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5.1 fs pulses generated by filamentation and carrier envelope phase stability analysis

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Abstract

Intense 5.1 fs CEO (carrier envelope offset) phase stable pulses were generated through two-fold filamentation in a noble gas at atmospheric pressure. The preservation of the CEO phase during the filamentation process was investigated. We show that generating these short pulses using filaments is not detrimental for the CEO phase stabilization, and that the more than one octave-spanning spectrum intrinsically generated by the process is feasible, and offers certain benefits, for direct use in single shot f–$2f$ spectral interferometry.

1. Introduction

In the last few years, strong interest has been shown in the new emerging fields of high-energy physics and attosecond science, mainly because of the attractive possibility of implementing time-resolved spectroscopy on an attosecond scale [1–3]. The attosecond pulse regime has been entered [4–6] by up-converting optical frequencies into the extreme ultraviolet (XUV) spectral region by the so-called high harmonic generation (HHG). By using sufficiently intense few-cycle infrared pulses as a pump, atoms can be driven into a strongly nonlinear regime where they can eject one of their outer electrons, which is then moving on trajectories determined by the residual field of the driving pulse. For trajectories starting at certain intervals in the electric field, the electron wave packet can return to its parent ion having gained additional kinetic energy from the field. In the case of recombination with the ion, an XUV photon is emitted. Employing such a scheme, pulses with durations of a few hundred attoseconds could be generated [6].

Using HHG as an approach to obtain isolated attosecond pulses, the fundamental laser pulses need to follow some strict requirements. To ensure that significant ionization probability is reached during only one cycle of the fundamental pulse, its intensity must be carefully controlled and its duration must be in the few-cycle regime.

Even then, the so-called sine pulses exhibit two field maxima of almost equal strength making control of the CEO (carrier envelope offset) phase one more necessary prerequisite [7].

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Until recently, the only known way to obtain such pulses was based on spectral broadening of phase-locked 30–50 fs pulses in a long guiding structure such as gas-filled hollow fibres with subsequent recompression using chirped mirrors [6–12]. The main drawback of this method is the sensitivity of the hollow fibre to alignment [13] and its limitation in the achievable energy throughput [14].

Our approach is based on our prediction of ultrashort pulse generation and self-compression [15] of infrared femtosecond laser pulses in noble gases, and recent experiments have indeed shown remarkable pulse compression [13].

When a sufficiently intense ultrashort laser pulse propagates in a transparent medium, a dynamical equilibrium of two main counteracting physical effects leads to the formation of a filament: focusing due to the optical Kerr effect and defocusing due to a plasma generated by multiphoton ionization.

The self-phase modulation in the filament provides the desired spectral broadening of the incident pulse up to more than one octave-spanning spectra while the self-guiding acts as a spatial filter leading to an excellent mode quality [13, 16–18]. This compression scheme is easily implemented because it does not require meticulous optical alignment of a hollow guiding tube. The energy of the resultant pulse is in the sub-millijoule range, comparable to that achieved with the hollow-fibre compression scheme [19] but expected to be scalable to the multi-mJ regime by using gases with higher ionization potential than argon. A most intriguing effect is self-compression to the few-cycle regime through filamentation which was predicted by our numerical modelling suggesting a simple design to yield even shorter pulses in the attosecond domain through the generation of high-order harmonics in a cell that acts both for pulse self-compression and for harmonic generation.

Up to now, the experiments employed cells filled with a uniform gas distribution. As we show further in this paper, this has turned out to be an efficient approach to generating few-cycle pulses and to be easy to implement. Nonetheless, there are possibilities for further significant improvements. Theoretical calculations as well as numerical simulations indicate that a non-uniform gas distribution inside the cell could yield significantly better pulse compression. This non-uniform gas distribution could avoid multiple compression and decompression stages, thereby improving the energy throughput, and allow the extraction of isolated single optical cycle pulses without the need of separate pulse recompression through negative dispersive elements [15].

2. Experiment

In our previous publication, we investigated the dependence of the generation of few-cycle pulses under the initial and environmental conditions with the perspective to produce high-fidelity ultrashort pulses [20]. Besides studying the influence of pressure and energy on the pulse duration, beam pointing stability and M^2 measurements were performed, demonstrating that the filamentation process does not significantly affect the pointing stability of the beam and that beams with exceptional quality ($M^2 \approx 1$) can be generated. This implies good focusability, an important prerequisite especially in high-field physics [20]. We also attributed the limit in achievable duration to the limited optical bandwidth of our chirped mirrors.

By employing an improved set of chirped mirrors, intense CEO-phase stabilized pulses as short as 5.1 fs can be obtained. The setup of this new experiment follows the guidelines that were found to be successful in the past and is shown in figure 1.

We started with the output of a commercial laser (CPA laser system, Femtolasers). It consists of a Ti:sapphire-based oscillator and an amplifier that can deliver up to 1 mJ,

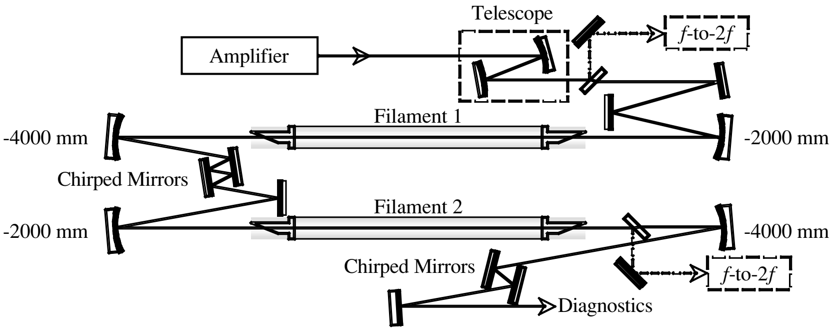

Figure 1. Experimental setup. Intense laser pulses are focused by a spherical mirror (ROC = −2000 mm) into the argon-filled 160 cm long cell (1) where filamentation occurs. After collimation with a curved mirror (ROC = −4000 mm) and recompression by four bounces on chirped mirrors (CM), the beam is refocused by spherical mirror (ROC = −2000 mm) for the formation of a second filament in the second cell and subsequent collimation through curved mirror (ROC = −4000 mm) and group-delay dispersion compensation by two bounces on ultra-broadband double-chirped mirrors (CM). SPIDER measurements are performed at three different positions: after the amplifier, after the first filament and after the second filament. The signals for the f–$2f$ interferometers are taken after the telescope and after the second cell.

approximately 30 fs long pulses with a repetition rate of 1 kHz. The amplified pulses are CEO-phase stabilized and the spectrum is centred at 800 nm.

The beam out of this system was imaged with a 2:1 telescope to a diameter (FWHM) of 6.4 mm. The beam was loosely focused with a −2000 mm ROC silver mirror into the first glass cell, which was filled with argon at a pressure of 850 mbar. Only part of the energy output of the amplifier, 0.68 mJ, was focused into the first cell leading to the formation of a short ($<$10 cm long) and clearly visible filament. Another small fraction of the amplifier output was sent to an f–$2f$ spectral interferometer in order to measure and correct for slow drifts of the oscillator CEO phase [21].

We characterized the pulse shapes before, between and after the two filaments using SPIDER [22]. The input laser pulse, shown in figure 2(a), had a duration of 34 fs and a slight chirp. The resulting broadened spectrum after the first filamentation stage was recompressed using negative-dispersion chirped mirrors (Femtolasers) yielding essentially transform-limited pulses as short as 11 fs (figure 2(b)) where the Ar pressure in the first cell was optimized to generate the shortest and cleanest output. The total output energy was 0.65 mJ, corresponding to $>$95% of the input energy. In order to select only the core part of the filament and cut out the reservoir part, the beam was apertured before being refocused into the second filament cell filled with argon at a pressure of 900 mbar.

The aperturing of the beam highlighted that the effective energy carried by the compressed pulse was 0.56 mJ ($\sim$82% of the initial energy), and the resulting spectrum is shown in figure 2(c). The spectrum spans more than one optical octave and supports transform-limited pulse durations of less than 2 fs. Using ultra-broadband chirped mirrors, we compressed the pulses to a duration of 5.1 fs, with an energy in the useable inner area of 0.18 mJ. This corresponds to a total compression factor of $>$6.8 with a total energy efficiency of 26% in an excellent central mode. A pressure scan inside the second cell revealed that the filamentation process is insensitive to gas pressure variations within approximately 100 mbar. Compared with our previous results, this confirms that the chirped mirrors remain the main limiting component in the generation of few-cycle laser pulses using filamentation.

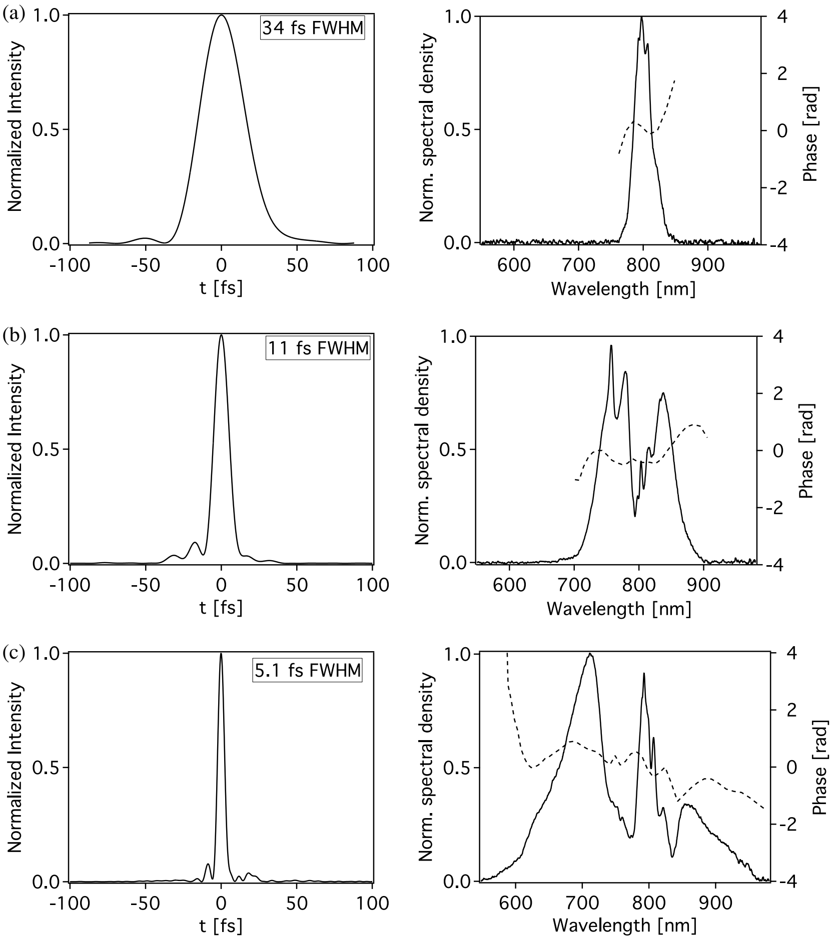

Figure 2. Pulse shapes and associated spectra and phases of (a) the input amplifier pulse, (b) after the first filament cell (11 fs) and (c) after the second filament cell (5.1 fs).

Since an octave-spanning spectrum is available at the output of the second cell, it suggests the possibility of employing it for the measurement of the slow drift of the CEO phase of the amplified pulses. In the traditional configuration, the CEO phase stability of amplified pulses is measured by f–$2f$ spectral interferometry (SI) directly after the amplifier, requiring an octave-spanning spectrum [21], which is typically obtained by focusing the amplifier pulses into a thin sapphire plate, leading to white-light generation. The information on the CEO phase can be retrieved from an analysis of the fringe pattern arising from the interference in the spectral domain, of two overlapping spectral regions: one component ($2f$) originating from the frequency-doubled red wing of the spectrum and the other (f) already present in the blue wing of the octave-spanning spectrum itself [23]. The fringe pattern intensity $I(f)$ for a single laser shot is then characterized in terms of spectral frequency (f) as

$$I(f) \propto \cos[2\pi f\tau_0 + \theta + \alpha(f)] = \cos[\Theta(f)], \qquad (1)$$

where τ_0 is the average time delay between the interfering frequency components, θ is the CEO phase and the constant $\alpha(f)$ reflects shot-to-shot fluctuations in τ_0 to the overall

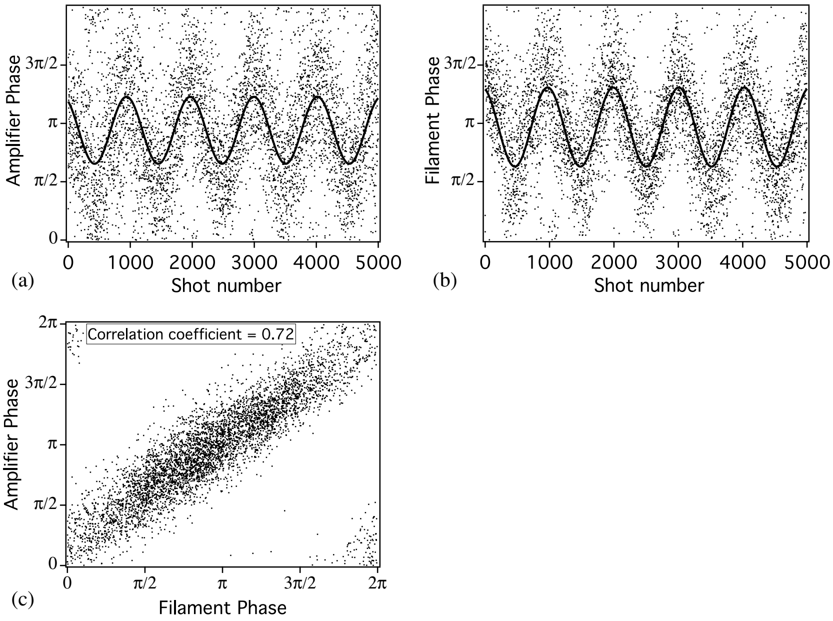

Figure 3. CEO fringe phase modulation measured simultaneously by (a) the amplifier f–$2f$ setup and (b) the filament output, together with fitted sinusoids (solid lines). The distributions have rms deviation: (a) 1.30 rad and (b) 1.07 rad. (c) Correlation of the shot-to-shot phase values, linear correlation coefficient of 0.72.

phase Θ, as well as higher order phase variations resulting from, e.g., the chirp of the input pulse and nonlinear pulse propagation.

We constructed a second f–$2f$ interferometer after the filaments, as shown in figure 1, to explore the feasibility of using the octave-spanning spectrum generated by the filaments directly for SI, to investigate the CEO phase behaviour and to discover whether this system could be used as a feedback to correct for slow CEO drifts in the oscillator.

The interference spectra of the two f–$2f$ interferometers were coupled into separate Acton Spectra Pro 300i spectrometers and read out by 2048-pixel Basler L101k-2k line-scan CCD cameras. The spectral resolutions of f–$2f$ were 0.05 nm/pixel for the first and 0.025 nm/pixel for the second. Both systems could acquire synchronized spectra at the full repetition rate of the laser (1.007 kHz). Note that only the ‘fast’ CEO feedback loop of the oscillator was in operation since we used the ‘slow’ feedback loop for our CEO measurement.

In figure 3, we show that the filamentation process does not detrimentally affect the CEO phase stability of the incoming pulse. We applied a 1 Hz sinusoidal modulation to the oscillator CEO phase while only one (oscillator) stabilization loop was active and the slow loop was switched off. Figures 3(a) and (b) show the measured CEO phases before and after the filaments, respectively, for corresponding laser shots. A correlation plot of the two measurements, figure 3(c), shows that there is excellent agreement between the two measured values. By comparing the widths of the distributions, we find that the CEO phase measurement after the filaments has an rms spread of 1.07 rad, compared to 1.30 rad after the amplifier for the same laser shots. The smaller spread may be linked to a lower sensitivity to fluctuations of input pulse characteristics of the white-light generation process in argon compared to the sapphire.

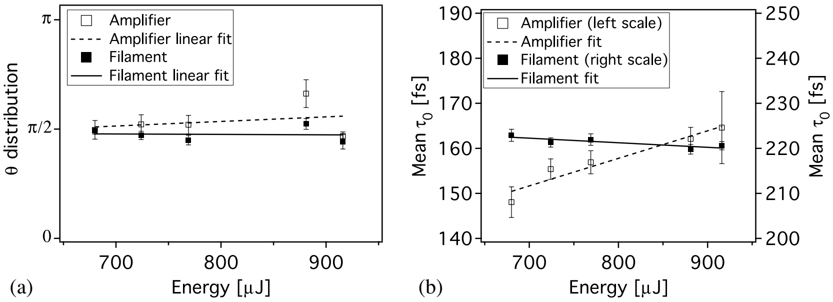

Figure 4. (a) Simultaneously measured CEO phase (θ) distribution Gaussian widths from the amplifier f–$2f$ fringes and the filament fringes with oscillator CEO phase lock on, for a range of pulse energies from the amplifier, where error bars represent the error in the width of the Gaussian fit to the θ distribution. (b) Simultaneously measured mean τ_0, where the error bars represent the width of τ_0 distribution. Each data point corresponds to a sample of 2000 shots.

Due to the nonlinear nature of the white-light generation process, it is important to study the behaviour of the interference fringes with respect to the input energy, since pulse energy fluctuations can be a primary source of noise when interpreting the CEO phase measurements. It is therefore useful to compare simultaneously the effects on the two f–$2f$ interferometers for different incident energies. We chose to vary the energy using silvered normal density filters instead of varying the amplifier pump laser power to ensure unchanged input pulse duration. The amplifier power fluctuations at the operating pump level were measured to be $\sigma = 1.4\%$. Although the energy variation can affect the filamentation process changing the pulse duration achievable after recompression, we verified that this does not significantly affect the fringe quality and stability measurable using the spectrum generated during the filamentation process. This is probably since the generated spectrum is much broader than one optical octave for a wide range of incoming pulse durations. Spectra spanning more than two octaves were recorded for input pulse durations ranging from 11 to 26 fs.

With the oscillator phase lock loop on, the CEO fringes from the amplifier f–$2f$ and from the filament were recorded simultaneously. The distribution of the CEO fringe phase θ was analysed for 2000 shots for different amplifier energies and then fitted with a Gaussian distribution. The results are shown in figure 4(a) where the error bars represent the error in the width of the Gaussian fit to the θ distribution.

As can be seen from figure 4(a), the width of the CEO fringe phase distributions measured by the two setups is similar within the fitting error bars. This is an indication of the reliability of the measurement performed employing the filament spectrum. Furthermore, the spread of the Gaussian distribution is smaller in the measurement employing the filament spectrum than in that using the sapphire plate, which may be linked to a lower sensitivity to pulse parameter fluctuations in the white-light generation process by filamentation as compared to that employing the sapphire plate.

This insensitivity represents one significant advantage of using the filament-broadened spectrum for the f–$2f$ measurement.

For the same data set, the comparative behaviour of τ_0 was also examined and the mean value arising from 2000 acquisitions is shown in figure 4(b), where the error bars represent the width of the τ_0 distributions. Besides the different mean values measured by the two setups, figure 4(b) shows that, for our conditions, the spread of the τ_0 distributions as a

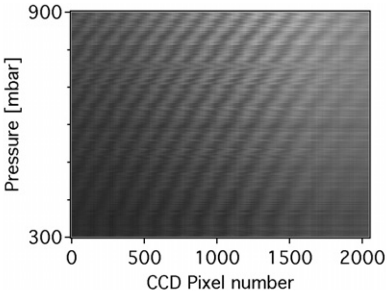

Figure 5. Spectral interference fringes recorded by the filament f–$2f$ setup for Ar pressure varying from 300 to 900 mbar.

function of laser energy is significantly lower from the second filament than that from f–$2f$ employing white-light generation in sapphire. Since, as can be seen from equation (1), shot-to-shot fluctuations in τ_0 will directly translate into uncertainty in determining the CEO phase θ; this means that the employment of the octave-spanning spectrum arising from the filamentation process will have a beneficial influence on the reduction of this uncertainty as well.

To test the insensitivity of the filament formation to environmental conditions, we recorded the f–$2f$ interference fringes after the second filament while scanning the Ar pressure from 300 to 900 mbar. As shown in figure 5, fringes remained visible over the entire range, indicating that the spectral broadening at all times yielded an octave-spanning spectrum, and that the CEO phase was preserved. The apparent discontinuity at 760 mbar was related to the oscillator phase lock. Small changes in the beam energy and/or focusing conditions could slightly modify the operating conditions for the shortest and cleanest pulse, but figure 5 indicates that the flexibility of the two-stage filamentation process can be adjusted to yield ultrashort, CEO phase stable pulses for a wide range of input parameters.

3. Conclusion

In conclusion, CEO-stabilized pulses as short as 5.1 fs were obtained employing a dual-stage filamentation setup and an enhanced set of chirped mirrors. The current limitation of this alignment-free method is still the use of double-chirped mirrors with limited mirror structures to compensate for the system dispersion. These shortcomings could be overcome by carefully adapted mirror structures and fine-tuning of the gas pressures, or with the design of a cell establishing an appropriate pressure gradient, which would lead to the feasibility of obtaining full self-compression avoiding the employment of chirped mirrors.

By measuring the CEO phase before and after filamentation, we were able to demonstrate that the CEO phase changes of the input amplified pulses are well measurable by f–$2f$ spectral interferometry of the octave-spanning spectrum produced from the filaments. The CEO phase variations into and out of the two stages of filamentation were well correlated. For the setup used, the CEO fringes produced by the filamentation process demonstrated smaller fluctuations in τ_0, as well as a less sensitive dependence on the energy variations of the amplifier output. In particular, CEO fringes were present under a wide range of conditions of pulse energy and cell pressure, suggesting the serious advantageous possibility of utilizing the spectrum for CEO stabilization of the amplifier output when making short pulses via filamentation.

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