New Journal of Physics
The open-access journal for physics

Short-pulse optical parametric chirped-pulse amplification for the generation of high-power few-cycle pulses

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New Journal of Physics **9** (2007) 438
Received 14 September 2007
Published 12 December 2007
Online at http://www.njp.org/
doi:10.1088/1367-2630/9/12/438

Abstract. We report ultrabroadband optical parametric chirped-pulse amplification (OPCPA) with an output pulse energy of up to 250 μJ from an OPCPA stage pumped by short pulses of ∼100 fs duration at 395 nm wavelength. In order to generate ultrahigh-power pulses in the few-cycle regime, such a short-pulse-pumped OPCPA scheme appears to be a promising route, by virtue of its inherently advantageous features. Firstly, the stretching and compression fidelity as well as the pulse contrast are increased due to the short pump- and seed-pulse durations. Additionally, the higher pump powers allow for using thinner OPA crystals, thereby increasing the amplification bandwidth that will support

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PII: S1367-2630(07)60024-X

even shorter pulse durations. We present experimental results where the effective bandwidth of the seed pulses was increased in the OPCPA process resulting in a shortened transform-limited pulse duration in addition to the energy gain. The amplified pulses from OPCPA have been compressed to the sub-10-fs, few-cycle range by using chirped mirrors. Scaling of this short-pulse-pumped OPCPA technique for few-cycle-pulse generation to the highest (TW–PW) power levels is also planned (Petawatt Field Synthesizer project at the Max-Planck-Institut für Quantenoptik).

Contents

1. Introduction

Few-cycle laser pulses with unprecedented peak powers are crucial for the investigation of laser-driven strong-field phenomena [1] having become an accessible field of research with the advent of suitable laser systems. Conventional laser-amplification technology can provide very high power levels, but the pulse duration determined by the laser material is limited to the tens-of-femtoseconds range. However, in recent years the technique of optical parametric amplification (OPA), which does not suffer from this limitation and allows for very large amplification bandwidths, has opened up a new path towards generating ultra-high power, few-cycle laser pulses [2]–[9].

In the OPA process amplification takes place in a nonlinear optical crystal, where phase matching in noncollinear geometry allows for extremely large amplification bandwidths [2], and thereby enables the amplification of broadband seed pulses using relatively narrowband pump pulses delivered by conventional laser-amplification technology. This amplification bandwidth can be significantly larger than that of any known laser-amplifier medium, constituting the main advantage of OPA over conventional laser technology. In addition, the gain can be very high even in a short length of material, which results in a lower B-integral, i.e. the accumulation of nonlinear phase by passing through material. Furthermore, in contrast to laser amplifiers no energy is converted into heat, thereby eliminating thermal distortions of the beam.

In order to reach the highest possible peak intensities, in high-power systems OPA is usually combined with chirped-pulse amplification (CPA), called optical parametric chirped-pulse amplification (OPCPA), a scheme first proposed by Dubietis *et al* [10]. In such a system pulse energies as high as 35 J were achieved in 84-fs pulses [11]. In the few-cycle regime 10-fs pulses with 90 mJ energy were demonstrated from an OPCPA system [12]. In typical ultrabroadband OPCPA systems the duration of pump pulses obtained from a Nd : YAG laser are usually on the order of 100 ps [8, 9, 12]. Some smaller-scale systems use shorter pump pulses of ∼100 fs duration and were able to generate sub-10 fs pulse durations, however, only with energies in the microjoule range [5]–[7], [13]–[15]. Recently, 300 μJ output pulse energy has been reported from an OPCPA system [16] pumped by 150 fs pulses, however, the signal pulse duration was about 20 fs.

We believe that OPCPA pumped by such short pulses could be a promising route towards boosting the output pulse energy up to the Joule level while keeping the shortest pulse durations. In order to combine few-cycle pulse durations with highest pulse energies we propose to pump an OPCPA system with TW-scale short pulses (100 fs to 1 ps instead of $\sim$100 ps of previous OPCPA systems) delivered by a conventional CPA system. This approach improves the conditions for high-power few-cycle pulse generation as compared to long-pulse-pumped OPCPA in several ways. Firstly, the short pump-pulse duration reduces the necessary stretching factor for the seed pulse, thereby increasing stretching and compression fidelity and allowing the use of simple, high-throughput stretcher-compressor systems, consisting of a block of glass for stretching and chirped multilayer mirrors for compression. Secondly, the short pump-pulse duration results in a short amplification time window and hence a dramatically enhanced pulse contrast outside this window. Finally, the significantly increased pump power permits the use of thinner OPA crystals, which implies an increase of amplification bandwidth as compared with OPA driven by longer pulses.

However, this novel approach also imposes a number of challenges that need to be overcome. Firstly, as in the case of long-pulse OPCPA the amplification sensitively depends on the uniformity of the pump-beam profile. In addition, for such short pulses the constraints on the pump–signal synchronization are even stricter than with $\sim$100 ps or longer pulses. Furthermore, the matching of pump and signal pulse fronts has to be considered, which is critical especially in OPCPA systems with sub-ps pump-pulse duration and with relatively large beam diameters.

Short-pulse-pumped OPCPA (SPP-OPCPA) systems in the few-cycle regime reported to date (cf [5]–[7], [13]–[15]) were pumped by microjoule-scale pulses. Here, we present first experimental results towards verifying the scalability of the SPP-OPCPA concept to higher power levels. Our proof-of-principle study draws on the ATLAS TW-scale Ti : sapphire CPA laser for both seeding and pumping of an OPCPA chain. Our system design consists of three OPA stages which aim for an amplified output pulse energy in the range of 50–100 mJ. Currently, the first stage is in operation and we present our results and findings in terms of amplification and pulse compression.

2. Experimental set-up

The experimental set-up is shown in figure 1. The ATLAS Ti : sapphire laser system was used as a drive laser for our OPCPA stage. In ATLAS the nanojoule-level pulses of a broadband oscillator (Femtolasers) are preamplified in a multipass Ti : sapphire amplifier stage by a factor of $\sim 10^4$. The preamplified pulses are used for seeding a regenerative amplifier which boosts the pulse energy to almost 20 mJ with a bandwidth of about 30 nm around the 790 nm central wavelength. The pulses are then further amplified in a first post-amplifier stage to $\sim$500 mJ and compressed in vacuum by a grating compressor. The compressed output pulses have an energy of 350 mJ ($\pm$5% rms) and a pulse duration of 43 fs ($\pm$3 fs) at a repetition rate of 10 Hz.

Both seed and pump pulses for OPCPA were derived from the ATLAS laser. This optical synchronization was essential since on a timescale of the pulse durations, i.e. $\sim$100 fs, no electronic method could have provided sufficient accuracy. The strict requirements on femtosecond-scale synchronization of the OPA process were still met even after separately

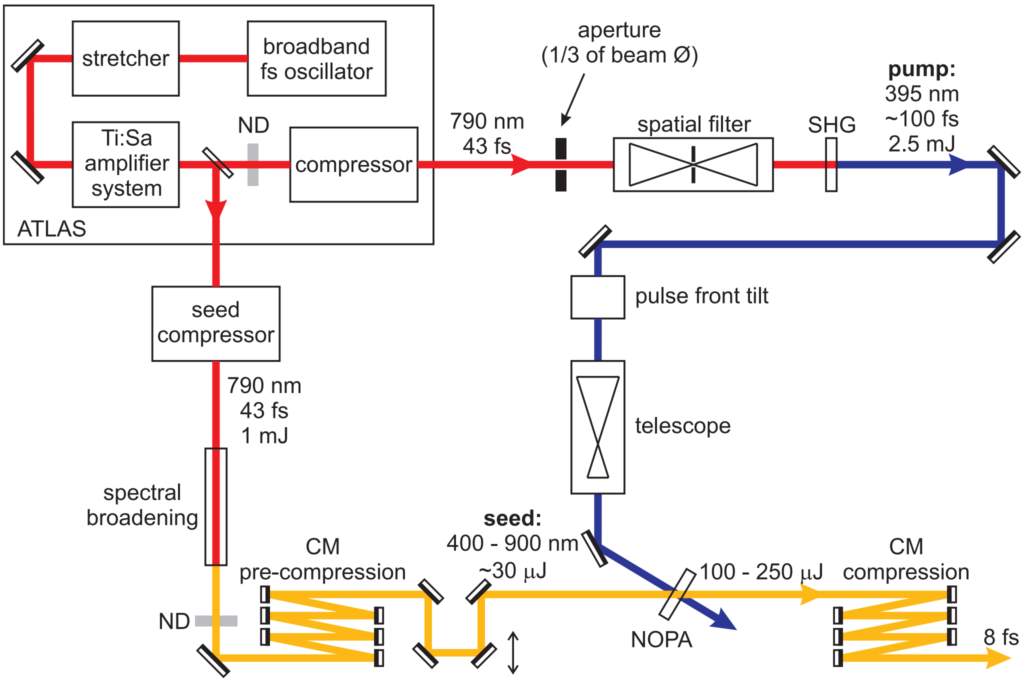

Figure 1. Schematics of the experimental set-up. ND: neutral density filter, CM: chirped mirror.

propagating the two beams along path lengths of about 20 m before overlapping them in the OPA crystal.

In order to provide a seed beam for OPCPA a small fraction of the ATLAS pulse energy was split off by a glass plate inserted into the uncompressed beam just before the compressor chamber. The split-off pulses were compressed by a separate grating compressor from ∼350 ps to ∼43 fs. The compressed pulses were focused into a gas cell by a 1.75-m focal length mirror for spectral broadening. The cell was filled with 1.4 bar of Ar gas. By using 1 mJ input pulse energy in a 1-cm diameter beam and an iris diaphragm for aperturing the incoming beam, a stable filament was generated. The output pulse energy was 800 μJ. A typical broadened spectrum is shown in figure 2. A comparison with the input spectrum reveals a broadening factor of about four, giving a transform-limited pulse duration of about 10 fs which is in good agreement with the results of Hauri *et al* [17] obtained for similar parameters. Behind the gas cell the seed beam was recollimated by a 1.5 m focal-length mirror.

For our amplification experiments it was of crucial importance that, apart from the intense part between ∼750 and ∼850 nm, the broadened spectrum also consisted of a very broad but weak tail with an exponentially decreasing intensity extending on the short-wavelength side down to 400 nm, which is shown in figure 2(a). The filament also acted as a spatial filter resulting in an excellent output beam quality (cf figure 2(b)). We used the spectral phase interferometry for direct electric field reconstruction (SPIDER) [18] technique for characterizing our seed pulses. According to these SPIDER measurements the seed pulse duration at the position of the OPA crystal was about 100 fs. Since this was comparable to the duration of the pump pulse, in some cases chirped-mirror precompression was used in order to avoid spectral narrowing in the OPA process.

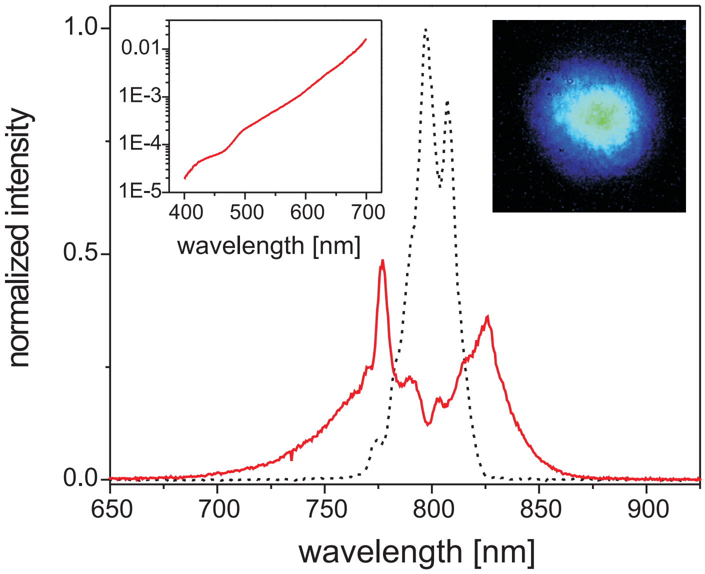

Figure 2. The seed spectrum before (dashed line) and after (solid line) broadening. Inset (a) shows the short-wavelength tail of the broadened spectrum in a semi-logarithmic plot. Inset (b) shows the beam profile after the plasma filament.

The OPCPA stage was pumped by frequency doubling a small fraction of the output of the ATLAS laser (figure 1). Since in the OPA process the resulting gain sensitively depends on the pump intensity [19]–[21], it was of fundamental importance to provide a pump beam with a sufficiently smooth beam profile. This is critical especially in case of large gain factors typically obtained in the first stage of an amplifier chain. Therefore, efforts were made to generate a pump with a beam profile as uniform as possible with a pulse energy sufficient for our first OPCPA stage.

From earlier experiments [22] it was clear that using the frequency-doubled output of ATLAS would not meet the stringent requirements on the uniformity of an OPA pump beam without any further precautions. In order to improve the spatial profile of our pump beam we have inserted a soft aperture (diameter $\sim 1/3$ of the beam diameter) and a vacuum spatial filter into the beam path before the second-harmonic generation (SHG) crystal. The filtering element was a cone-shaped quartz capillary pinhole [23] which allowed for stable operation over an extended period of time owing to its high damage threshold. With the current set-up we were able to obtain a pulse energy in the fundamental beam of about 6 mJ with an acceptable beam profile. Subsequent frequency doubling in a 2-mm-thick KDP crystal with ~40% conversion efficiency gave 2.5 mJ pulse energy at 395 nm. The bandwidth of the second harmonic (SH) pulses was ~ 3 nm, limited by the relatively thick KDP crystal, yielding an estimated pulse duration of 85 fs.

In order to obtain an amplification bandwidth as large as possible a noncollinear geometry was used for the parametric amplification [2]. This geometry along with short pump and seed-pulse durations and the relatively large beam diameters of a few millimetres, required matching of the pulse fronts of the pump and the seed beams by tilting one of them [6, 13].

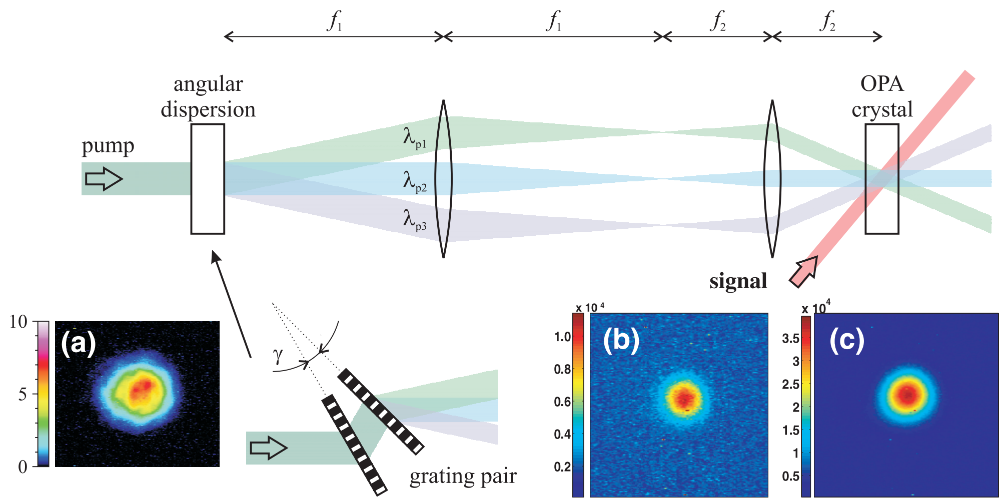

Figure 3. Pulse-front matching between pump and signal using a pair of transmission gratings. In the experiment, the telescope consisted of two concave mirrors, which, for clarity, have been substituted by lenses in the schematic picture. The beam profiles shown are (a) the ∼1 mJ pump beam before entering the transmission gratings; (b) the focused seed profile before and (c) after amplification. Note that the images (b) and (c) are not shown on the same scale, but are adjusted to the same maximum value.

We investigated the following two routes for the pulse-front matching by tilting the pulse front of the pump beam: (i) a 45°-apex-angle prism followed by a reflective Galilean telescope; (ii) a pair of transmission gratings followed by a reflective Keplerian telescope. Approach (i) allowed us to use higher pump energies due to the absence of a focus in the telescope. However, it led to a spectral lateral walk-off [13] at the crystal which also affected the amplification process (i.e. by resulting in a spatial chirp in the amplified signal).

The schematic of the alternative approach (ii) is shown in figure 3. The gratings used are fused-silica transmission gratings with 2500 lp mm^{-1} groove density optimized for high diffraction efficiency (>90%) into the -1st order. They were fabricated by electron beam lithography and reactive ion beam etching similar to those in [24]. The two gratings are used in tandem with a small tilt angle between them. Tuning the tilt angle allows an easy variation of the residual angular dispersion and hence a tuning of the pulse-front tilt. The distance between the gratings was kept at minimum to suppress temporal pulse-distortion effects due to dispersion and spatial chirp. The role of the telescope behind the gratings is two-fold. Firstly, it eliminates spectral lateral walk-off at the OPA crystal by imaging the grating surface on to the crystal (cf approach (i)). Secondly, a demagnifying telescope also reduces the beam diameter. Hence, the intensity can be kept at a low value at the gratings and at the same time increased to a much higher value in the OPA crystal. This reduces the B-integral in the grating substrates and also the risk of damage to them on one hand, and allows for high gain and efficient amplification in the crystal on the other hand.

We note that using a single grating rather than a pair of gratings would suffice for pulse-front tilting but would lack tunability. In our experiment, however, the ability to tune the pulse-front tilt was of great importance as it allowed us to vary the pump intensity on the

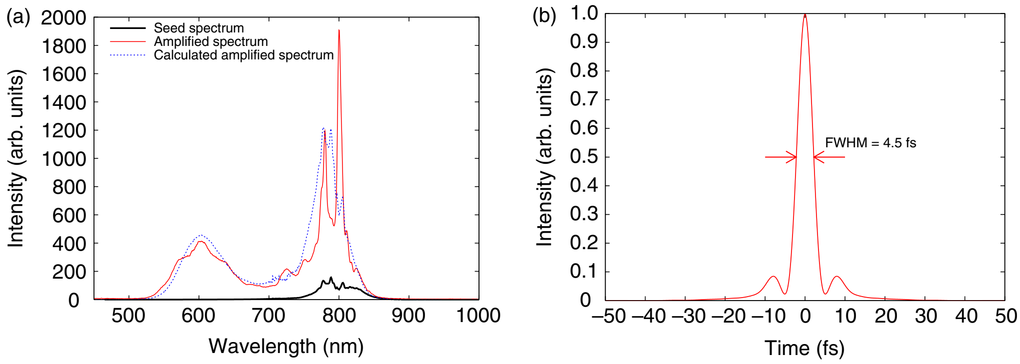

Figure 4. (a) Spectra of seed (black) and amplified signal (red) from our experiment. Simulation results for the amplification based on the measured seed spectrum as input are shown as well (blue/dashed). (b) Transform-limited pulse duration corresponding to the measured amplified signal spectrum shown in (a).

OPA crystal by changing the telescope parameters, while keeping the pulse fronts matched in the same set-up and without having to alter the beam size and intensity on the gratings.

3. Results and discussion

In our OPCPA experiments we have used BBO crystals of different thicknesses (0.5, 1 and 2 mm) with all of which we were able to observe ultrabroadband amplification. The amplified signal energy varied between 100 and 250 μJ depending on the amplification range and bandwidth, which was tunable by changing the orientation of the crystal and thereby the phase-matching condition.

The noncollinear angle between pump and seed was close to $\alpha = 3.7^\circ$ inside the crystal set for type I phase matching in the visible and near-infrared spectral range with the broadest possible bandwidth. By changing the crystal orientation the amplified region of the spectrum could easily be tuned. The diameter of the pump beam at the OPA crystal was about 3 mm. In order to prevent optical damage at the last telescope mirror and at the OPA crystal the pump energy was attenuated to $\sim$1 mJ. With 85 fs pulse duration this gave a pump peak intensity of about 150 GW cm^{-2}. Owing to the limited pump energy used in this experiment the seed energy was also attenuated to $\sim$30 μJ before entering the OPA crystal.

Figure 4(a) shows a typical amplified signal spectrum obtained with the 1-mm BBO crystal. It shows a very large amplification bandwidth ranging from below 550 nm to above 800 nm. The spectrum corresponds to a Fourier-limited pulse duration of less than 5 fs (cf figure 4(b)). In this case the pulse-front matching was achieved by using a 45°-apex-angle-BK7 prism (geometry as in approach (i) in section 2). The pump energy was 560 μJ, the seed energy was approx. 20 μJ, which resulted in an amplified pulse energy of approx. 150 μJ. Each of these energies were subject to shot-to-shot fluctuations of about 7% (rms).

Comparing the seed and the amplified signal spectra in figure 4(a) clearly indicates the large variations in gain between different spectral ranges. Between 750 and 850 nm where the spectral intensity of the seed is large, we see a gain on the order of 3–4, whereas in the weak exponential tail below 700 nm (cf figure 2) the gain is several orders of magnitude larger. The reason for this is that for different seed intensities along the spectrum the amplification takes

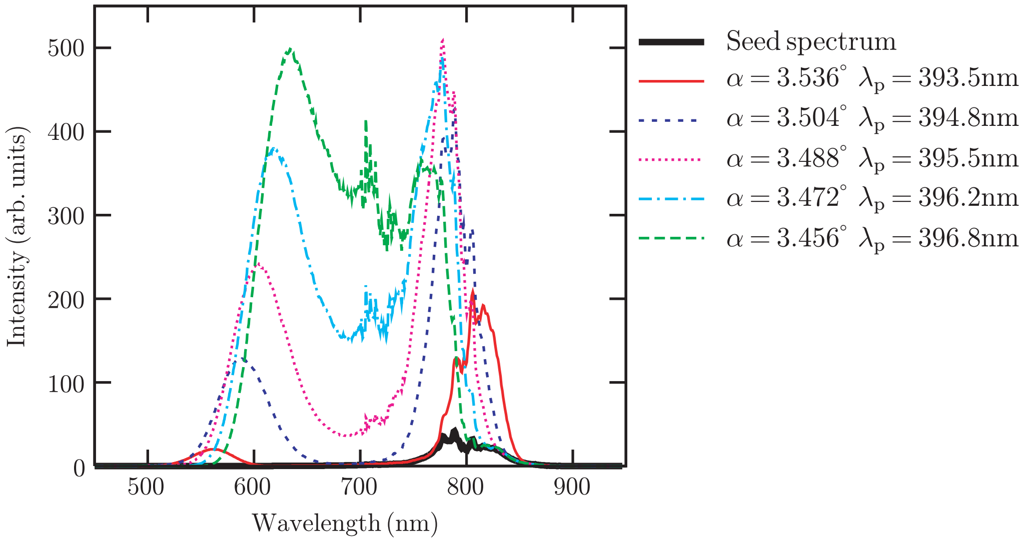

Figure 5. Calculated amplified spectra for a set of different noncollinear angles α and pump wavelength λ_p corresponding to the angular chirp and bandwidth of the pump pulse in the experiment using the experimental seed spectrum as input. The noisy peak at 700 nm is an artifact of the measurement of the short-wavelength tail of our seed spectrum.

place in different regimes, i.e. while the amplification is strongly saturated around the 800 nm peak, the short-wavelength wing, being several orders of magnitude smaller, is unsaturated and experiences a much larger (small-signal) gain. The important consequence of the resulting enhanced bandwidth is the considerably reduced transform-limited pulse duration (from about 10 fs for the seed to below 5 fs for the signal) as demonstrated in figure 4(b).

In order to understand the origin of the exact shape of the amplified signal spectrum, we have performed numerical calculations of the OPA gain using a simplified 1D model [19]. For the calculations we used the measured seed spectral intensity as an input. Our model assumes a monochromatic pump pulse, however, we superimposed several such monochromatic calculations in order to account for the bandwidth and the angular chirp of the pump pulse to a first approximation. The dashed line in figure 4(a) shows the resultant amplified spectrum for one particular set of noncollinear and phase-matching angles, which are close to the experimental values. It is clear from figure 4(a) that for this choice of noncollinear and phase-matching angles a dip in the middle of the spectrum (around 680 nm) appears, allowing for a good qualitative agreement between experiment and calculations. In addition, the calculation reproduces the saturated (around 800 nm) and unsaturated (below 700 nm) gain regimes as observed in the experiment, for input parameters close to those in the experimental set-up.

As mentioned above, our model assumed a monochromatic pump pulse, and therefore is not able to account for the possible effects of the angular chirp introduced by the pulse-front matching. For a qualitative demonstration of how sensitive the shape of the amplified spectrum is to the choice of the pump wavelength λ_p coupled with the angle α, in figure 5 we show the monochromatic calculations at a number of $\alpha-\lambda_p$ pairs, the superposition of which resulted in the calculated amplified spectrum in figure 4(a). The $\alpha-\lambda_p$ pairs correspond to the experimental values for the pump-pulse bandwidth (FWHM of 3 nm) and angular chirp originating from the

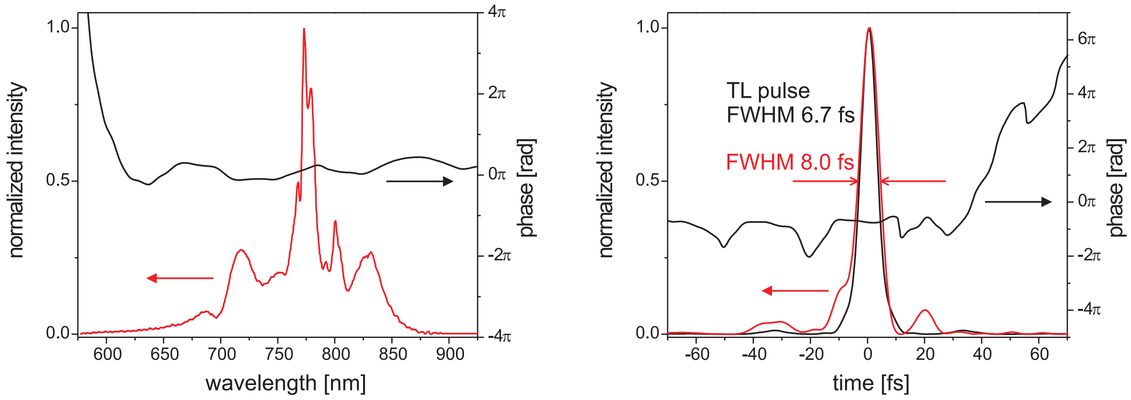

Figure 6. Reconstructed spectral (left) and temporal (right) phase and intensity of amplified and compressed signal pulses.

pulse-front tilt. We can clearly see that over this range of pump-pulse parameters the shape of the amplified spectrum changes from being almost flat in the centre (around 700 nm) to having a dip in the same way as we observe it in our experiment, indicating an extreme sensitivity to the exact choice of parameters.

In order to test the compressibility of the amplified spectrum the output pulses were reflected off a set of chirped mirrors and characterized by our SPIDER apparatus (cf section 2). In these measurements the thickness of the OPA crystal was chosen to be 2 mm, because of the larger output energy. Owing to the limited bandwidth of the chirped mirrors on the short-wavelength side, the OPA crystal was tuned to a narrower amplification bandwidth centred around 770 nm. The amplified spectrum (cf left panel of figure 6) in this case supported a transform-limited pulse duration of 6.7 fs. The pump, seed and resulting amplified energies were 370, 60 and 150 μJ, respectively. The results of the pulse reconstruction can be seen in the right panel of figure 6. The reconstructed spectral phase is nearly flat across the pulse spectrum indicating a good compression. A Fourier transform into the time domain reveals a compressed amplified-pulse duration of 8.0 fs which is reasonably close to the transform-limited pulse duration of 6.7 fs.

It shall be noted that pulses delivered by the first stage of our OPCPA chain based on the ATLAS system do not yet represent a new regime in terms of pulse duration and achieved peak powers. Alternative methods, such as the compression of a spectrally broadened hollow-fibre output [25], have been shown to deliver pulses in the few-cycle regime with a pulse duration of down to 5.6 fs and pulse energy up to 1.2 mJ [26]. However, the scalability of these methods is a known limitation if PW-power levels are considered.

We also investigated how the amplification and pulse-front matching affected the focusability of the signal pulse. This is particularly important when future applications of such a system are considered, such as the investigation of high-field laser-matter interactions. Figure 3(b) shows the far-field beam profile of the seed before (left) and after (right) amplification. It is clear that the excellent beam quality delivered by the plasma filament is not visibly distorted in the amplification process using the pulse-front-matched pump beam.

4. Conclusion

In summary, we have demonstrated ultrabroadband OPCPA with an output energy on the 100 μJ level pumped by short pulses ($\sim$100 fs) and compression of the amplified pulses to the sub-10-fs, few-cycle regime. The reported experiments constitute the first step towards a multi-TW, few-cycle SPP-OPCPA system.

In order to scale up such short-pulse OPCPA to higher output energies, high-power pump lasers are necessary, which are capable of delivering pulses with a very good spatial uniformity of the beam that meet the strong requirements of OPCPA pumping. However, as the system size increases, it also becomes more and more challenging to provide a suitable pump beam. The ATLAS-pumped OPCPA system is expected to become a multi-TW-level milestone on the way towards boosting the power of few-cycle optical pulses to the PW level (Petawatt Field Synthesizer project at MPQ [27]) based on the short-pulse OPCPA concept. This large-scale system is planned to be pumped by few-ps pulses, an intermediate regime where the advantages of the short-pulse OPCPA concept will be combined with less critical requirements, e.g. for pulse-front matching, making the system design more robust.

Acknowledgments

Financial support from the Marie-Curie Individual Fellowship no. MEIF-CT-2005-024150 (ZsM) as well as the PFS grant of the Max-Planck Society is acknowledged.

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