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Direct carrier–envelope phase stabilization of a soliton-effect compressed sub-two-cycle pulse source through nonlinear mixing of solitonic and dispersive waves

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Abstract We present a carrier–envelope phase (CEP) stabilized sub-two-cycle 5.2 fs pulse source based on soliton-effect self-compression of femtosecond laser pulses in millimetre-long highly nonlinear photonic crystal fibres. We employ a simple and efficient scheme to generate a strong (40–60 dB, configuration dependent) CEP beat signal directly from the pulse source via *f*-to-2*f* interferometry where the second harmonic of the main soliton pulse is mixed with the isolated dispersive blue/green radiation peak that is also generated in the compression process, obviating the need for additional spectral broadening mechanisms.

1 Introduction

Carrier–envelope phase (CEP) stabilization techniques have experienced remarkable progress since the first time-domain measurements of the CE-phase shift between successive pulses from a Ti:Sapphire oscillator using a second-order cross-correlator [1]. This early method was not sufficiently sensitive to the very small phase errors that rapidly accumulate at the high repetition rate of laser oscillators, which prevented long-term CEP coherence and resulted in incorrect offset frequency measurements [2]. Just a few years later, various innovative schemes for the phase-coherent determination of the carrier–envelope offset (CEO) phase were proposed [3]. The simplest and more straightforward approach, known as *f*-to-2*f* self-referencing, involved only one nonlinear process—second harmonic generation (SHG)–for obtaining a CEP beat signal, although it required broadening of the original laser spectrum over an octave in bandwidth, which at the time was still inaccessible directly from the low-energy femtosecond pulses delivered by laser oscillators. Such large spectral broadening became feasible via supercontinuum generation in microstructured fibres [4] and the first experimental demonstrations of the method were soon reported [5, 6].

Over the last decade, many efforts have been done to stabilize and control the CEP in femtosecond Ti:Sapphire laser oscillators and several detailed studies have been performed over a wide range of topics, from CEP dynamics to phase coherence and the mechanisms behind the origin of noise. More recent improvements in CEP stabilization techniques, such as direct phase measurements based on multi-photon-induced photoelectron emission from a gold surface [7], direct locking in the time domain [8], direct CEP stabilization of low-noise prismless octave spanning Ti:Sapphire oscillators [9, 10] and the recent feed-forward approaches [11] give us a hint of the importance of this research field for applications where low-noise CEP-stabilized pulses are required, such as attosecond science, frequency metrology, precision spectroscopy and extreme nonlinear optics.

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In the time domain, CEP-dependent effects become noticeable at very short pulse durations approaching the single-cycle limit. Therefore, an important characteristic of a few-cycle pulse source is the possibility of stabilizing its CEP. In earlier work, we experimentally demonstrated the direct generation of 4.6 fs sub-two-cycle broadband optical pulses by soliton-effect compression of low-energy pulses from a standard Ti:Sapphire laser in a short highly nonlinear photonic crystal fibre (PCF) [12]. Figure 1 shows a typical output spectrum obtained using sub-nanojoule 42 femtosecond pulses propagating in a 4.85-mm-long PCF, where it can be seen that its main portion is not octave spanning, at least on a linear scale [13, 14]. Interestingly, this spectral profile is nevertheless adequate for direct CEP stabilization using the *f*-to-2*f* technique.

More exactly, the presence of an isolated peak of non-solitonic (dispersive) radiation in the blue region of the spectrum near 470 nm, together with a particular component in the IR region extending from 900 to 1,000 nm, both with high spectral power densities, allows for the generation of the beat signal that determines the carrier-envelope offset (CEO) frequency with a signal-to-noise ratio (SNR) of more than 50 dB (400 kHz resolution bandwidth) directly from this soliton-effect compression sub-two-cycle pulse source [15]. Nevertheless, only short-term (several seconds) CEP stabilization of the pulse source was previously achieved since some issues related to the stability of the unlocked signal had to be solved, namely by reducing mechanical and acoustical noise sources.

Several steps towards this goal have been performed: the complete water circuit used to cool the Ti:Sapphire crystal within the laser oscillator was improved to eliminate vibration coupling to the optical table (which is also actively vibration isolated), the closed-loop water chillers were removed from the laboratory room, several optical mounts were adapted and more firmly adjusted to the table, and the *f*-to-2*f* interferometer was placed inside an isolating box to provide shielding against external perturbations such as air currents, acoustic noise and thermal drift. It is known that any vibrations of the optical mounts, or temperature and/or pressure variations induce a random phase shift between the interfering beams that is transferred through the stabilization loop to the carrier–envelope phase instability of the pulse source [16, 17], although the last modification mentioned above only resulted in minor improvements in the overall stability of our CEP stabilization scheme.

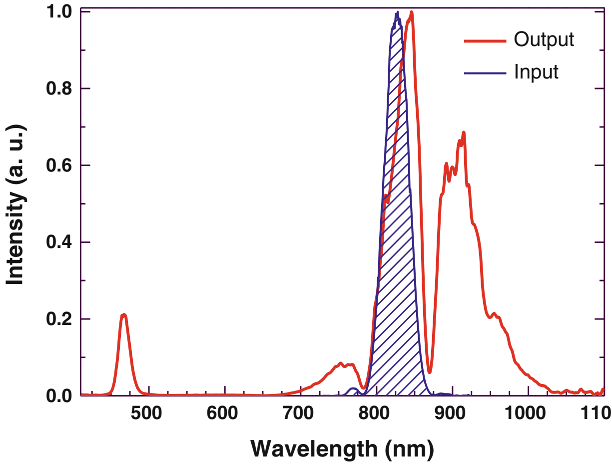

Fig. 1 Measured input and output spectra of the sub-two-cycle pulses generated by soliton-effect compression in millimetric highly nonlinear PCFs, showing the "isolated" non-solitonic radiation peak centred around 470 nm [15]

2 Experimental method

The fundamental scheme used for generating CEP stabilized sub-two-cycle pulses is based on the setup described in previous work [15]. In the present setup, several parameters and components (namely the PCF length) have been optimized for the generation of a strong CEO beat note while simultaneously obtaining and characterizing the resulting sub-two-cycle pulses in the time domain. Our home-built 85 MHz, 10 fs Ti:Sapphire oscillator [18] was spectrally narrowed using a slit placed between its intracavity prisms to generate the ~30 nm bandwidths and correspondingly longer pulses required for optimum soliton-effect compression (Fig. 2). Pulse chirp was controlled with an extracavity fused-silica prism pair prior to coupling to a 2.85-mm-long highly nonlinear PCF (Crystal Fibre A/S, core diameter of 1.6 µm, zero-dispersion wavelength $\lambda_{ZD} = 670$ nm) using a standard 40 × (0.65 N.A.) microscope objective. In spite of the short length of our PCF, we were able to use it for several days in continuous operation time with input average powers of more than 185 mW (peak power of 53 kW) before noticing any deterioration or long-term damage to the fibre end-faces [19] capable of affecting our measurements. Care was taken to ensure that the oscillator was operating in the single pulse regime, with no continuous-wave (cw) component or multiple pulsing also because the later operation modes tend to potentiate damage to the PCF. Input energy and polarization control were performed with a polarizing beamsplitter cube (PBS) placed between two achromatic half-wave plates.

The broadband PCF output pulses were collimated with a broadband aspheric lens L ($f = 2$ mm) and reflected off a new ultra broadband double-chirped mirror (DCM) pair, with a group-delay dispersion (GDD) of approximately -82.5 fs^2 per bounce at 800 nm. These DCMs have a twofold purpose since (1) they preserve the short pulse

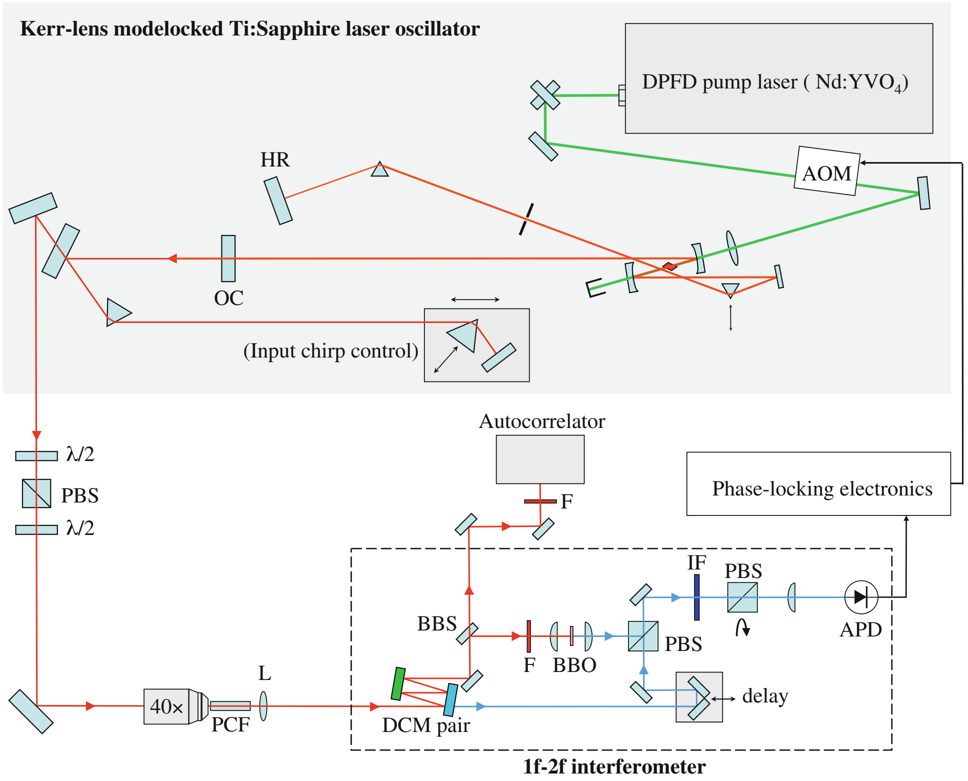

Fig. 2 CEP stabilized soliton-effect compressed sub-two-cycle pulse source: experimental setup for direct generation and simultaneous temporal characterization of the pulses (see text for details)

duration expected at the output of the PCF by pre-compensating the dispersion of the subsequent optical setup in the branch created for temporal characterization, as mentioned previously, and (2) in addition, separate the two frequencies for the f-to-$2f$ interferometer. Actually, we used one of the mirrors as a dichroic mirror to transmit the blue portion of the spectrum (a peak of non-solitonic radiation centred at around 470 nm), and made it interfere with the blue pulses obtained after frequency doubling the infrared (IR) band at 940 nm using a 2-mm-long type-I BBO crystal. The resulting beat signal gives the CEO frequency f_0 of the comb. An adequate variable delay stage in one arm of the interferometer assures that both the visible band and the second harmonic of the IR band ($1f$ and $2f$ frequencies) can be temporally and spatially overlapped with the required high precision. A polarizing beamsplitter (PBS) is used to collinearly combine the orthogonally polarized $1f$ and $2f$ pulses, and a second rotating PBS allows us to balance the power of both interferometer arms for optimal contrast, further increased by the interference filter (IF) that removes the background out of the SHG signal. The beat signal was detected with an avalanche photodiode (APD210, Menlo Systems GmbH), monitored with an rf spectrum analyzer and then coupled to phase-locking electronics (XPS 800, Menlosystems GmbH) to control an acousto-optic modulator (AOM, IntraAction) that regulates the power in the pump beam of the Ti:Sapphire laser oscillator.

The offset frequency was locked to ¼ of the repetition rate of the laser oscillator, which was experimentally measured as $f_{\mathrm{rep}} = 85.44$ MHz, so that every fourth pulse leaving the oscillator would have the same CEP when the offset frequency was locked.

Furthermore, using a 0.65-mm-thick ultrabroadband beamsplitter (BBS, Venteon GmbH) with constant splitting ratio and equalized dispersion in transmission and reflection, we managed to simultaneously characterize our soliton-effect compressed pulse source in the time domain. Once filtered with a 0.5-mm-thick lowpass filter to eliminate residual short-wavelength (sub-610-nm) light, the PCF output pulses were coupled to a second-harmonic autocorrelator equipped with a 20-μm β-barium borate (BBO) crystal (Femtolasers GmbH). The output spectra were measured by two calibrated fibre optic spectrometers (Ocean Optics), covering the IR/red and red/blue spectral regions, providing a measurement range extending from about 210 to 1,050 nm, limited by their own spectral sensitivity.

3 Results and discussion

Measuring the temporal duration of our pulse source and at the same time directly generating the CEO beat signal for its stabilization restricts our results to a trade-off between the best possible autocorrelation (i.e., the shortest pulses) and the best CEO beat signal for phase stabilization (the one with the highest SNR), since these conditions are not forcibly fulfilled by the same generated spectrum. In fact, by adjusting the delay between both interferometer arms, as well as the central wavelength, chirp, energy and polarization of the input Ti:sapphire laser pulses, and by fine-tuning the whole alignment of the *f*-to-2*f* interferometer, a clear CEO beat signal with a SNR of 60 dB (400 kHz resolution bandwidth) was obtained, well above the 30 dB usually required for standard CE-phase stabilization electronics to track and lock the signal (Fig. 3). Even though the CEO frequency beat note is quite strong, its trace is still contaminated with noise that we were not able to eliminate completely.

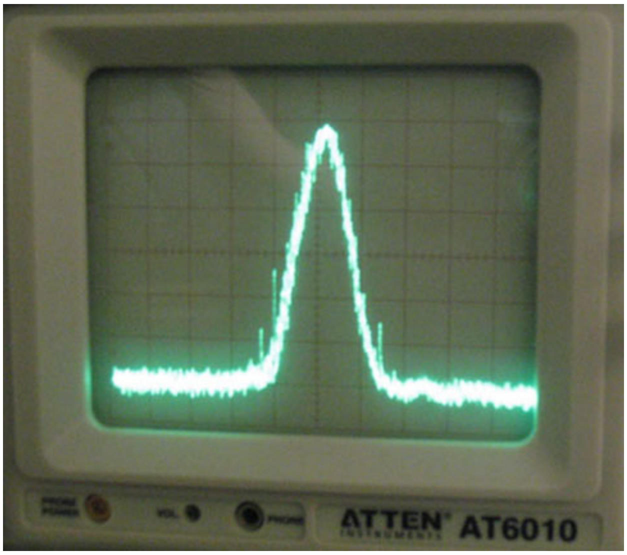

Fig. 3 CEO beat signal with a SNR of 60 dB (at 400 kHz resolution bandwidth over a 10 MHz span)

However, it is essential to assure that we still have a soliton-compressed pulse source in the sub-two-cycle regime, which for our spectrum corresponds to a temporal width forcibly shorter than 5.7 fs.

As formerly stated, the best compromise resulted in soliton-effect compressed pulses as short as 5.2 fs (FWHM) obtained for moderately chirped input pulses with 41.0 fs and 161 mW average power at the entrance of the PCF, which corresponds to a compression factor of ~8. The input spectrum has a 35 nm bandwidth centred at 838 nm; the output reveals the presence of an isolated band of non-solitonic radiation in the blue region of the spectrum centred at 470 nm, and a broadband component that extends from 600 to 1,100 nm, as shown in Fig. 4a. Even though the resulting blue band has a significantly smaller intensity compared to that in Fig. 1, it still has enough spectral power density to achieve a good signal strength in the CEO beat signal.

The measured interferometric autocorrelation (IAC) trace of the compressed pulses is shown in Fig. 4b. The sub-two-cycle duration is consistent with our previous measurements where the full electric field of the compressed pulses was retrieved with a genetic algorithm applied to spectral and IAC measurements and further corroborated by numerical simulations [12]. The collimated output power is 48 mW, corresponding to an overall efficiency of 30 %, slightly higher than our previous results [12] as expected from using a collimating lens with a larger numerical aperture.

With this sub-two-cycle pulse source and beat note, we achieved locking of the CE beat signal to some degree for more than 1 h. A CEO frequency with a SNR of 45 dB (6.5 kHz resolution bandwidth) centred at $f_0 \approx 21.3$ MHz was measured with a digital spectrum analyzer (Signal Hound USB-SA44B), as shown in Fig. 5. The observed

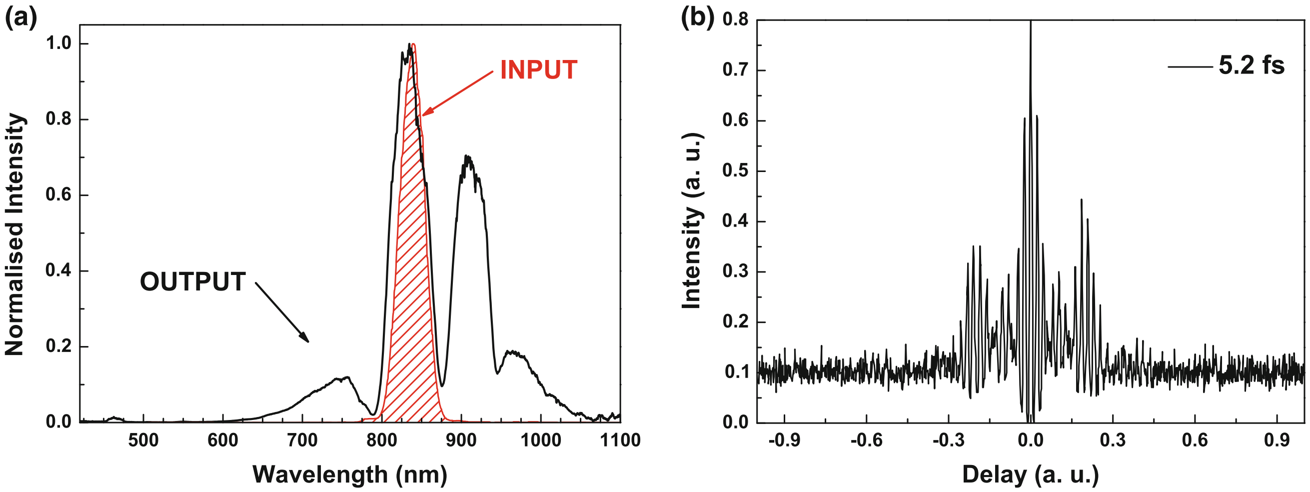

Fig. 4 **a** Measured PCF input and output spectra of the pulses. **b** Measured IAC trace of the soliton-effect compressed pulses

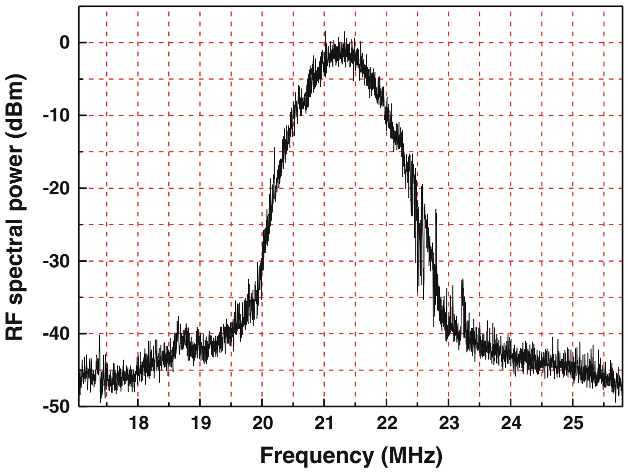

Fig. 5 Locked beat signal with a SNR over 40 dB (at 6.5 kHz RBW over a 5 MHz span)

lower SNR compared to Fig. 3 is due to the different instrument and settings used. Although the central frequency of the beat note was successfully locked, its bandwidth is still too wide (approximately 2.25 MHz), possibly due to the uncompensated noise in the unlocked beat note mentioned above.

Apparently the stabilization is effective only at low frequencies, but fails to remove some high-frequency modulation from the CEP beat note. There are several possible sources for the observed residual phase noise. For example, it is known that the use of a multi-longitudinal-mode diode-pumped frequency-doubled (DPFD) Nd:YVO4 laser for pumping the femtosecond laser presents some disadvantages since mode competition and beating effects increase the relative intensity noise (RIN) of the output [19, 20] which, through nonlinear effects, couple directly to CEO phase noise in the mode-locked Ti:Sapphire oscillator. These multimode pump lasers show significantly larger RIN in higher-frequency ranges (typically from 1 kHz to 10 MHz) and our feedback loop, with its 20-kHz bandwidth, may not be able to correct the induced extra CEP fluctuations. Improved noise performance could be obtained by extending the feedback loop bandwidth to 100 kHz since the larger RIN values are precisely in the 1–100 kHz range, but still this limited bandwidth would be insufficient to fully suppress the intensity-dependent high-frequency CEP noise, and therefore stabilization electronics should be further optimized [20].

Beam pointing instability is another major drawback of DPFD pump lasers when dealing with CEP stabilization techniques and it was found that this increases the fluctuations of the CEO frequency beat signal, resulting in a wider bandwidth of the locked central frequency, although it does not affect the SNR [19].

The CEO frequency is also susceptible to intracavity laser energy variations and to vibrations of the laser cavity. Ti:Sapphire lasers with intracavity prisms for dispersion compensation are especially susceptible to vibrations due to optical path length changes. Thus, in order to accomplish CE-phase stabilization, the laser should have very high passive stability. Due to the need to properly tune the input pulses for soliton-effect compression, the use of a different oscillator, namely a mirror-dispersion controlled laser, is not practical here, unless external spectral filtering is performed at the cost of pulse energy.

Another possible source of noise would be the *f*-to-2*f* interferometer itself. It is known that simple and versatile common-path interferometers that use a calcite Babinet–Soleil compensator for precise adjustment of the delay between the two overlapping beams [21] can strongly reduce phase noise when compared to typical Mach–Zehnder configurations and even actively stabilized interferometer versions [16]. Still, we could not clearly identify our interferometer configuration as a source of residual phase noise in the CEO beat signal.

The particular nonlinear dynamics of the soliton compression process may also play a role in the observed residual CEP noise, since we obtain the CEP beat signal by mixing the second harmonic of the main soliton with the fundamental non-solitonic (dispersive) radiation in the blue spectral region. Our system, therefore, relies upon the mutual coherence between solitonic and dispersive radiation in the sub-two-cycle regime. Bergé et al. [22] have recently shown numerically that the CEP of higher frequencies generated by filamentation and self-compression of femtosecond laser pulses in gases (a process that shows some similarity to soliton compression in negative dispersion fibres) exhibits a very high sensitivity to input amplitude fluctuations for output pulses near the maximum compression region, with intensity changes of a few percent resulting in large excursions of the CEP and consequent blurring when averaged over several laser shots. A similar degradation in coherence could be taking place in our experiment thus affecting the quality of our CEP signal, and this possibility will be analyzed in future experimental and numerical work.

4 Conclusions

We presented a CEP stabilized sub-two-cycle pulse source based on soliton-effect self-compression of standard femtosecond laser pulses in millimetre-long highly nonlinear PCFs. By coupling moderately chirped input pulses centred at 838 nm, with 41.0 fs and 161 mW average power to a 2.85-mm-long PCF, we obtained 5.2 fs (FWHM) output pulses corresponding to a compression factor of ~8. A simple and efficient scheme was employed to generate a strong CEP beat signal directly from the pulse source. By mixing an isolated band of non-solitonic (dispersive) radiation around 470 nm with the second harmonic of the main (solitonic) broadband component that extends from 600 to 1,100 nm in a *f*-to-2*f* interferometer, beat signals with a 40–60 dB SNR (configuration dependent) were obtained. Simultaneous measurement of the temporal duration of the pulses required a trade-off between the best possible autocorrelation (i.e., the shortest pulses) and the best CEO beat signal for phase stabilization (the one with the highest SNR). The central frequency of the beat note was successfully locked for about 1 h with a 45 dB SNR, although its bandwidth is still too wide (approximately 2.25 MHz for a 6.5 kHz RBW) possibly due to uncompensated phase noise outside the bandwidth of the locking electronics used in this work. Identification and further reduction of the residual CEP noise will be pursued in the upcoming work, and conducting an out-of-loop phase noise analysis may give additional insight to sort out the reason for the peculiar behaviour.

This method should further extend the usefulness and applicability of few-cycle soliton-compressed laser pulses with the added advantage that the CEO frequency is obtained by *f*-to-2*f* interferometry directly from the pulses to be used in experiments without the need of standard octave-spanning spectra nor additional nonlinear spectral broadening mechanisms.

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