order to improve the heat dissipation.\[94–96\]

Another possibility to reduce the thermal load onto a single nonlinear crystal is to distribute the thermal load to multiple crystals. They can be arranged either serially\[97\] or in the Fourier plane of a grating arrangement.\[50\]
Table 1. Overview on current state-of-the-art high average power few-cycle OPCPA systems

| Author [Ref.]   | Nonlinear crystal | Central wavelength | Pulse duration (Fourier-Limit) | Phase-matching concept       | Pulse energy | Repetition-rate | Average power | CEP stability |
|-----------------|-------------------|--------------------|-------------------------------|-----------------------------|--------------|----------------|---------------|---------------|
| Baltuska et al. [43] | BBO              | ~600 nm            | <4.0 fs (3.5 fs)              | Angular-dispersed pump      | 0.5 µJ       | 1 kHz          | 0.5 mW        | no            |
| Rothhardt et al. [36] | BBO             | 880 nm             | 5.0 fs (3.8 fs)               | Phase mismatch engineering  | 22 µJ        | 1 MHz          | 22 W          | 0.41 rad (95 s) |
| Harth et al. [45]   | BBO              | 667 nm             | 4.6 fs (2.5 fs)               | Two-color pump              | 1 µJ         | 0.2 MHz        | 0.2 W         | no            |
| Prinz et al. [98]   | BBO              | ~850 nm            | 5.7 fs                       | Close to magic angle        | 50 µJ        | 0.3 MHz        | 15 W          | 0.24 rad (20 min) |
| Furch et al. [99]   | BBO              | ~800 nm            | 6.3 fs (5.8 fs)               | Close to magic angle        | 12.5 µJ      | 0.4 MHz        | 5 W           | yes           |
| Manzoni et al. [48] | BBO              | ~700 nm            | 3.8 fs (3.65 fs)              | Pulse synthesis             | <4 µJ        | 1 kHz          | <4 mW         | no            |
| Budriunas et al. [91] | BBO             | ~880 nm            | ~9 fs                        | Close to magic angle        | 53 mJ        | 1 kHz          | 53 W          | 0.22 rad      |

Figure 19. Schematic layout of nonlinear compression.

2.5. Summary and Overview on State-of-the-Art High Average Power Few-Cycle OPCPA Systems

The architecture, concepts and main limitations of high average power few-cycle OPCPA systems have been presented in the previous subsections. Table 1 provides an overview on state-of-the-art few-cycle OPCPA systems. It can be seen that all systems rely on BBO as nonlinear crystal material. Very short pulses can be achieved by angularly dispersed pump beams, two-color pumping and pulse synthesis. However, a high average power has so far only been achieved by systems relying on the simple and efficient non-collinear phase matching scheme with slight modifications and further power scaling towards 100 W remains challenging.

3. Nonlinear Pulse Compression to Few-Cycle Durations

As mentioned earlier nonlinear compression is an alternative to OPCPA systems. It relies on spectral broadening and subsequent temporal compression, rather than utilizing large amplification bandwidths. Its principle has been first proposed in 1969 by Fisher et al., and is illustrated in Figure 19. A laser pulse with a certain bandwidth and pulse duration emitted from a laser system is propagated in a nonlinear medium, where it undergoes the process of self-phase modulation (SPM). This imposes a temporal chirp on the pulse, which leads to spectral broadening. The removal of the chirp leads to shorter pulses and, at the same time, to a higher peak power if the losses are sufficiently low.

Over the years many realizations of such experiments have been presented. Most commonly the process of SPM is achieved in a waveguide, which has the advantage that the propagation in the waveguide leads to a spatially uniform spectral broadening, i.e. no spatial chirp. Additionally, longer interaction lengths and, therefore, larger spectral broadening is possible. In fact, the first compression to femtosecond pulses was achieved in an optical fiber in 1982. Optical fibers can be used for sub-ps pulses up to approximately one microjoule pulse energy. At higher pulse energies self-focusing and associated damage can occur. The next important step in nonlinear compression was established by Nisoli et al., which used a noble-gas-filled hollow waveguide (a capillary) for compression. The noble gas filling and the large inner diameter allowed the use of higher energies up to the millijoule range. The realization of this type of nonlinear compression was an important step for attosecond physics. Still hollow core compression of Ti:Sapphire lasers to sub-2cycle CEP stable pulses is the standard technique for attosecond experiments. However, for a long time a gap remained for the medium energies of a few tens of microjoule, where a standard optical fiber is damaged and the necessary small diameters of capillaries lead to unacceptable high losses. With the advent of so-called Kagome photonic crystal fibers this situation changed. Kagome fibers behave like capillaries in terms of their dispersive properties, but exhibit significantly lower propagation losses.

Therefore, they are now routinely used for nonlinear compression experiments with few-microjoule to 100 microjoule-level pulses.

Besides the use of waveguides, standard bulk material can be used for achieving SPM. Several demonstrations have been made over the years and with recent progress the efficiencies have significantly increased. This approach seems to be particularly valuable when aiming at nonlinear compression of high intensity lasers. Very recently, a multi-pass configuration has been utilized to achieve several passes through the nonlinear medium easing some of the experimental difficulties associated with multi-plate arrangements.
3.1. Nonlinear Pulse Compression in Gas-Filled Hollow Waveguides: Design Criteria, Limitations and Mitigation Strategies

A nonlinear compression experiment aims at shortening the input pulses in their duration either to a certain value or to the shortest possible duration. The following paragraph will briefly outline some important design criteria. Due to its simplicity, it does not account for effects like self-steepening, dispersion or temporal pulse quality, but it gives a good guideline on how to design nonlinear compression experiments.

Pinault et al. have shown that the compression factor \( F \) can be expressed with the bandwidth of the initial \( \Delta \omega_0 \) and broadened spectrum \( \Delta \omega_{\text{bro}} \) as:

\[
F = \frac{\Delta \omega_0}{\Delta \omega_{\text{bro}}} = \left(1 + \frac{4}{3\sqrt{3}} \Phi_{nl}^2 \right) \approx 0.88 \Phi_{nl}^2, \quad \text{if } \Phi_{nl} \gg 1
\]

where \( \Phi_{nl} \) is the nonlinear phase. Obviously, the larger the broadening factor the shorter the compressed pulses can be. In turn, this requires to accumulate as much nonlinear phase as possible. The latter one follows the simple textbook expression\(^{[122]}\):

\[
\Phi_{nl}^{\text{max}} \propto n_2 P_{\text{peak}} M F D^2 L_{\text{eff}},
\]

where \( n_2 \), \( P_{\text{peak}} \), \( M F D \) and \( L_{\text{eff}} = \left(1 - \exp(-a L)\right) \) are the nonlinear refractive index, the peak power, the mode field diameter and the effective length (\( a \) - loss coefficient, \( L \) - length of fiber), respectively. This simple equation already allows to discuss some of the limitations. The first one is a rather practical, in particular, for capillaries where the length, or more precisely the effective length, cannot be arbitrarily long. Since capillaries are very sensitive to bending losses they must be kept straight, which is difficult for long lengths. There has been work on so-called stretch capillaries of a few meter length,\(^{[123-126]}\) but still most experiments use a maximum length of 1 m.\(^{[127]}\)

Another limitation is given by self-focusing, which sets a limit to the product \( n_2 P_{\text{peak}} \) of the nonlinear refractive index and peak power of the incident laser pulse. For gas-filled waveguides the nonlinear refractive index is pressure dependent and can be written as\(^{[122]}\):

\[
n_2 = n_2 \cdot P.
\]

where \( n_2 \) is the nonlinear refractive index at standard conditions and \( P \) is the pressure of the gas.\(^{[128]}\) Although self-focusing sets an upper limit on the achievable nonlinear phase, the equation above shows that the required broadening can be achieved by an arbitrary gaseous nonlinear medium by adapting its pressure.

However, this assumption neglects the gas ionization potentially occurring for the high intensities experienced in the fiber core. This puts a limit on the smallest possible inner radius, which was found to follow the relation\(^{[127]}\):

\[
a_{\text{min}} = A T_{\text{opt}}^\alpha E_0^\beta,
\]

where \( A \) is a numerically obtained constant depending on the gas, \( T_{\text{opt}} \) is the pulse duration as defined in,\(^{[127]}\) \( E_0 \) is the pulse energy and \( \alpha \cong 0.45 \) and \( \beta \cong 0.51 \).\(^{[127]}\)

For a capillary equation (3) can be used to deduce an optimal inner radius for a given fiber length (\( L \)) and wavelength (\( \lambda \)).\(^{[129]}\)

For the highest compression factor this value is

\[
L_{\text{opt}}^{\text{compression}} = 0.61 \cdot \left(\frac{L \lambda^2}{L_{\text{eff}} \Phi_{nl}}\right)^{1/3}
\]

while it is

\[
L_{\text{opt}}^{\text{peak power}} = 0.98 \cdot \left(\frac{L \lambda^2}{L_{\text{eff}} \Phi_{nl}}\right)^{1/3}
\]

for the highest peak power enhancement.

Equations (5)–(7) can now be used to determine an optimal configuration for a capillary based compressor. For a given set of pulse parameters (energy and duration) the minimum possible radius for compression according to eq. (5) is calculated. If this value is larger than either (6) or (7) the compression scheme is ionization limited, otherwise self-focusing restricts the achievable shortest pulse duration. In fact, the ionization limit can be avoided by either choosing a noble gas with high ionization potential or by making the capillary longer. In that case eqs. (6) and (7) dictate that a larger inner radius can be used, which reduces the intensity inside the capillary. This might be achieved by employing the stretched capillary approach introduced by Nagy et al.\(^{[121,126]}\)

3.1.1. Limitations and Mitigation

For a general nonlinear compression experiment utilizing gas-filled waveguides and, particularly, for operation at very high average power the most important limiting factors and their potential mitigation must be studied. For capillary based approaches these are (some of them have already been mentioned above)

(a) Self-focusing
(b) Ionization
(c) Propagation loss of the waveguide
(d) Absorption losses in employed optics

The effects of a) and b) have already been discussed in the paragraphs above, since they are generally important for the design of capillary-based pulse compression experiments. In the following point c) and d) will be discussed, since they turn out to be of utter importance for experiments at high average power.

It is well known and established that standard optical fibers can supply kW average power level. Consequently, it is not surprising that nonlinear compression experiments with standard optical fibers have already reached 250W of average power.\(^{[130]}\) However, for using few-cycle pulses in strong field experiments an energy of a few tens of microjoule is required. Therefore, only capillary- or Kagome-based compression schemes will be considered in the following. Both approaches need to ensure stable operation at average powers up to 1kW, which can nowadays be delivered by femtosecond ytterbium based laser architectures.\(^{[14,15,27,131]}\) This includes the waveguides itself as well as all the involved components, such as lenses, laser windows and mirrors. For that purpose a testbed with a 1kW CW laser has been implemented to study average power related (c) and (d) of above list) effects in
nonlinear compression.\cite{132} Using a CW laser allows to investigate purely average power related effects without the possible interference of phenomena such as self-focusing or ionization, i.e. to separated points c) and d) from a) and b).

In a first experimental test capillary and Kagome waveguides were tested. Such a test is particularly important for capillaries, which exhibit a high transmission loss. As described by Marcatili and Schmeltzer the propagation losses (for the electrical field) can be written as:\cite{133}

\[
\alpha = \left(\frac{2.405}{\pi}\right)^2 \cdot \frac{\lambda^2}{a^2} \cdot \frac{1}{\sqrt{\nu^2 + 1}} \cdot \frac{\nu^2 + 1}{\nu^2 - 1},
\]

where \(\nu = n_{\text{glass}}/n_{\text{core}}\) is the ratio of the refractive indices of the glass and core, \(\lambda\) the wavelength and \(a\) the inner radius, respectively. For a standard capillary with 250\(\mu\)m diameter and a length of 1 m, already, more than 20% of the power is lost upon propagation. Naturally, this leads to large thermal loads when going to high average powers. Therefore, a specific water-cooled mounting of capillaries and Kagome fibers is necessary and described in more detail in.\cite{132}

In a first step the cw laser has been coupled to both waveguides. Subsequently, the average power is increased stepwise and the transmitted power as well as the near field beam profiles are monitored.

Figure 20 shows the results of this transmission test for a capillary a) and a Kagome b) waveguide. Due to the lower transmission losses of the Kagome fiber, it exhibits a transmission value of \(\sim 90\%\) in comparison to \(>70\%\) in a capillary. Nevertheless, both waveguides can be easily operated up to the full available average power of 1kW leading to 712 W (capillary) and 900 W (Kagome) of transmitted average power, respectively. Over the whole range of average powers stable operation was possible, due to the water-cooled mounting and the beam profiles (near-field) remained unchanged (Figure 20).

In terms of optical elements (point d) on the list), such as lenses or windows, it has already been shown for cw lasers that low absorption glasses and anti-reflection coatings have to be used.\cite{134} These elements present no limitation to the power levels (1 kW) discussed here.\cite{132} Even more important with respect to few-cycle pulse generation are mirrors. Commonly, metal optics are used for near-octave spanning bandwidths, since they offer a reasonable reflectivity and low dispersion. However, it has been observed that at 50W average power these metal optics already show significant heating and, therefore, cannot be used for high average power few-cycle lasers.\cite{135} Alternatively, gold-coated sapphire substrates or group delay dispersion (GDD) - optimized dielectric mirrors can be used. The former ones start to show thermal effects at power levels exceeding 500W, while GDD-optimized mirrors have been operated up to 1kW of average power already.\cite{132}

In conclusion, it has been demonstrated that nonlinear compression is an average power scalable concept that can be used to generate few-cycle laser pulses from kilowatt class femtosecond ytterbium based laser systems.

3.1.2. Multiplexing Approaches: Scaling to Higher Energy

High intensity experiments require the highest possible pulse energy potentially combined with high average power. Therefore, several techniques have been proposed over the recent years to circumvent some of the limitations mentioned above. Certainly, for high energy input pulses the ionization of the gas medium becomes the most limiting factor. As discussed above a potential way to avoid ionization is to use large inner diameter capillaries with long lengths.\cite{17,125,126} Another potential way is to use differentially pumped capillaries, where the entrance is kept under vacuum while the gas is filled at the backend of the fiber.\cite{136}

This way there is no ionization induced defocusing at the entrance of the capillary and higher pulse energies can be achieved. A third way is the use of circular polarization, which allows for controlled behavior of the ionization induced instabilities and, therefore, the operation at higher pulse energies as compared to linear polarization.\cite{137,138}

A different approach is the use of so-called multiplexing schemes, where the energy of the incoming pulse is distributed over two or more spatially or temporally separated replicas, where each of the sub-pulses undergoes spectral broadening before all replicas are re-combined and compressed. Figure 21 displays an experimental setup which has been used for generating temporally separated replicas to be spectrally broadened, recombined and recompressed.\cite{139}

Both spatial and temporal multiplexing essentially represent interferometric approaches, which might have to be combined with some sort of active stabilization. Their feasibility has first
been studied using standard solid-core optical fibers with very encouraging results,[139–141] After a first proposal, [139,140,142] the temporal multiplexing approach has also been demonstrated for hollow fiber compressors operated at high energy, few-cycle pulse duration and proved even carrier envelope phase-stability.[143,144] Although the first experiments have only been performed with two replicas, recent experiments with four replicas hint at scaling towards a larger number of pulse copies and potentially a significant increase in pulse energy of few-cycle hollow core compressors.[145]

3.2. High Average Power Few-Cycle Pulses via Hollow-Fiber Compression

Based on the findings above nonlinear compression experiments can be designed and performed. The first important approach in that regard is the use of Kagome fiber as waveguide for nonlinear compression. It has been shown that few-cycle pulses can be generated with a two-compression stage approach using a 38W, μJ thin-disk oscillator.[111] In that experiment 14.5W, 9.1 fs pulses have been achieved (Figure 22), although, soliton-effect self-compression was utilized in the second compression stage.

Experiments with higher average power have only been performed with one compression stage with thin-disc oscillators[113] or fiber lasers, respectively. Emaury et al. have shown that more than 100W of average power can be obtained after transmission through one Kagome fiber. In that experiment only partial compression to ~10W, 88fs has been demonstrated.[113] In a later experiment that value was increased to 46W, 108fs.[114] The highest average power for Kagome based nonlinear compression has been demonstrated with fiber lasers, where 76W, 31fs have been achieved and successfully been used for high harmonic generation at 10 MHz repetition rate.[114]

Experiments towards few-cycle pulses have been performed with a two-stage compression approach based on capillaries. In these experiments a fiber laser frontend has first been compressed to 30fs and subsequently to few-cycle pulse durations.

The first experiment in that regard reached 53W, sub-8fs at 150kHz.[135] However, in that experiment the thermal limitations of employed silver mirrors hindered further power scaling. In fact, subsequent experiments have been limited to ~30W of average power, because of this effect. As described in section 3.2 the use of appropriate mirrors with low absorption and optimized dispersion properties should allow for higher power levels.

Figure 21. Experimental setup used for divided pulse nonlinear compression. The input pulses are split into a series of pulses by transmission through birefringent crystals and spectrally broadened in a fiber. After polarization rotation with a Faraday-rotator the spectrally broadened pulses are temporally recombined by the same set of birefringent crystals and recompressed. Reproduced from.[139]

Figure 22. Experimental realization of few-cycle pulses with Kagome fibers. The average power at the output is 14.5W. (a) Experimental SHG-FROG trace at the output of the two-stage compression setup. (b) the retrieved SHG-FROG trace. (c)(i) Corresponding measured (blue line) and retrieved (gray line) spectra after the chirped mirror compressor (c)(ii) Experimental output spectrum before the chirped mirrors with increased argon pressure (29 bar) enhancing the dispersive wave emission. (d) The retrieved intensity (solid-blue line) and phase (solid-green line) profiles corresponding to (c)(i). The Fourier-transform-limited pulse duration for the retrieved spectrum is represented by the gray dots. (Reproduced from[111])
Indeed, the predicted scaling properties could be experimentally validated. The frontend in that experiment was an 8-channel coherently combined fiber laser system delivering 660 W at 1.27 MHz.\textsuperscript{[14]} In the first stage compression to 30 fs pulses at 408 W of average power is achieved, which already constitutes the highest average power ultrashort pulse laser demonstrated to date.\textsuperscript{[147]}

The second compression stage is utilized to compress the pulses down to 6.3 fs in duration (Figure 23). At a central wavelength of 980 nm this corresponds to a laser pulse that is shorter than two optical cycles. At the same time 216 W of average power is achieved in this short pulses surpassing OPCPA systems by an order of magnitude.\textsuperscript{[147]} At the used repetition rate this corresponds to 170 μJ of pulse energy, which is sufficient for many strong-field experiments (peak power of >17 GW). Therefore, it can be expected that this source has unique potential to advance many experimental fields, e.g. attosecond science.

Recently, a new compression scheme based on a multi-pass cell has been proposed. The basic idea is to achieve multiple transmissions through a bulk material in a compact and reliable setup. This way, the 850 fs of a >400 W Yb:YAG inoslab amplifier could be compressed to 170 fs at 375 W of average power.\textsuperscript{[120]} However, due to the free propagation this scheme cannot provide spatially uniform spectral broadening and pulse shapes with Gaussian beams.

### 3.2.1. Potential for Carrier-Envelope-Phase Stabilization

As mentioned already, the stabilization of the carrier envelope phase (CEP) is a crucial pre-requisite for many experiments that are performed with few-cycle laser pulses. As outlined in section 3.1 the process of SPM is associated with a nonlinear phase-shift, which is responsible for spectral broadening. Obviously, additional instabilities of the CEP are introduced by any fluctuations of this phase, e.g. by pulse-to-pulse intensity fluctuations. The larger the nonlinear phase is (or the spectral broadening) the more sensitive is the CEP stability of the output pulses to fluctuations. Basically, this dictates the need for very stable laser systems in terms of beam-pointing stability, peak-to-peak intensity variations and, obviously, CEP stability of the input pulses itself. Therefore, it is not surprising that over the years many demonstrations of CEP stable pulse compressors have been reported and various studies on the influence of several aspects on the CEP stability have been performed.\textsuperscript{[75,125,148–152]} For the case of the high average power compression experiments outlined above, a CEP stable fiber chirped pulse amplification system is required. Current research efforts showed that this requires synchronized radio-frequency drivers for the employed acousto-optical pulse pickers\textsuperscript{[155]} resulting in the recent demonstration and stabilization of a 80 W, μJ-level fiber CPA system with subsequent nonlinear compression to 30 fs and less than 100 mrad CEP noise.\textsuperscript{[154]} These results should pave the way for achieving >100 W, sub-2 cycle pulses with stabilized CEP and high energy.

### 3.3. Summary and Overview on State-of-the Art High Average Power Few-Cycle Lasers Based on Nonlinear Compression

In summary, gas-filled-hollow-waveguide-based nonlinear compression can be considered as a flexible and versatile method for various pulse parameters (μJ-mJ), repetition rates and pulse durations, which can easily be addressed by choosing appropriate experimental conditions, such as fiber length, inner diameter, gas type and gas pressure. Table 2 summarizes parameters that have been achieved by state-of-the-art laser systems based on nonlinear compression. Obviously, compression in capillaries has been the method of choice for Ti:Sapphire based laser systems for many years. As shown in Table 2 the average power and pulse energy for CEP-stable few-cycle pulses have been steadily increased over the years. With the availability of high average power ytterbium-based laser systems new average power levels in both Kagome and capillary compressors are attainable today.

Moreover it has been proven that nonlinear compression is an average power scalable concept when using appropriate optical elements. Based on the presented findings the average power of few-cycle pulses could be increased by one order of magnitude as compared to current OPCPA systems (Tab. 2). As of now, it seems that nonlinear compression offers some favorable scaling properties and is the method of choice to address unprecedented performance levels of few-cycle lasers. It has been shown that it is expected to combine this outstanding performance with CEP stability further extending the potential application areas of such unique laser sources. In fact, the realization of the Extreme Light Infrastructure’s high repetition rate laser system (HR1), delivering 100kHz, 1mJ, sub-2 cycle pulses with stable carrier-envelope phase will be based on nonlinear compression of fiber lasers.\textsuperscript{[156]}

### 4. Selected Applications

Few-cycle laser pulses enable exciting studies on ultrashort timescales. High average power and high repetition rates are

**Figure 23.** Few-cycle pulses at high average power. (a) Spectrum (blue) and spectral phase (red) of the pulses after the second compression stage. (b) Temporal pulse profile of the compressed pulses obtained from (a). The inset shows the collimated beam profile. Reproduced from.\textsuperscript{[147]}
Table 2. Overview on current state-of-the-art high average power few-cycle lasers systems based on nonlinear compression

| Author [Ref.] | Wave-guide type | Central wavelength | Pulse duration (Fourier-Limit) | Pulse energy | Repetition-rate | Average power | CEP stability |
|---------------|----------------|--------------------|-------------------------------|--------------|----------------|---------------|---------------|
| Nisoliet al. [104] | Capillary | 0.8 μm | 5.0 fs | 0.5 mJ | 1 kHz | 0.5 W | no |
| Baltuska et al. [148] | Capillary | 0.8 μm | 5.0 fs | 0.5 mJ | 1 kHz | 0.5 W | < 50 mrad |
| Schultze et al. [105] | Capillary | 0.8 μm | < 4 fs | 0.4 mJ | 3 kHz | 1.2 W | < 100 mrad |
| Böhle et al. [125] | Capillary | 742 nm | 4 fs (2.6 fs) | 3 mJ | 1 kHz | 3 W | 360 mrad |
| Bohman et al. [155] | Capillary | 780 nm | 5.0 fs | 5 mJ | 1 kHz | 5 W | no |
| Mak et al. [111] | Kagome | 1 μm | 9.1 fs (5 fs) | 0.4 μJ | 38 MHz | 14.5 W | no |
| Rothhardt et al. [135] | Capillary | 980 nm | 6.3 fs | 170 μJ | 1.27 MHz | 216 W | no |
| Hädrich et al. [147] | Capillary | 980 nm | 6.3 fs | 170 μJ | 1.27 MHz | 216 W | no |

usually required when the efficiency of a process is very low or the count rates / statistics of a particular experiment have to be increased. This combination of short pulses and high average power not only brings existing experiments and applications to a new quality. It also enables novel exciting studies, which so far have not been possible.

4.1. High Harmonic Generation with Few-Cycle Pulses at High Repetition Rate

High harmonic generation (HHG) in noble gases offers the possibility to generate laser-like radiation in the XUV and X-ray spectral region. However, the conversion efficiencies are typically very low (< 10⁻⁶). Few-cycle pulses are beneficial for driving HHG since they allow the process to be driven at higher intensities without exceeding the critical ionization fraction which is crucial for phase-matching. Thus, few-cycle pulses allow for the highest cutoff energy (see Figure 24a) and the highest efficiency with a particular laser wavelength. Since modern few-cycle lasers can nowadays provide these pulses at very high average power, as presented in this article, a high photon flux in the XUV can be expected.

However, due to the limited pulse energy, which ranges from a few- to a few-hundreds of μJ for high repetition rate (> 10 kHz) few-cycle systems, tight focusing of the pulses is required to achieve the necessary intensity (> 10¹¹ W/cm²) for HHG. This tight focusing reduces the size of the generation volume and increases the Gouy-phase gradient along the propagation direction. However, both effects can be compensated by providing the generating medium with sufficiently high density. By focusing the pulses of a high repetition rate few-cycle OPCPA system into a xenon gas jet, a high conversion efficiency of 8·10⁻⁶ has been achieved, despite tight focusing to only 30 μm focal diameter. Although, only ~1 W of average power from the few-cycle laser system has been employed in these experiments, more than 10¹² photons/s have been generated already within the strongest harmonics H17 and H19 at ~25 eV. Figure 24 displays the generated photon flux versus the applied backing pressure at the gas nozzle delivering xenon gas to the interaction region. Interestingly, several maxima and minima can be seen in this pressure scan which can be employed to analyze the phase matching conditions.

Heavier noble gases allow to increase the cutoff energy and high harmonics have been generated up to the so-called water window (> 288 eV) with a high power few-cycle laser system. In future, the photon flux can be further increased by utilizing the high average power, which is available from cutting-edge few-cycle lasers. Thus, many application, which so far have been restricted to large-scale short wavelength light sources, such as synchrotrons and free-electron lasers, will be feasible with tabletop light sources in the future. The interested reader shall be directed to a comprehensive review article on high photon flux HHG sources.
4.2. Generation of Isolated Attosecond Pulses at High Repetition Rate

Isolated attosecond pulses (IAPs) are the fastest controllable electromagnetic events nowadays. Thus they allow investigating matter on shortest timescales and have become an indispensable tool for the investigation of ultrafast processes in atoms, molecules and solids.\(^{[12,163,164]}\)

Isolated attosecond pulses are generated by means of high harmonic generation in combination with suitable filter mechanisms, which restrict the harmonic emission to a single half-cycle of the driving fundamental laser field. A straightforward method is amplitude gating: Basically, the cutoff harmonics are spectrally selected by suitable metal filters and lower photon energies are suppressed. If few-cycle pulses with well-controlled CEP are used to drive this process, the electric field-strength strongly varies from half-cycle to half-cycle and the cutoff harmonics are exclusively generated by the most intense half-cycle of the driving field, which generates an isolated emission event with sub-fs pulse duration.

Certain applications, such as photoelectron spectroscopy or coincidence detection of ionization fragments, require high repetition rate sources of IAPs that so-far have not been available. The commonly used laser technology namely Ti:Sa lasers operated at \(\sim\)kHz repetition rates simply cannot provide the required driving pulses at higher repetition rates and thus higher average powers, due to thermo-optical constraints. Consequently, the repetition rate of IAP sources has not exceeded a few kHz so far.\(^{[105,163]}\)

As illustrated in the previous sections of this article few-cycle laser technology nowadays allows generating the required few-cycle pulses at up to MHz repetition rates with peak powers in excess of 1 GW. The experimental setup for the first experimental realization of isolated attosecond pulse generation with such a high repetition rate driving laser is shown in Figure 25.\(^{[165]}\)

For high harmonic generation, 14 \(\mu\)J pulses delivered by a high repetition rate OPCPA system have been focused by an off-axis parabola to a focal spot diameter of 30 \(\mu\)m x 35 \(\mu\)m. The target gas (argon) is emerging from a 150 \(\mu\)m-diameter nozzle backed with several bar of pressure. The harmonic signal has been analyzed by a flat-field grating spectrometer. The backing pressure and the position of the gas jet have been optimized for the highest harmonic signal at \(\sim\)60 eV photon energy. In addition, the laser intensity has been fine-tuned by an iris placed at the entrance of the HHG chamber to adjust the cutoff energy to be located between 60 eV and 70 eV. A spectral bandpass between 55 eV and 73 eV has been created by placing a 200 nm thick Zr and a 200 nm thick Al filter into the XUV beam path.

The CEP of the laser pulses has been controlled and scanned by a pair of fused silica wedges. Two representative harmonic spectra are illustrated in Figure 26. Figure 26a) displays a spectrum recorded for a certain CEP \(\phi_0\). It shows distinct harmonic lines indicating the generation of an attosecond pulse train. In contrast, if the CEP is changed by \(\pi/2\) the spectrum changes to a continuous shape, as shown in Figure 26b), indicating the generation of a single isolated attosecond pulse. These findings are supported by a numerical simulation of the HHG experiment. More details on the methods can be found in.\(^{[165]}\)

4.3. Studies of Strong-Field Ionization with Coincidence Detection

Few-cycle pulses delivered by a 400 kHz repetition rate OPCPA system\(^{[99,173]}\) have been utilized for strong-field ionization of argon. For this purpose, the 6 fs pulse has been focused to \(\sim 7 \times 10^{13}\) W/cm\(^2\) peak intensity. Coincidence detection of the released electrons and the ionic fractions allow for discrimination of electrons origination form single- and double-ionization from argon, argon dimers and the background gas. The electron momentum map corresponding to single ionization of argon atoms is displayed in Figure 27.

It shows a clear fan-like structure which for electrons with very low kinetic energy has been associated with the interference of classical electron trajectories leaving the atom at different times during the pulse evolution.\(^{[174,175]}\) The fine structure for momenta \(|p| > 0.2\) a.u. is identified as resonantly enhanced ionization through intermediate Rydberg states, whereas the reasons for the huge number of minima for \(|p| > 0.3\) a.u. is not yet clear.\(^{[173]}\)

Remarkably, this map has been extracted from \(10^7\) events, which have been acquired in only 40 minutes thanks to the high repetition rate of the laser system. In future, the CEP dependence of strong-field ionization can be studied by such experiments in great detail.
4.4. Towards Ultrafast Dynamics with Pump-Probe Experiments

Ultrafast dynamics in atoms, molecules or ions can be investigated by few-cycle laser pulses. One particular class of studies are pump-probe experiments, which in a first step excite the system and in a second step probe the system with a delayed pulse.

For example, charge transfer processes in molecules can be investigated by applying synchronized pump pulses in the infrared and probe pulses in the XUV. Such studies have e.g. been performed on iodomethane molecules at the free-electron-laser LCLS. Few-cycle lasers and high harmonic generation can in principle enable such experiments on a lab scale and allow accessing much faster dynamics, due to the short pulse duration and the inherent synchronization of the fundamental laser and its harmonics.

The detection of all molecular fragments in coincidence, as introduced in the previous section, is crucial for gaining a maximum information of the investigated processes. To keep the detector counts per laser pulse low enough, a high repetition rate XUV source is needed for such experiments. This combination has recently been demonstrated by inner-shell excitation of iodomethane molecules with a 70 eV high-photon-flux HHG source. The HHG source is based on a high-power fiber laser system with subsequent nonlinear compression. Coincidence detection of all fragments has been achieved. Figure 28 displays a photoion-photoion-coincidence (PIPICO) map showing the momentum of the iodine (I+) fragments versus the momentum of the CHx+ fragments. Due to momentum conservation, these coincidence events, which resulted from a (quasi-) two-body fragmentation clearly lie on diagonal lines in the PIPICO spectrum. This experiment demonstrates the feasibility of such studies, but the integration time for recording the displayed PIPICO spectrum was as long as 20 hours. In future, the XUV photon flux has to be further increased to enable pump-probe experiments with reasonable integration times. High average power few-cycle lasers will be the ideal choice for this purpose and enable exciting studies on few-femtosecond to attosecond time scales.

5. Conclusion & Outlook

Few-cycle laser technology has come of age. Due to the recent advances in Yb-based solid state laser technology, femtosecond laser...
pulses are nowadays available at much higher average power and repetition rate. Optical parametric chirped pulse amplification and nonlinear pulse compression are two complementary methods to transfer the high average power of Yb-based femtosecond lasers to few-cycle pulses. OPCPA provides few-cycle pulses with very good CEP stability and average powers up to 22 W at 1 MHz repetition rate[36] and up to 53 W at 1 kHz repetition rate.[97] The concept is particularly well suited to provide wavelength-tunable pulses for spectroscopic applications[178,179] and to explore wavelength ranges that lack of powerful lasers. However, their power scaling is hindered by thermo-optical effects arising from absorption of the interacting waves.

Nonlinear compression employing noble-gas-filled hollow capillaries represents a promising alternative to overcome these limitations. It demonstrated few-cycle laser pulses with more than 200 W of average power already[180] and scaling beyond 1 kW of average power seems feasible.[132] In future, the stabilization of the CEP of such laser systems will be achieved to ultimately meet the requirements of ambitious projects such as the ELI-ALPS HR laser beamline.[156]

High power few-cycle laser systems have already enabled unique applications such as efficient high harmonic generation[147,159] the generation of isolated attosecond pulses at up to 0.6 MHz repetition rate.[165] Furthermore, the high repetition rate of these systems enabled coincidence detection of fragments after strong-field ionization at 0.4 MHz repetition rate[173] and recording photoion-photoion-coincidence spectra of molecular fragments after inner-shell XUV excitation.[172] In the XUV, the spectral region around 2 μm has already been explored extensively. High average power of modern few-cycle lasers and secondary sources will benefit from the increased average power of modern few-cycle lasers and secondary sources in the XUV.

Moreover, few-cycle lasers at longer wavelengths will gain increasing importance. The spectral region around 2 μm wavelength has already been explored extensively. High average power OPCPA systems, delivering 6 W of average power have been demonstrated in this spectral region already[183] but also uncovered thermo-optical limitations in further power scaling. As an alternative Ti:sapphire-based lasers[186–190] in combination with nonlinear compression will provide energetic few-cycle pulses with average powers approaching the 100 W-level soon and can be employed as pump lasers for longer mid-IR wavelength systems based on parametric frequency conversion. A detailed overview on high average power few-cycle lasers in the mid-IR will be provided in a separate review article soon.

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Conflict of Interest

The authors have declared no conflict of interest.

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