High-contrast, intense single-cycle pulses from an all thin-solid-plate setup

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High-contrast, intense single-cycle pulses are highly desirable tools in ultrafast science, enabling highest temporal resolution, pushing matter to extreme conditions, and serving as drivers in petahertz electronics. In this Letter, we use thin solid plates in a double multi-plate supercontinuum configuration, delivering a broadband spectrum spanning from ~400 to ~1000 nm at the −20 dB intensity level to produce a single-cycle pulse. We show that the spectral broadening by self-phase modulation with few-cycle pulses is more suitable for compression than the single-cycle limit than with multi-cycle pulses. The pulses are compressed to 2.6 fs pulses, close to the transform limit of 2.55 fs, with an energy of 0.235 mJ. They exhibit an excellent power stability of 0.5% rms over 3 h and a beam profile. The obtained single-cycle pulses can be utilized in many applications, such as generation of isolated attosecond pulses via high-order harmonic generation, investigation of ultrafast phenomena with extreme temporal resolution, or high-intensity laser-solid experiments. © 2020 Optical Society of America

At the heart of the interaction between a laser pulse and matter is the response of the electrons of the material to the incident electric field. The light–matter interaction depends strongly on the ratio of the pulse duration to the characteristic response time of the medium (polarization response), in addition to the pulse intensity and energy. Recently, it has been noted that there exist fundamental inherent peculiarities in the interaction physics in a single-cycle pulse limit, which are not manifested in usual multi-cycle pulses: the strong-field ionization dynamics in a single-cycle pulse is quite different from that in a multi-cycle pulse [1,2]. Accordingly, single-cycle pulses are desirable in various areas of ultrafast optical science. For example, in high-order harmonic generation (HHG), attosecond pulses are produced every half cycle of a driving laser pulse. Various methods [3–5] have been devised and successful in producing isolated attosecond pulses using multi- or few-cycle laser pulses. However, single-cycle pulses promise to generate an isolated attosecond pulse more easily and with higher efficiency. Furthermore, single-cycle pulses are powerful tools for the development of petahertz electronics, in particular for wide bandgap materials [6–8]. Consequently, experimental efforts have been pushing pulse durations into the single-cycle limit [9–12] in the optical or near-infrared spectral region.

To obtain high-contrast single-cycle pulses, a broadband supercontinuum and proper compensation of dispersion are required. Commonly, such a broadband spectrum has been obtained from the nonlinear processes of multi-cycle pulses with gas-filled hollow-core fibers (HCF) [13–15]. However, in general, such HCF setups have a relatively low throughput of ~50% and limitations in long-term stability without a beam-locking system.

As an alternative, the spectral broadening by solid thin plates, called multiple-plate supercontinuum generation (MPSC), has been studied in recent works [16–18]. Here, a strong laser pulse is focused onto a medium, and the nonlinear effects induced by the intense field, including self-focusing, self-phase modulation, and self-steepening (SS), are responsible for spectral broadening. In a MPSC, the first plate is placed near the focus of the driving laser for maximum spectral broadening, while avoiding damage on the plate. The other plates are then placed one by one until no further spectral broadening occurs. Namely, by adequately placing thin plates around the focal region of the laser, the formation of filaments can be avoided, and a dramatic increase of bandwidth can be achieved.

It has been known that SPM leads to symmetrical spectral broadening, allowing a broader full width at half maximum (FWHM); on the other hand, SS generates higher frequencies, leading to broad hump in the blue spectral region, not only breaking a spectral symmetry, but also introducing both complicated spatial chirp and high-order spectral dispersions [19,20]. This means that the broadened spectrum by SPM may be compressed successfully to a temporally transform-limited pulse.
One solution that enhances the SPM effect during spectral broadening is to use well-compressed few-cycle pulses as driving sources for MPSC, since the pulse duration of the input pulse has a significant effect on the relative contribution from SPM in spectral broadening [18–21]. In our study, we use a double MPSC configuration; the supercontinuum from the first MPSC stage is compressed to a few-cycle pulse, which is then used as a driving pulse for the second MPSC stage.

In this Letter, we demonstrate the generation of intense, high-contrast single-cycle pulses, using thin solid plates with a 0.42 mJ/30 fs Ti:sapphire femtosecond laser at 3 kHz. Two sets of broadband chirped mirrors with an ammonium dihydrogen phosphate (ADP) crystal were used to control the dispersion and compress the supercontinuum output from the second MPSC stage to 2.6 fs (single-cycle) pulses characterized by the dispersion scan (D-scan) method [22], which has been proven to be a suitable technique for characterizing few- or single-cycle pulses [9,22–25]. A power stability of 0.5% over 3 h and an M² value of 1.26 were achieved. The central brightest part of beam was used for pulse compression and beam characterization, such as measurement of energy, power stability, and M².

The experimental setup is illustrated in Fig. 1. The input pulses of 30 fs and 0.42 mJ at a central wavelength of 780 nm, from a multi-pass Ti:sapphire amplifier at 3 kHz, are focused into the first MPSC stage consisting of two 100-μm-thick fused silica plates. Since a too high intensity induces stronger high-order nonlinearity [26] leading to low conversion efficiency due to energy loss from ionization [27,28], the input beam size is reduced by a mirror telescope for loose focusing. The peak intensity of the incident pulses is $7.1 \times 10^{12}$ W/cm² at the beam waist without any plates, the beam diameter is 530 μm at the 1/e² width, and correspondingly, the Rayleigh length is 28.3 cm.

In a typical MPSC, SPM is a dominant mechanism for spectral broadening for the first few plates, whereas SS starts to play the main role in additional plates, giving rise to the formation of a broad blue hump [16–18] in the output spectrum. In other words, since the steepening effect occurs sequentially when adding more plates for MPSC, we can minimize the effect of SS during spectral broadening by controlling the number and position of the plates. Figure 2(a) are the change of the spectrum with respect to the number of plates. When more than three plates were used, the hump-like structure toward high frequencies starts to occur rapidly, breaking the symmetric profile. Hence, in the first MPSC stage, to obtain an SPM-driven symmetrically broadened spectrum, we installed two plates before SS becomes a dominant process. The plates were set at the Brewster angle (55.5°), and the first plate was located 16 cm before the beam waist for maximum broadening without damage. A second plate was located 14 cm from the first plate. The output energy was 0.395 mJ, corresponding to a conversion efficiency of 94.0%. After passing three pairs of chirped mirrors and a wedge pair, the pulses were characterized with a transient-grating frequency-resolved optical gating (TG-FROG) setup. To get a pulse duration as short as possible, the higher-order dispersions of the input pulse, namely, the third-order dispersion (TOD) and the fourth-order dispersion (FOD), were optimized with the Dazzler (Fastlite inc.) settings of the Ti:sapphire laser amplifier [9]. Optimal compression resulted in 8.8 fs pulses, as shown in Fig. 2(b), close to a transform limit of 8.0 fs. The retrieved pulse was reconstructed from FROG traces using a 256 × 256 grid size with a FROG error of 0.007. The output of the first MPSC stage was then sent into the second MPSC stage for further spectral broadening. After the first MPSC stage, the beam size was adjusted for loose focusing in the second MPSC stage by optimizing the distance and focal length between the concave mirrors FM1 and FM2 in Fig. 1. The intensity of pulses at the beam waist was $1.11 \times 10^{13}$ W/cm² without any plates, and the beam diameter was 700 μm at the 1/e² width for the second MPSC. Two pieces of 100-μm-thick fused silica plates with a separation of 19 cm were used in the second MPSC stage. After collimation with a concave mirror, the output pulses were compressed by a second set of chirped mirrors and wedge pair. The specifications of the second set of chirped mirrors supported shorter pulse durations, and the covered spectral range was shifted to the blue compared to the first set. The resulting spectrum after the second MPSC stage spanned from 460 nm to 950 nm at the −20 dB intensity level with four plates in total.

Fig. 2. Characterization of the output from the first MPSC stage. (a) Spectra (log scale) taken after each fused silica plate. (b) Retrieved pulse envelope (red curve). The output pulses are compressed to 8.8 fs, as measured by TG-FROG, close to the transform-limit of 8.0 fs (blue curve).

Fig. 1. Experimental setup for high-contrast intense single-cycle pulses with double-stage fused silica thin plate broadening and pulse compression. L, focusing lens (f = 2 m); FM1, FM2, and FM3, concave mirrors (f = 1 m, 1.5 m, and 0.5 m, respectively); CM1 and CM2, chirped mirrors (Ultrafast Innovations PC70 and PC1332, respectively). The output pulses are sent to a D-scan setup for characterization.
single-cycle pulses. The additional plates were placed 8 cm, additional plates for further spectral broadening to obtain high enough after the second plate. Hence, we installed three TOD compensation.

3.42 fs than a typical single MPSC configuration (5.35 fs) with double MPSC configuration provides shorter pulse durations of the same spectral range using fewer plates without additional configuration. The reconstructed phase [Fig. 3(a), red solid curve], this results in a spectrum extending from 460 nm to 950 nm at the −20 dB intensity level with a pulse energy of 0.34 μJ (conversion efficiency, 83%). Note that for compression, different numbers of chirped mirrors were used in the single or double configurations: six pairs for single and five pairs for double MPSC configuration.

Temporal characterization was done with a D-scan setup with 5-μm-thick Type I beta barium borate (BBO) crystal. Figure 3 also shows the results of D-scan measurements for each MPSC configuration. In the single configuration, the output spectrum has a broad hump toward high frequencies [Fig. 3(a), red solid curve], while the output spectrum from the double MPSC configuration does not exhibit such a broad blue hump. Figures 3(b) and 3(c) show that, in the single MPSC configuration, more residual negative TOD can be found than in the double MPSC configuration. The reconstructed phase [Fig. 3(a), red and blue dashed curves] confirms this and results in a satellite structure (red curve), where the pulse was compressed to 5.35 fs. The high-contrast 3.42 fs pulses result from the double MPSC configuration (blue curve).

[Fig. 3(a), blue solid curve], corresponding to a transform-limited pulse duration of 3.13 fs. We note that the spectral broadening remains symmetrical in the two MPSC stages. We infer that SPM plays the dominant role in spectral broadening in this case. The output energy was 0.34 μJ, resulting in a high conversion efficiency of 93% in the second MPSC stage. For comparison, we built a single MPSC configuration to reach the same spectral coverage, as in the double MPSC configuration. Seven plates in total were required. As shown in Fig. 3(a) (red solid curve), this results in a spectrum extending from 460 nm to 950 nm at the −20 dB intensity level with a pulse energy of 0.35 μJ (conversion efficiency, 83%). Note that for compression, different numbers of chirped mirrors were used in the single or double configurations: six pairs for single and five pairs for double MPSC configuration.

In the double MPSC configuration, the peak power is still high enough after the second plate. Hence, we installed three additional plates for further spectral broadening to obtain single-cycle pulses. The additional plates were placed 8 cm, 5 cm, and 3 cm, respectively, downstream from the second plate. After five pieces of 100 μm thick silica in the second MPSC stage, the output pulses have a spectrum spanning from 392 nm to 987 nm at the −20 dB intensity level [Fig. 4(a), black curve], supporting a transform-limited pulse duration of 2.55 fs [Fig. 4(d), black dashed curve]. The output energy was 0.3 μJ before compression with a beam profile shown in Fig. 5(a). The center spot contains over 90% of the total energy. The overall conversion efficiency was 71%. During further spectral broadening with the additional plates, the symmetry of output spectrum worsens but still has no hump structure like observed in a typical single MPSC configuration. We noted, however, that if we put more plates in addition to current five plates in the second MPSC stage, the asymmetry develops rapidly in the output spectrum, and energy loss is increased due to excessive conical emission. Hence, the five plates were found to be an optimum number for the second MPSC stage. The D-scan trace of the pulses compressed with six pairs of chirped mirrors and 4 mm fused silica (inserted via the wedge pair) is shown in Fig. 4(b). The reconstructed spectral phase is presented in Fig. 4(a) (red dashed curve). The residual TOD increases with seven plates in total in both stages, as shown in Figs. 3(c) and 4(b). Based on the reconstructed spectral phase [Fig. 4(a), red dashed curve], there is a residual TOD of around −50 fs. Further TOD compensation is required for the compression down to single-cycle limit. For TOD compensation, an ADP crystal was used, since it exhibits a relatively high ratio of TOD to GDD [29]. For compensating the residual negative TOD after seven plates in both MPSC stages, a 2-mm-thick z-cut ADP crystal (United Crystals, Inc.) was found to be suitable. At the same time, a similar amount of fused silica had to be removed via the wedges. The D-scan trace of pulses after adding the ADP crystal and removing 2-mm-thick fused silica for dispersion compensation is presented in Fig. 4(c). Note that the tilting of the trace is now removed, indicating that remnant TOD was well compensated. The almost flat reconstructed spectral phase
The development of petahertz electronics.

Such high-contrast, intense single-cycle pulses can be utilized in many applications, including generating bright isolated attosecond pulses by HHG, high-resolution investigation of ultrafast processes, and few-cycle pulses are more suitable for spectral broadening by SPM than multi-cycle pulses. The single-cycle pulse has an excellent beam profile and long-term power stability. A close-to-transform-limited single-cycle pulse of 2.6 fs FWHM with a spectral coverage from 400 nm to 1000 nm at a ∼20 dB intensity level is achieved at the 3 kHz repetition rate with a pulse energy of 0.235 mJ. The double MPSC configuration can also be adopted to higher pulse energies or different repetition rates by properly adjusting the pulse intensities with the location of thin plates together with a careful compensation of TOD via an ADP crystal. Our studies also show that spectral broadening by SPM is more favorable for optimal compression, and few-cycle pulses are more suitable for spectral broadening by SPM than multi-cycle pulses. The single-cycle pulse has an excellent beam profile and long-term power stability. A close-to-transform-limited single-cycle pulse of 2.6 fs FWHM with a spectral coverage from ∼400 to ∼1000 nm at a ∼20 dB intensity level is achieved at the 3 kHz repetition rate with a pulse energy of 0.235 mJ. The double MPSC configuration can also be adopted to higher pulse energies or different repetition rates or wavelengths by properly adjusting the pulse intensities with a looser focusing scheme, and/or modifying the plate material. Such high-contrast, intense single-cycle pulses can be utilized in many applications, including generating bright isolated attosecond pulses by HHG, high-resolution investigation of ultrafast phenomena, or high-intensity laser-solid experiments aimed at the development of petahertz electronics.

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