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Utilizing the temporal superresolution approach in an optical parametric synthesizer to generate multi-TW sub-4-fs light pulses

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Abstract: The Fourier-transform limit achieved by a linear spectral phase is the typical optimum by the generation of ultrashort light pulses. It provides the highest possible intensity, however, not the shortest full width at half maximum of the pulse duration, which is relevant for many experiments. The approach for achieving shorter pulses than the original Fourier limit is termed temporal superresolution. We demonstrate this approach by shaping the spectral phase of light from an optical parametric chirped pulse amplifier and generate sub-Fourier limited pulses. We also realize it in a simpler way by controlling only the amplitude of the spectrum, producing a shorter Fourier-limited duration. Furthermore, we apply this technique to an optical parametric synthesizer and generate multi-TW sub-4-fs light pulses. This light source is a promising tool for generating intense and isolated attosecond light and electron pulses.

1. Introduction

The generation of intense ultrashort laser pulses is important for various fields in basic science like femtochemistry [1], attosecond physics [2] and laser-plasma physics [3] or applied research like material processing [4]. For the production of these short light pulses, it is crucial to precisely control their spectral amplitude and phase. The technique of chirped pulse amplification (CPA) revolutionized the generation of high intensity laser pulses via controlling the spectral phase [5]. CPA lasers with titanium sapphire as laser medium are the most prominent example supporting down to 20-25 fs duration (8-10 optical cycles). To generate even shorter pulses, it is intensely explored how to extend the spectral bandwidth, via broadband amplification [6] or broadening techniques after amplification, called post-compression [7]. An alternative light amplification technique, optical parametric chirped pulse amplification (OPCPA), supports larger spectral bandwidths than any conventional laser medium reaching the few-cycle temporal regime [8,9]. Moreover, an optical parametric synthesizer (OPS) is utilizing multiple OPCPA stages that amplify different spectral regions [10]. Therefore, OPS makes the generation of intense pulses with sub-1–2 cycle duration possible [6,10,11].

For a broadband laser a constant or linear spectral phase leads to the highest peak intensity. Hence, high intensity laser systems commonly try to reach a linear spectral phase. Furthermore, the root-mean-square (RMS) pulse duration for a given spectrum is the smallest in the case of a linear spectral phase and hence, the RMS duration fulfills the well-known uncertainty relation for the time-bandwidth product [12]. Therefore, laser pulses with a linear spectral phase are referred to as Fourier-transform limited (FL).

However, for many applications in photonics the full width at half maximum (FWHM) of the pulse duration is more important than the RMS duration. This FWHM duration for a laser pulse with a certain spectrum can be shorter than the FL, i.e., sub-Fourier limited (sub-FL). We

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refer to temporal superresolution (TS) [12–15] as an approach to shorten the FWHM via spectral phase shaping (SPS) by creating sub-FL pulses, or alternatively via spectral amplitude shaping (SAS) by cutting the spectrum to reach a new, shorter FL duration. Both ways decrease the peak intensity characterized by the temporal Strehl ratio (I_{max}), i.e., peak intensity ratio of the shaped and original calculated FL pulse. TS was used so far to shorten the duration of laser pulses from oscillators [16] as well as conventionally amplified lasers [13]. However, the low pulse energy of the oscillators and the long duration of the amplified laser pulses hampered the applicability of those laser sources.

In this article, we present the implementation of TS in a multi-TW laser based on OPCPA as well as OPS. This allows us to generate laser pulses with less than 4 fs FWHM duration (~1.6 optical cycles) and a peak power of multiple TW. To the best of our knowledge, this is the most powerful sub-4-fs laser in the world.

The high power of the light source together with the modulated temporal structure of the obtained pulses are naturally tailored for nonlinear applications, in which the most intense sub-FL main peak dominates the interaction.

2. Experimental implementation and results

For the experiments presented in this article a multi-TW sub-5-fs laser system, the Light Wave Synthesizer 20 (LWS-20), was used [10]. This system (shown in Fig. 1) consists of a front end (Pharos & Orpheus, Light Conversion), which provides low energetic (15 µJ) laser pulses with a spectrum ranging from 580–1020 nm. Those pulses are temporally elongated with a grism (prism + grating) stretcher [17]. Then the light pulses pass through an acousto-optic programmable dispersive filter or shaper (Dazzler, Fastlite), which can change their spectral amplitude and phase. This shaper was used in the experiments to achieve compression of the laser pulses close to the FL as well as for the implementation of TS. Afterwards, the pulses are amplified in a two-color pumped (532 nm, 355 nm) OPS consisting of four OPCPA stages. Then the stretched pulses are temporally compressed by bulk material and chirped mirrors. The temporal characterization was accomplished using the chirp-scan technique [18,19]. The retrieval of the spectral phase from the measured chirp scan traces was accomplished via minimizing

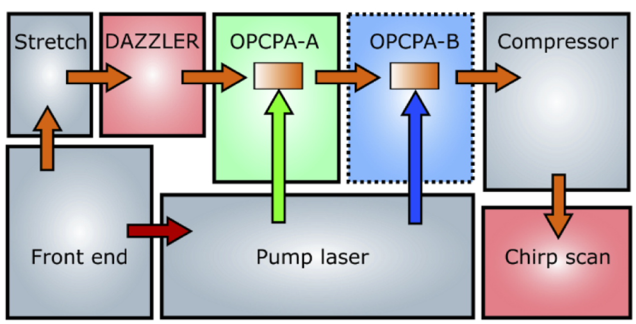

Fig. 1. Sketch of the experimental setup. Broadband laser pulses (580 - 1020 nm) from a commercial front end are temporally elongated with a grism stretcher (Stretch) and an acousto-optic programmable dispersive shaper (Dazzler). Then they are amplified in a two-color-pumped (532 nm, 355 nm) OPS (OPCPA-A for 532 nm and OPCPA-B for 355 nm) and temporally compressed with a bulk material compressor and chirped mirrors. For the implementation of spectral amplitude shaping as well as spectral phase shaping the Dazzler was used and the pulses were temporally characterized in a chirp scan setup. For the experiments shown in Fig. 2 and Fig. 3, only the green pumped OPCPA stages were in operation.

the error between a simulated chirp scan trace and the measured one with an genetic algorithm [20]. The implementation of TS via phase or amplitude shaping generates side peaks in the time domain that we optimized to have an intensity of <0.5 of that of the main peak.

To realize and study TS for an OPCPA system pumped with a single color, first, just the green pumped amplification stages of the LWS-20 system were used. It is highly relevant as many existing OPCPA systems are working with similar spectral bandwidth [21–23]. This supports a FL FWHM pulse duration of ~7.5 fs. With the shaper and without TS a FWHM pulse duration of 200-400 as above the FL could routinely be reached, as shown in Fig. 2.

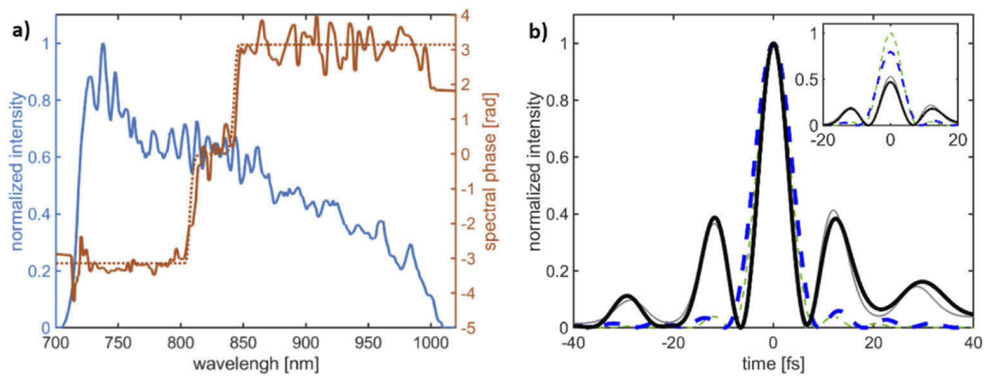

Fig. 2. Spectral phase shaping (SPS) to optimize the FWHM pulse duration of an OPCPA system. a) Spectral intensity and phase; blue: laser spectrum; dotted red: applied phase shift for SPS; red: measured spectral phase with SPS. b) Temporal intensity; thin dashed green: FL of the laser spectrum (FWHM: 7.48 fs); thick dashed blue: measured compressed laser pulse without SPS (FWHM: 7.9 ± 0.1 fs (105 ± 1%), I_{max}: 80 ± 1%); thin gray: theoretical laser pulse calculated with the applied phase with SPS (FWHM: 6.32 fs (85%), I_{max}: 53.4%); thick black: measured laser pulse with SPS (FWHM: 6.4 ± 0.1 fs (86 ± 1%), I_{max}: 48 ± 3%). The % value after the pulse duration is the ratio to the FL of the spectrum, while the I_{max} value is the temporal Strehl ratio, i.e., the intensity decrease to the FL. The inset on the right upper side shows the temporal intensity of the same four pulses, all normalized to the maximum of the FL pulse.

To implement TS, first, SPS was used by employing a spectral phase with the shaper additionally to the phase for temporal compression. The used added spectral phase is defined in Eq. (1).

$$\varphi(\lambda) = \begin{cases} \left(e^{\ln(0.5)\left(\frac{2(\lambda-\lambda_0)}{\Delta\lambda_{\mathrm{FWHM}}}\right)^{10}} - 1\right)\pi, & \lambda \leq 825 \text{ nm} \\ \left(1 - e^{\ln(0.5)\left(\frac{2(\lambda-\lambda_0)}{\Delta\lambda_{\mathrm{FWHM}}}\right)^{10}}\right)\pi, & \lambda > 825 \text{ nm}, \end{cases} \tag{1}$$

Where $\varphi(\lambda)$ is the added spectral phase, λ is the wavelength, λ_0 is the central wavelength of the added spectral phase (825 nm), which corresponds to the approximate central wavelength of the used OPCPA spectrum and $\Delta\lambda_{\mathrm{FWHM}}$ is the FWHM of the added spectral phase (35 nm). This phase is practically equivalent with a π phase jump in a limited spectral range as used by others [14].

Figure 2(a) shows the spectrum of the green pumped OPCPA stages as well as the applied and the measured spectral phase to realize sub-FL pulses. Figure 2(b) shows the temporal structure of the FL, the measured compressed pulse, the pulse with a spectral phase according to Eq. (1) and the measured sub-FL pulse.

With SPS the FWHM pulse duration was reduced from the measured 7.9 ± 0.1 fs to 6.4 ± 0.1 fs, which corresponds to 86 ± 1% of the calculated FL FWHM. To accomplish this shortening

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the (temporal Strehl ratio) measured peak intensity was reduced to 48 ± 3% of the peak intensity of the FL.

The fast changes in the spectral phase, which are necessary for effective shortening of the pulse duration, can cause unwanted side effects in the shaper. E.g., it can reduce the overall transmission of the Dazzler or cause pre- and post-pulses, which render high intensity interactions with high density plasma targets from solids impossible [17]. Therefore, the implementation of TS via SAS was investigated, which does not cause those side effects.

To shorten the FWHM of the laser pulses, a hole was cut in the spectrum by reducing the transmission of the shaper to its minimum from 775–875 nm. This was found to be optimal via numerical optimization. The laser spectra with and without SAS as well as the measured spectral phases for both spectra are shown in Fig. 3(a). The temporal structures of the FL, the measured compressed laser pulse, the FL with SAS and the measured laser pulse with SAS are shown in Fig. 3(b). The FL FWHM of the original spectrum was 7.47 fs, which was reduced to the FL with SAS of 6.34 fs, corresponding to 85%. To achieve this the maximal intensity reduces to 47.5% compared to FL. The FWHM of the measured compressed original pulse was 7.7 ± 0.1 fs and the FWHM of the measured laser pulse with SAS was 6.35 ± 0.13 fs, which also corresponds to 85 ± 2% of the original FL. The measured temporal Strehl ratio was thereby reduced to 34 ± 2%.

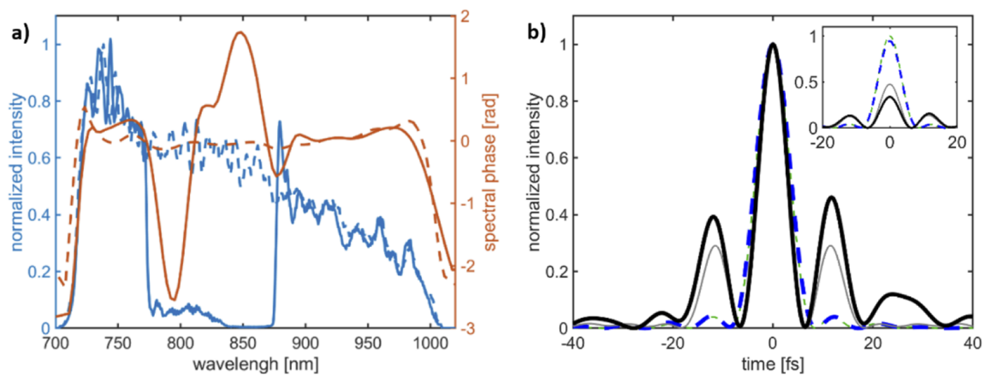

Fig. 3. Spectral amplitude shaping (SAS) to optimize the FWHM pulse duration of an OPCPA system. a) Spectral intensity and phase; dashed blue: original laser spectrum; dashed red: measured phase of the original compressed laser pulse; blue: spectrum with SAS; red: measured spectral phase with SAS. b) Temporal intensity; thin dashed green: FL of the original laser spectrum (FWHM: 7.47 fs); thick dashed blue: measured compressed laser pulse without SAS (FWHM: 7.7 ± 0.1 fs (103 ± 1%), I_{max}: 94 ± 1%); thin gray: FL of the spectrum with SAS (FWHM: 6.34 fs (85%), I_{max}: 47.5%); thick black: measured laser pulse with SAS (FWHM: 6.35 ± 0.13 fs (85 ± 2%), I_{max}: 34 ± 2%). The % value after the pulse duration is the ratio to the FL of the original spectrum, while the I_{max} value is the temporal Strehl ratio, i.e., the intensity decrease to the original FL. The inset on the right upper side shows the temporal intensity of the same four pulses, all normalized to the maximum of the FL pulse.

As the experiments and the theoretical predictions from Fig. 2 and Fig. 3 show, for the same shortening of the FWHM pulse duration SPS leads to slightly higher peak intensities than SAS. The implementation of SPS does not reduce the pulse energy, while that of SAS reduces it to about 60%. The expected temporal Strehl ratio for SPS is 53.4% and for SAS is 47.5%, which is a rather small difference and the implementation of SAS is in many cases much easier. SPS causes some side effects in the Dazzler [17], which lead also to a slight change of the spectrum. This is avoided by SAS and it can be implemented easily in systems without an acousto-optic programmable dispersive shaper via blocking certain wavelengths in a grating or prism stretcher or (custom made) spectral filters. Furthermore, the temporal characterization of a pulse with SAS is simpler (see supplementary material). Due to the fact, that the shaper in our system is located before the OPCPA stages, which are saturated and provide a gain of $\sim 10^5$, the suppression of the spectral intensity for certain wavelengths requires to set the shaper transmission to minimum. Still the amplifiers can pull up the intensity in these spectral regions to a significant level, as it is the case around 800 nm in Fig. 3(a). This not completely suppressed spectral range degraded the temporal Strehl ratio in the measurement. If complete suppression of this spectral region were possible, the FL pulse duration would be 5.96 fs. In a following experiment with the spectrum of Fig. 3(a), we combined SPS and SAS and utilized this region around 800 nm to achieve measured 6.1 ± 0.1 fs pulses (81 ± 1% of the original calculated FL) with a temporal Strehl ratio of 38 ± 2% (see supplementary material). In principle, this combined shaping is applicable to many other OPCPA systems, which significantly enhances effects caused by the carrier envelope phase (CEP) [24].

It is also possible to transfer the concept of TS to shorten the FWHM of laser pulses produced by an OPS system with even broader spectrum. To demonstrate this, SAS was applied using the entire spectrum of the LWS-20 laser system (all green and blue pumped OPCPA stages). The original spectrum of the LWS-20 and the spectrum with SAS as well as the measured spectral phases with and without SAS are shown in Fig. 4(a). It should be noted that the original laser pulse has a small hole around 700 nm, but it does not influence the demonstrated effects. The temporal structures of the FL, the measured compressed laser pulse, the FL with SAS and the measured laser pulse with SAS are shown in Fig. 4(b). To shorten the FWHM duration of the laser pulses the transmission of the shaper was reduced to its minimum for the spectral components from 720 nm – 800 nm. This resulted in a shortening of the FWHM duration from 4.64 fs FL and 5.0 ± 0.3 fs of the measured compressed pulse to 3.87 fs FL with SAS and 3.9 ± 0.2 fs for the measured pulse with SAS. This means, the measured FWHM duration with SAS is 84 ± 5% of the original FL with a temporal Strehl ratio of 39 ± 2% and 78 ± 8% of the measured compressed pulse (not FL) with a peak intensity ratio of 46 ± 4%. There is no significant modulation outside

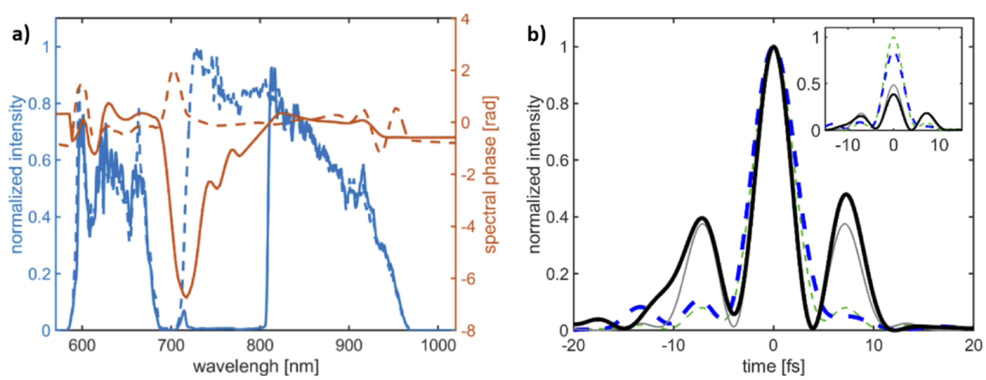

Fig. 4. SAS to optimize the FWHM pulse duration of LWS-20, a sub-5-fs optical parametric synthesizer. a) Spectral intensity and phase; dashed blue: original laser spectrum; dashed red: phase of the original compressed laser pulse; blue: spectrum with SAS; red: measured spectral phase with SAS. b) Temporal intensity; thin dashed green: FL of the original laser spectrum (FWHM: 4.64 fs); thick dashed blue: measured compressed laser pulse without SAS (FWHM: 5.0 ± 0.3 fs (107 ± 6%), I_{max}: 84 ± 4%); thin gray: FL of the spectrum with SAS (FWHM: 3.87 fs (83%), I_{max}: 48.9%); thick black: measured laser pulse with SAS (FWHM: 3.9 ± 0.2 fs (84 ± 5%), I_{max}: 39 ± 2%), representing a sub-4-fs multi-TW light source. The inset on the right upper side shows the temporal intensity of the same four pulses, all normalized to the maximum of the original FL pulse.

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of the shown temporal window (see supplementary material). The corresponding peak power is 5.1 ± 0.3 TW. This source represents the first sub-4-fs laser with >5 TW peak power.

In addition to shortening the FWHM of the laser pulses, TS also increases the asymmetry of the half-cycles of the electromagnetic wave around the peak. The reduction of the measured pulse duration form 5 fs to 3.9 fs decreases the height of the second highest half-cycle of the electromagnetic wave in respect to its highest half-cycle from 92% to 87%. It makes this novel multi-TW sub-4-fs light source an ideal driver for the generation of intense isolated attosecond pulses [10,25] and has the potential to increase the spectral bandwidth of generated isolated attosecond pulses by approximately 50%. Furthermore, it significantly boosts CEP effects.

3. Conclusion

We have demonstrated the application of TS to a broadband multi-TW OPCPA system that supports sub-8-fs pulses as well as its application to a multi-TW OPS system supporting sub-5-fs pulses. The spectral shaper, which is located before the main amplification stages of the laser system, was used to realize spectral amplitude as well as phase shaping. This allowed to produce sub-FL pulses and a shortening of the FWHM pulse duration of the OPCPA system of more than 15%, which can be applied in similar form to shorten the FWHM of many existing OPCPA systems.

Amplitude shaping of the OPS system produced pulses with less than 4 fs FWHM duration and >5 TW peak power. This makes it up to now the most powerful sub-4-fs laser source in the world.

The quasi-single-cycle pulse duration of this new light source in combination with its high peak power are optimal for nonlinear applications such as nonlinear optics, high-harmonic generation or relativistic laser-plasma physics. It will enable the generation of isolated attosecond pulses with high pulse energies and add a new viable tool to the toolbox of attosecond physics [25]. Furthermore, tight focusing of the sub-4-fs pulses facilitates the generation of relativistic intensities and the study of waveform-dependent relativistic laser-plasma phenomena [26].

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Data Availability. Data underlying the results presented in this paper are not publicly available at this time, but may be obtained from the authors upon reasonable request.

Supplemental document. See Supplement 1 for supporting content.

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