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Generation of intense, carrier-envelope phase-locked few-cycle laser pulses through filamentation

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ABSTRACT Intense, well-controlled light pulses with only a few optical cycles start to play a crucial role in many fields of physics, such as attosecond science. We present an extremely simple and robust technique to generate such carrier-envelope offset (CEO) phase locked few-cycle pulses, relying on self-guiding of intense 43-fs, 0.84 mJ optical pulses during propagation in a transparent noble gas. We have demonstrated 5.7-fs, 0.38 mJ pulses with an excellent spatial beam profile and discuss the potential for much shorter pulses. Numerical simulations confirm that filamentation is the mechanism responsible for pulse shortening. The method is widely applicable and much less sensitive to experimental conditions such as beam alignment, input pulse duration or gas pressure as compared to gas-filled hollow fibers.

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1 Introduction

Time resolved measurements of electron dynamics in atoms and molecules, which ultimately govern any chemical reaction [1], wake-field particle acceleration, and the production of energetic proton beams [2], benefit tremendously from laser sources delivering intense, well-controlled light pulses with pulse durations of only a few optical cycles [3–5]. Such sources fulfill a central task in the generation of attosecond extreme ultraviolet (XUV) pulses [6]. Single attosecond pulse generation exploits infrared (IR) pulses of only 2–3 cycles that carry enough energy to be loosely focused to intensities in excess of several 10^{14} W cm^{-2}. The availability of attosecond XUV pulses in turn opens the way towards attosecond-time domain spectroscopy [7]. As a first demonstration in this new field, the inner-shell decay in a noble gas atom could recently be directly determined [8].

The main obstacle in these fields is the difficulty in producing suitable driving laser pulses [3]. This requires not only intense, reproducible few-cycle pulses, but also a control of the carrier phase with respect to the pulse envelope [9–11]. Indeed, with intense few-cycle laser pulses, the electric field amplitude rather than the intensity envelope becomes the determining factor. The electric field oscillations must then be identical from pulse to pulse; i.e. they must be carrier-envelope-offset (CEO) phase-locked; and currently only a few laser laboratories master the technology. To become routinely available, simplification in the techniques of few-cycle laser pulses production is necessary.

2 Generation of phase-locked intense few-cycle laser pulses

So far, the only successful demonstration of CEO phase-locked intense few-cycle laser pulses starts with chirped-pulse amplification (CPA) Ti:sapphire laser systems [12] typically providing 25- to 30-fs pulses (9 to 11 optical cycles at a center wavelength of 800 nm) with energies of a few millijoules per pulse [13]. In order to generate few-cycle pulses, the pulse spectrum is artificially broadened through self-phase modulation via propagation inside a gas-filled hollow capillary [14], and then recompressed spectrally. The capillary works in two ways: Firstly, the light pulse is tightly confined inside the capillary, which acts like an extended spatial filter resulting in a high spatial coherence, and secondly its high intensity is maintained over an extended distance, leading to large spectral broadening through the nonlinear interaction with the gas medium. The emerging pulse is then spectrally recompressed typically with chirped mirrors. Typically pulses in the range of 6- to 8-fs can be routinely generated with pulse energies in the range of 100–400 μJ. The shortest pulses obtained with this technique yield pulses as short as 3.8 fs with an energy of 15 μJ in the near-IR [15] and more recently, 2.8 fs with 500 nJ [16]. However,both energies are too low to be used directly in the above mentioned fields. CEO phase-stability requires the implementation of two phase-locked loops: a fast one for the laser oscillator [9, 17] and a slow one to correct for drifts in the laser amplifier [18, 19].

While gas-filled hollow fibers constitute the key ingredient to produce few-cycle pulses, they have serious inherent limiting factors: Energy scalability is limited (to about 0.4 mJ); beam pointing fluctuations of the incoming beam directly translate into unwanted

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energy and pulse parameter fluctuations of the outgoing pulses [20], a bane for high field physics experiments; and the performance critically depends on the quality of the hollow fiber. Therefore alternative approaches such as chirped pulse optical parametric amplification are being pursued [21]; the generation of sub-3-cycle pulses, however, remains to be demonstrated.

3 Self-generation of few-cycle pulses through filamentation

We have found an experimental approach, which considerably simplifies the procedure. Instead of relying on the external guiding effect of the capillary, we exploit the self-guiding effect that an intense optical pulse can experience when it propagates in a transparent gas [22]. The self-guiding effect removes the constraints of the capillary, while it keeps and even expands on their beneficiary aspects. These effects are spatial filtering via self-guiding, spectral broadening via self-phase modulation, pulse self-shortening [23] and CEO phase preservation [24]. As will be shown, under suitable conditions, that are easy to implement, one can obtain intense, reproducible, CEO phase-locked, few-cycle laser pulses with excellent beam quality by simply focusing a laser beam in a gas cell and recompressing the output. Under optimal conditions, our modeling predicts that filamentation can even lead to pulses very close to their fundamental limit of a single cycle without the need of a final recompression stage [25].

Filamentation, or self-guiding, occurs when a sufficiently intense ultrashort laser pulse propagates in a transparent medium. Two physical effects play a major role in the formation of a filament: the self-focusing effect due to intensity-dependence of the refractive index of the medium, and defocusing due to the formation of a plasma [26]. Both effects are schematically illustrated in Fig. 1.

It is important to stress that a filament is not a steady state. It has rich temporal dynamics involving also other effects such as group velocity dispersion, self-phase modulation, pulse self-steepening and the Raman effect [27]. Despite the complex spatio-temporal coupling that occurs from their combined action, the pulse takes the form of a narrow beam (100 μm) surrounded by a reservoir of laser energy. This reservoir feeds the filament core which becomes largely insensitive to initial conditions. It maintains high peak intensity around 10^{13} W/cm [24] over a long distance, exceeding by orders of magnitude the Rayleigh length even if the underlying dynamics is complex and changing rapidly.

In addition to the pulse spatial contraction, which is induced by the intensity dependence of the refractive index, broadening of the pulse spectrum arises due to the time variation of the refractive index ($n = n_0 + \Delta n_{\mathrm{Kerr}}(I) = n_0 + n_2 I$). The corresponding phase variation adds new frequency components to the spectrum: Red frequency components on its ascending, and blue components on the descending side. This results in a variation of the instantaneous frequency with time (chirp). The addition of new frequencies develops to the point of creating a quasi-continuum extending over the entire visible spectrum and into the infrared. This opens the possibility to obtain shorter pulses, by a suitable retardation of the red components with respect to the blue ones.

One additional aspect of femtosecond filamentation, which has no counterpart in external guiding (hollow fiber), is pulse self-compression [25, 28, 29]. The different nonlinear effects occurring in filamentation lead to an important restructuring of the pulse time profile. Some of these effects, such as self-focusing, pulse self-steepening, and self-phase modulation, act instantaneously. These effects tend to accumulate laser energy to the ascending part of the pulse, whereas the time-delayed effects, such as photo-ionization and the Raman effect, tend to cut off its trailing part. Their combination eventually leads to the formation of pulses that are significantly shorter than the incident pulse. One important facet, previously overlooked, is the fact that pulse reshaping by filamentation can be effective down to the fundamental limit of nearly one optical cycle.

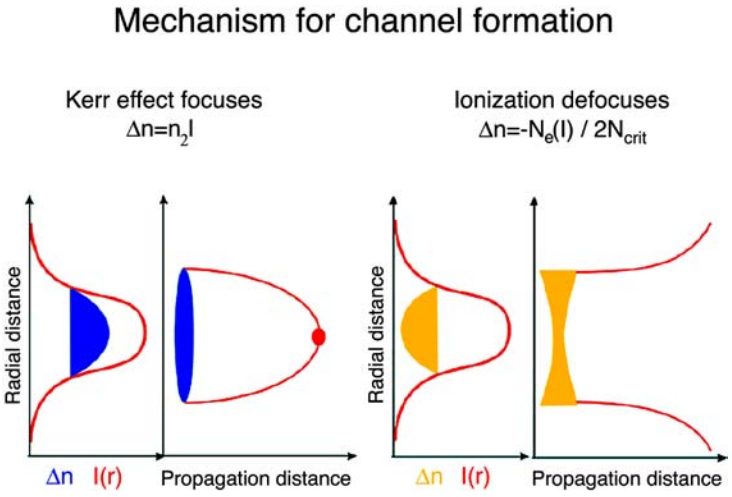

FIGURE 1 Mechanism for channel formation: For a typical beam intensity profile with a maximum on axis, the intensity dependent refractive index ($n = n_0 + \Delta n_{\mathrm{Kerr}}(I) = n_0 + n_2 I$) acts like a succession of increasingly converging lenses. In the absence of a limiting effect, this would lead to a beam collapse on axis if the pulse power exceeds a critical value $P_{\mathrm{cr}} = \lambda^2/2\pi n_0 n_2$, which is several Gigawatt in air at 800 nm. On-axis beam collapse is arrested by multi-photon ionization. This occurs typically at intensities around 10^{13} W/cm^2 in gases, giving rise to a weakly ionized plasma with an electron density typically around 10^{16} cm^{-3}, and a corresponding reduction of the local refractive index $\Delta n = -N(I)/2N_{\mathrm{cr}} \sim 10^{-4}$. Here, $N(I)$ denotes the intensity dependent free electron density and N_{cr}, the density above which the plasma becomes opaque. Thus, multi-photon ionization acts as a strong regulating mechanism, limiting the beam intensity on axis and the dissipation of laser energy during propagation

4 Experimental results

We found that optimum pulse shortening from 43 fs to 5.7 fs could be achieved by using two successive gas cells at pressures of 840 mbar and 700 mbar of argon, as shown in Fig. 2.

The CEO-phase-locked input pulse with an energy of 0.84 mJ was loosely focused into the first cell where it generated a 10–15 cm long filament roughly

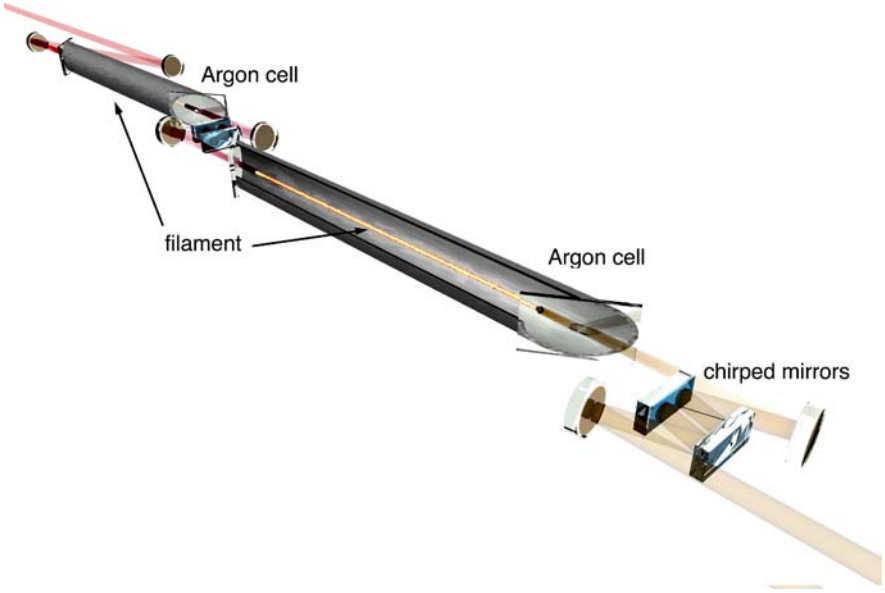

FIGURE 2 Experimental setup: Two cells are filled with argon at 840 mbar and 700 mbar respectively. CEO-phase-locked 43-fs IR pulses with 0.84 mJ energy form a 10–15-cm long filament in the middle of the first cell and are compressed with chirped mirrors to 10.5 fs at 790 μJ. Sending those pulses into the second cell results in a filament also 15–20 cm long that, after final recompression, leads to a 5.7-fs pulse with 45% of the initial pulse energy in an excellent spatial profile

in the center of the cell, as estimated from the length of the scattered broadband continuum. The emerging spectrum was recompressed with chirped mirrors, resulting in a shortening by a factor four while retaining 94% of the input energy. Sending the 10.5-fs pulse into the second gas cell, another 15–20 cm long filament was formed, leading after chirped-mirror recompression, to a 5.7-fs pulse with 45% of the initial pulse energy,i.e. 0.38 mJ, in an excellent spatial profile. Figure 3a and b show a spectral phase interferometry for direct electric-field reconstruction (SPIDER [30]) measurement of the pulse temporal profile, spectral phase and spectrum measured at a kHz single shot rate [31]. The excellent spatial quality of this pulse is revealed in the inset of Fig. 3a. CEO phase stability is confirmed through f-2f spectral interferometry [9, 19] with the persistence of fringes confirming the phase-preserving nature of the filaments. Figure 3c shows a time series of such a measurement where the CEO-lock, with one feedback for the oscillator only, has been switched on after roughly 1.2 s. The measurement clearly confirms the formation and steadiness of fringes, hence confirming CEO-phase conservation. Figure 3e shows the spectrum measured after the second gas-cell (before the chirped mirror compressor), optimized for maximal spectral width at a pressure of 1060 mbar. It supports a transform-limited pulse duration of 1.75 fs shown in Fig. 3d, assuming a flat spectral phase. The spectrum after the first cell, at a pressure of 840 mbar, is shown in Fig. 3f.

We would expect that with better dispersion compensation (i.e. better chirped mirrors) we could obtain even shorter pulses than the demonstrated 5.7 fs.

We attribute the observed differences between results shown in Fig. 3a

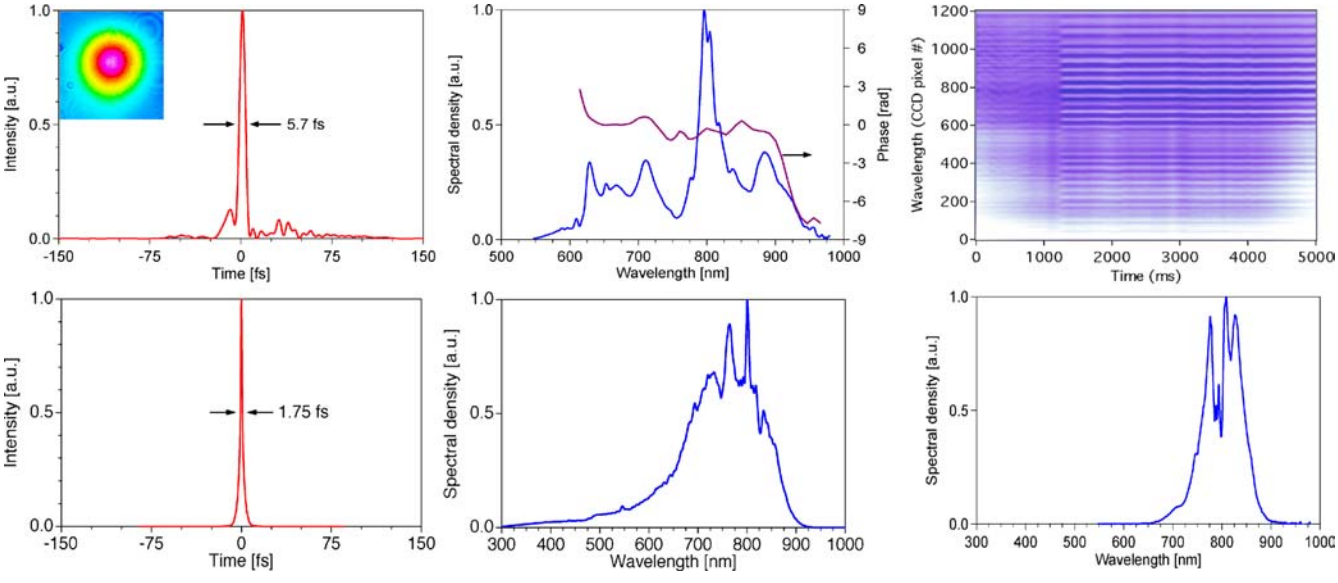

FIGURE 3 (**a**) Temporal intensity profile of compressed pulses as obtained with SPIDER for pressures of 840 mbar and 700 mbar of argon. The full width at half maximum (FWHM) pulse duration of 5.7 fs corresponds to 2.1 optical cycles. The spatial profile, measured with a high resolution CCD (WinCam, DataRay) is excellent, as can be seen in the *inset*. The associated spectrum and spectral phase are given in (**b**). (**c**) Time series from an f-2f spectral interferometry measurement. The CEO-phase lock is switched on after roughly 1.2 s with the emergence of fringes confirming the CEO phase lock. (**d**) Transform-limited pulse duration supported by the normalized spectrum after the second gas-cell, shown in (**e**), before the chirped mirror compressor, assuming a flat spectral phase, perfect bandpass and phase characteristics of the chirped mirrors. (**f**) Normalized pulse spectrum after the first cell. The pressures in the first and second cell were optimized for maximal spectral broadening and were measured to be 840 mbar and 1060 mbar respectively

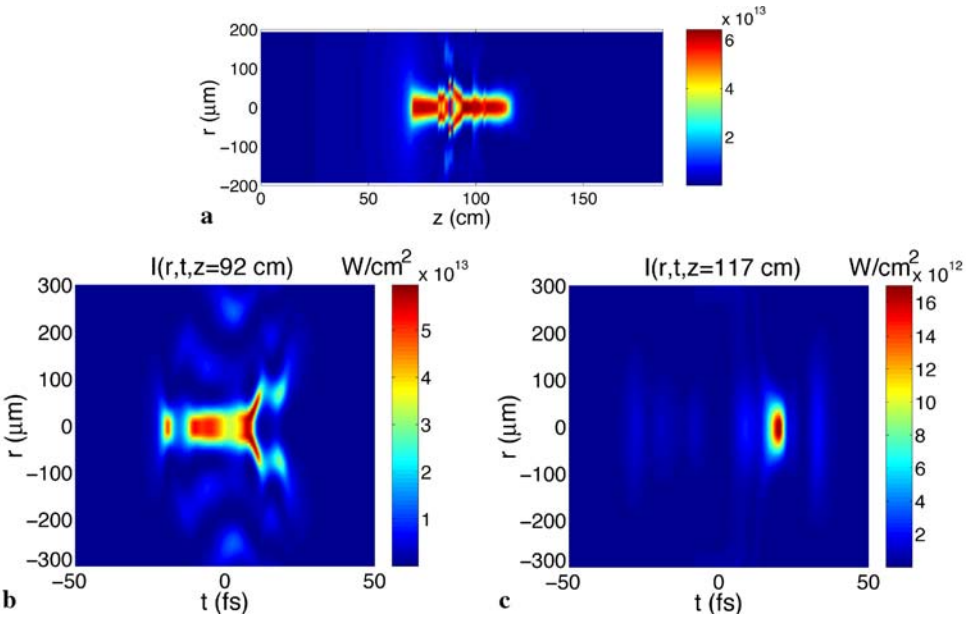

FIGURE 4 Modeled pulse propagation for second gas cell: (**a**) Intensity profile confirming the formation of a filament. (**b**), (**c**) Space-time profile of the pulse at two locations inside the filament. (**b**) is calculated 92 cm from the focusing mirror for the second gas cell. Further propagation (25 cm) leads to a tightly spatially and temporally confined light bullet

and d, respectively Fig. 3b and e to a large extent as follows: First, the difference in the spectra is obviously due to the chirped mirrors. The phase characteristics of the chirped mirrors used in the experiment are insufficient to produce a pulse much shorter than 6 fs. This limitation not only modifies the spectrum but, more importantly, limits the compression. The dispersion introduced by the mirrors being known, it will be possible to design and fabricate appropriate chirped mirror [32, 33] structures to obtain better compression. Secondly, the dispersion in the exit window, consisting of a 0.7 mm thick fused silica plate at Brewster angle, has a significant effect on such ultra broadband pulses. For instance, it increases the duration of a transform-limited single-cycle pulse seven-fold.

The robustness of the present experimental technique to obtain shorter pulses with respect to variations in input pulse and operating parameters is essential. We have therefore carefully checked the influence of these changes on the final pulse energy, duration and spatial profile and have found that this method is surprisingly insensitive to such changes. Therefore self-compression through filamentation is a very robust and reliable method to generate intense few-cycle pulses, e.g. a change in pressure of 100 mbar influenced the pulse duration by 0.2 fs only.

5 Numerical simulation

The measured results can be well reproduced by a code calculating the propagation of intense short laser pulses in a transparent medium [34]. Our code solves the 3D nonlinear envelope equation describing the evolution of the field envelope. This approach has been demonstrated to be valid down to the single-cycle limit [35]. It accurately reproduces experimental results on filamentation in gaseous and solid media. The model includes the physical effects of diffraction, group velocity dispersion, self-focusing, self-steepening, space-time focusing, Raman scattering, ionization, plasma defocusing as well as photo-absorption and plasma recombination.

We have calculated the propagation inside the second gas cell, by using various input conditions close to the measured input pulse parameters, such as the pulse amplitude and phase (obtained from a SPIDER measurement). Figure 4a gives the calculated spatial distribution of laser intensity, which confirms the formation of a filament. Figure 4b and c show the spatio-temporal intensity distribution at two different propagation distances. The temporal evolution is illustrated by the transition between Fig. 4b and c. We find specific locations where self-shortening of the laser pulse takes place over several centimeters with a minimum pulse duration nearing 1.3 optical cycles. Interestingly, the calculated phase front of an isolated self-compressed pulse such as in Fig. 4c reveals a near flat surface over several centimeters, evolving into a parabolic shape (diverging beam). In fact, the comparison between numerical and experimental results shows that the pulse of Fig. 3a with a diverging beam of excellent quality was collected at a location close to optimal. It suggests that the final external compression stage is required by the presence of the exit window.

6 Conclusions and outlook

We have demonstrated experimentally, and confirmed theoretically, that it is possible to obtain nearly single cycle, CEO phase-locked pulses via filamentation in a gas for the first time. This technique presents a robust and simple alternative to hollow fiber pulse compression. The following aspects are unique to this technique:

Flat phase front and intensity clamping: filamentary propagation has been shown to yield an output pulse with a flat phase front, a feature confirmed by our

simulations. Furthermore, because filamentation is arrested by multi-photon ionization, a process scaling as intensity to the eleventh power in argon, a very effective intensity clamping is achieved. The fact that a light pulse with nearly constant high intensity and with a flat phase front interacts over an extended distance with a noble gas leads to very efficient generation of high order harmonics [36]. It eliminates the influence of the Gouy phase shift [37], which occurs when a beam goes through a focusThis may give a decisive advantage in experiments involving a coherent interaction between several waves at different frequencies where phase matching is required [38].

Energy up-scaling: for an input pulse of power well above critical for self-focusing, multiple filaments tend to form, each filament carrying the same energy of about 1 mJ [28]. This appears at first sight to be a serious limitation for energy up-scaling. However, it has also been shown recently [34] that it is possible, with simple amplitude or phase masks, to organize the multi-filaments according to a prescribed pattern. If closely spaced, such mutually coherent filaments have been shown to coalesce [39].

In our opinion, our experimental simplification will help making well-controlled, i.e. CEO phase-locked, single-cycle optical pulses much more accessible, which is a crucial issue in attoscience and other emerging fields.

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