Generation of intense few-cycle laser pulses through filamentation – parameter dependence

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Abstract: Intense few-cycle laser pulses as short as 5.1 fs are generated though selffilamentation in a noble gas atmosphere. We study the dependence of the laser pulse fidelity on the driving pulse profile and chirp as well as on the gas parameters, quantify their pointing stability and spatial quality.

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1. Introduction

In recent years intense few-cycle pulses in the near infrared have enabled many novel experiments, particularly in high-field science, and paved the way towards time-resolved spectroscopy on an attosecond time scale [1-3]. Intense driving pulses, comprised of only two or three electric field oscillations have led to the successful generation of single attosecond pulses [4], which were used to track motions of inner-shell electrons in atoms [5]. Most experiments in high-field science are sensitive to the electric field amplitude of the laser pulse making stabilization of the carrier to envelope (CEO) phase [6] a necessity for reliable attosecond pulse generation [4]. In the past, the generation of such intense CEO-phase stable few-cycle pulses has solely been achieved by spectral broadening of an amplified pulse in a gas-filled hollow-core fiber and subsequent compression with chirped mirrors [7-9]. We have recently shown [10] that intense few-cycle pulses as short as 5.7 fs can be produced with their

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CEO phase preserved, simply through filamentation [11] thereby making arduous coupling into a fiber unnecessary.

2. Experiment

We studied the dependence of the generation of few-cycle pulses on the initial and environmental conditions with the perspective to produce a high-fidelity ultrashort pulse. The experiment consisted of taking the output from a commercial amplifier (CPA laser system, Femtolasers) and generating filaments in argon in two stages (Fig. 1). We characterized the laser pulse at three different positions in the beam line: out of the amplifier, just before the first gas cell, after the first cell and chirped-mirrors, and finally after the second cell and chirped-mirrors (labelled $\bigcirc, \oslash, \bigcirc,$ in Fig. 1). Measuring the pointing stability after the filament, we confirm that the self-guidance process of the laser beam does not measurably influence the pointing stability. We have made caustic measurements to determine the beam quality parameter M² and have found it, as expected from self-guiding, to be close to the diffraction limit, indicating excellent focusability, an important prerequisite especially in high-field physics.



Fig. 1. Experimental setup. Intense laser pulses are focused by a spherical mirror (R1, ROC = -2000 mm) into the argon-filled 160-cm-long cell (1) where filamentary propagation occurs. After recompression by four bounces on chirped mirrors (CM), the beam is refocused by spherical mirror R2 (ROC = -2000 mm) for the formation of a second filament in cell 2 and subsequent group-delay dispersion compensation by 6 bounces on ultra-broadband double-chirped mirrors (CM). SPIDER measurements are performed at three different positions (label $\bigcirc, \bigcirc, \bigcirc$): after the amplifier, after the first filament and after the second filament. Beam pointing fluctuations are measured behind the second filament with a position sensitive detector (PSD) and a focusing lens L (f=50 cm).

The experimental setup is shown in Fig. 1 with the laser system delivering CEO phasestable 33 fs pulses with energy of 0.85 mJ at a repetition rate of 1 kHz. The beam width was decreased to a FWHM of 6.4 mm before being loosely focused with a -2000 mm ROC silver mirror into the first argon-filled cell where the pulse filaments through a dynamic balance between Kerr-lensing and plasma-induced defocusing. Nonlinear effects lead to the generation of new frequency components and to a spectral redistribution in energy [12]. The emerging pulse is positively chirped, which is compensated for by commercially available chirped mirrors. This process is repeated in the second cell and, after chirp compensation, the pulses are characterized by spectral phase interferometry for direct electric field reconstruction (SPIDER)[13, 14].

Since filamentation is a highly non-linear process, it is, for practical purposes, necessary to investigate its influence on the pointing stability. We recorded the beam's intensity profile after the second gas cell with a high-resolution 14-bit CCD camera (Wincam, Dataray Inc.) and determined the (x,y) coordinates of the intensity centroids for 1500 consecutive laser shots (position, see Fig. 1). Two different measurements were applied to distinguish the

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intrinsic beam pointing instability from the one caused by filamentation: First, the laser propagated through the two interaction cells under vacuum and therefore without undergoing filamentation. Second, the cells were filled with argon (at 800 mbar, and 700 mbar respectively) and the pulse filaments. The results are shown in Fig. 2 and indicate that no significant influence is caused by filamentary propagation. Without filamentation, the beam pointing variation was 0.5% rms and with filamentation 0.6% rms, whereas the shot-to-shot pointing fluctuations were below 0.8%. Similar results have been obtained at other gas pressures.



Fig. 2. Centroid movement of the laser beam at the exit of the filamentation setup for 1500 consecutive laser shots. The data is recorded with a 14 Bit CCD camera (Wincam, DataRay Inc.) for the evacuated cell ((a); no filamentation) and for the case when the pulse filaments (b).

We observed that the initial temporal pulse shape significantly influences the process of spectral broadening during filamentation. To investigate this correlation we systematically varied the input pulse shape by altering the compressor settings of the amplifier. The gas pressure in the first cell was set to 800 mbar.



Fig. 3. Temporal shape of the laser pulses with different chirps (left) creating the first filament. Group delay dispersion of the input pulse is altered systematically by varying amplifier compressor settings (see Table 1) yielding pulses from 59.8 fs to close to transform-limited 33.2 fs pulses. The shortest pulse after filamentary spectral broadening and compression by chirped mirrors (right) was measured for a positively chirped 41.7-fs input pulse (bold, number 3). Pulse 3 exhibits the smallest prepulse-to-mainpulse peak ratio and yields broadest spectrum after the first filament.

Both, the initial (Fig. 3, left) and the pulse emerging the first filament (Fig. 3, right) were reconstructed with SPIDER (Fig. 1 pos (1) and (2)). Comparing corresponding input and

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emerging pulse durations no clear correlation was observed. To identify the key factor a set of parameters was deduced from the initial pulse such as peak power, group delay dispersion (GDD), slope of the rising pulse edge, and the contrast ratio of pre-to-main pulse for the input laser beam. A summary of these quantities is shown in Table 1 for the four different pulses labeled in Fig. 3. Shortest input pulse duration (33.2 fs, pulse 1) expected to cause strongest non-linear spectral-broadening response in the gas yields a spectrum supporting only 15.2 fs (transform-limited (TL)) after filamentary propagation. Even though carrying highest peak power (21.5 GW) and steepest pulse slope, the measured filament pulse duration was 14.8 fs, which was slightly shorter than its TL due to spectral phase distortions.

Table 1. Table of laser parameters for amplifier and filament output for four different pulses. The amplifier output was optimized to achieve shortest pulse duration after the filament. Largest spectral broadening, supporting a transform-limited (TL) 9.8-fs pulse was achieved by a clean, positively chirped input pulse (no. 3). The crucial input parameter for best compression is neither peak power nor initial pulse duration and slope, but seems to be best pulse contrast ratio (i.e. smallest prepulse-mainpulse peak ratio). After filamentation peak power is increased by more than 360% for the optimum pulse number 3.

Amplifier output						Filament output		
Pulse	T _{pulse} FWHM [fs]	Peak- power [GW]	GDD [fs ²]	Slope [fs ⁻¹)	Ratio Pre/Main pulse[%]	TL [fs]	T _{pulse} FWHM [fs]	Peak- power [GW]
0	33.2	21.5	-208	0.042	2.8	15.2	14.8	38.0
0	37.7	18.9	-96	0.024	2.7	12.4	13.0	52.5
€	41.5	18.2	424	0.027	2.1	9.8	11.2	66.4
0	49.1	15.2	669	0.030	3.1	11.3	12.4	61.5

Pulse number 3 generated the broadest filament spectrum. In contrast to others, this pulse exhibits the smallest pre-to-main pulse ratio (2.1%), which turned out to play the major role for optimum SPM during filamentation. The corresponding emerging filament pulse had a 9.8-fs theoretical limit and a measured duration of 11.2-fs. These measurements indicate that a *clean* temporal pulse shape without any pre-pulses seems to be best driver pulse for filamentation and for largest spectral broadening, even though the provided pulse intensity is not the highest possible in this case.



Fig. 4. Pressure scan in the first gas cell filled with Ar. The temporal pulse shapes (a) are reconstructed by SPIDER for a set of different gas pressures, ranging from 140 mbar up to 940 mbar. Pulse shortening is observed with a limit of 11.2 fs at 900 mbar. The measured pulse duration ((b), solid line) decreases almost linearly with higher pressure and are compressed close to the transform-limit (dashed line) by chirped mirrors which remained unchanged during the experiment. The large leap in pulse duration beyond 900 mbar is caused by beam profile break-up yielding significant laser beam distortion.

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Due to the slightly asymmetric spectrum of the amplifier pulse a moderate positive GDD $(+400 \text{ fs}^2)$ was necessary to provide the cleanest temporal pulse shape. For other spectral shapes, however, a driver pulse might carry no or even negative GDD to offer best pulse contrast ratio.

Next, we kept the input pulse parameters fixed for a 41.3-fs pulse and studied the process as function of gas pressure in both cells. The argon pressure was varied therefore from 100 mbar to 940 mbar and the results are shown in Fig. 4. The pulse duration after the first cell decreases from 34.3 fs to a minimum of 11.2 fs and then increases again, for constantly increasing gas pressure (Fig. 4(a)). This near linear decrease on the pulse full-width-at-half-maximum (FWHM) on increasing pressure is shown in Fig. 4(b) (dashed lines) and the corresponding transform limited (TL) pulse FWHM (solid line). Pulse shortening is significant already at the lowest gas pressure and then decreases with decreasing pulse duration. Optimum pulse shortening is observed at the maximum pressure of 900 mbar, beyond which the beam spatial profile is spoiled and makes pulse characterization unreliable.

The most astounding fact is the energy throughput of 800 μ J, which corresponds to 94% of the input pulse energy. Since our goal is to generate a pulse as clean and short as possible, we used the 11.2 fs pulse as input to the second cell and studied the dependence on the gas pressure as well (Fig. 5).



Fig. 5. Pressure dependence of output pulses from the second gas cell. a) The reconstructed temporal pulse profile after filamentary propagation in argon are plotted for various gas pressures. b) The Fourier-transfom limited (TL) pulse duration (dashed line) is decreasing almost linearly for higher pressures to a minimum of 4.1 fs at 750 mbar. The measured (solid line) pulse shows shortest duration (≈ 6 fs) at 650 mbar. The deviation from the TL pulse is attributed to imperfect GDD compensation by double-chirped mirrors.

Varying the gas pressure inside the second cell from 450 mbar to 750 mbar, we observe that the pulse spectrum nearly linearly increases, as indicated by the linearly decreasing TL width in Fig. 5(b). The increase in spectral width is however not as pronounced as in the first cell. The measured pulse FWHM does not linearly decrease with increasing pressure and follow the predicted TL FWHM simply because of limitations in the performance of the chirped mirrors. This is confirmed in Fig. 5(a) and indicated by the appearance of post-pulses in the temporal domain. Clearly, the currently limiting factors are the chirped mirrors, and improvements in their performance will result in shorter pulses.

We demonstrate this fact with a recent measurement of intense CEO-phase stabilized 5.1fs pulses by filamentation in an excellent spatial mode (Fig. 6) by using an improved set of chirped mirrors. Complete characterization of the pulses, in terms of duration, phase and associated spectra where performed employing a 1-kHz single-shot SPIDER setup.



Fig. 6. Pulse and associated spectrum and phase a)after the first filament cell (11 fs) b) after the second filament cell (5.1 fs)

A 0.68-mJ laser pulse from the amplifier system was focused into the first cell filled with argon at a pressure of 850 mbar leading to the formation of a short (< 10 cm long) and clearly visible filament. The input laser pulse was a 35-fs pulse with a slight positive chirp and the pressure in the first cell was optimized such as to generate the shortest and cleanest pulse as described above. After the first cell and subsequent chirped-mirror compression, pulses as short as 11-fs where obtained (Fig. 6(a)) with a substantially flat phase and with an energy of 0.65 mJ (conserving > 95% of the input energy). The 11-fs pulse was then refocused into a second cell filled with argon at a pressure of 900 mbar. An aperture was inserted before the entrance of the second cell in order to select only the center part of the incoming beam thereby reducing the energy to 0.56 mJ (~ 82% of the initial energy). Refocusing the 11-fs pulse into the second cell resulted, after recompression with the better chirped-mirrors, in a 5.1-fs pulse with energy of 0.18 mJ (Fig. 6(b)). This is 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 reveals that the generation is insensitive to gas variations within approximately 100 mbar.

3. Conclusions

Intense few-cycle laser pulses are generated though self-filamentation inside a noble gas atmosphere. We show that the energy dissipated inside the gas does not significantly alter the beam pointing stability and that the beam spatial quality is considerably improved though filamentation. The initial M^2 of 1.3 improves to a near perfect value of between 1.05 and 1.07, which is a direct consequence of filamentation generating its own guiding channel. Scans of the input pulse parameters and environmental conditions are carried out with the objective of identifying parameters, which result in the highest fidelity and shortest pulse duration: Scans of the pressures of both cells reveal a near-linear relation between pulse duration and pressure. Varying the chirp of the generating input pulse results in a modification of its temporal profile, which in turn influences the filamentation process. We have found that the chirp should be adjusted such as to result in a clean temporal pulse shape to yield the shortest output pulse. The potential of generating even shorter pulses than the

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initial 5.7-fs pulse [10] is demonstrated with improved chirped mirrors to 5.1 fs. The current limitation of this alignment-free method is the use of double-chirped mirrors with inadequate 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.

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