Pulse self-compression to the single-cycle limit by filamentation in a gas with a pressure gradient

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Received April 13, 2005; revised manuscript received June 2, 2005; accepted June 3, 2005

We calculate pulse self-compression of a 30 fs laser pulse traversing gas with different pressure gradients. We show that an appropriate density profile brings significant improvement to the self-compression by filamentation. Under an optimal pressure gradient, the pulse duration is reduced to the single optical cycle limit over a long distance, allowing easy extraction into an interaction chamber. © 2005 Optical Society of America

OCIS codes: 320.5520, 190.5530.

It was shown recently that filamentation of infrared femtosecond laser pulses in argon leads to a remarkable pulse self-compression.[1,2] The filamentation of ultrashort laser pulses is sustained by the combined action of beam self-focusing by the optical Kerr effect, beam defocusing that is due to the plasma created by multiphoton ionization, pulse self-steepening, and beam diffraction.[3] The propagating pulse experiences significant reshaping in both time and space.[4,5] This reshaping is so efficient that, by starting with a pulse of a few tens of femtoseconds' duration, one could obtain reproducible pulses with excellent beam quality and a duration as short as a few femtoseconds, close to the single optical cycle limit.[1] 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 submillijoule range comparable to that achieved with the hollow-fiber compression scheme.[6] Self-compression by filamentation suggests a simple design to produce even shorter pulses, in the attosecond domain, through the generation of high-order odd harmonics in a cell that acts both for pulse self-compression and harmonic generation. Recent experiments performed at ETH Zürich have shown interesting first steps in this direction.[7]

For many applications, however, a gas with uniform density is not practical. Optimal pulse shortening is reached after a specific distance, which depends on initial laser conditions and gas pressure. It is not easy to locate the distance of optimal compression and to extract the short pulse, even if one can adjust the optimal location to a certain extent by varying the gas pressure. The shortest structures are usually obtained after a plasma defocusing stage. The filamentation dynamics involves successive cycles with self-focusing and defocusing stages owing to the interplay between the optical Kerr effect and the plasma generated at each nonlinear focus. Between these points, the pulse broadens again because the trailing part of the pulse is refocused.[4] Recompression stages may occur at each successive cycle, after which the pulses reach an even shorter duration.[2,8] However, each recompression cycle is accompanied by diffraction and significant loss of energy owing to multiphoton absorption.

The purpose of this Letter is to describe a significant improvement of the self-compression scheme. Instead of producing a filament in a cell filled with a gas of uniform density, we investigate the case of a gas density gradient along the propagation axis. We show that this design gives even better pulse compression, avoids cyclic compression stages, and therefore limits the energy losses. The scheme permits easy extraction of the isolated single optical cycle pulse for delivery to an interaction chamber. To evaluate the pulse's temporal characteristics, we perform a numerical simulation with a three-dimensional nonlinear envelope propagation code that models the evolution of a laser pulse. This code has been tested in a large number of cases and faithfully reproduces the experimental results.[2,9–11] It is able to describe the evolution of a pulse even in the

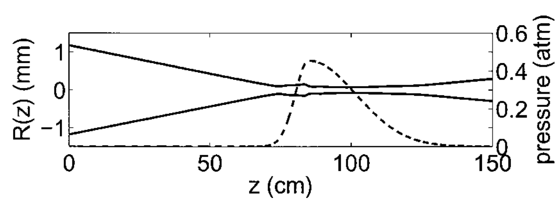

Fig. 1. Pressure distribution (dotted curve, scale on the right-hand axis) and computed beam width (continuous curve, scale at the left) along the propagation axis. The linear focus is at $z=80$ cm.

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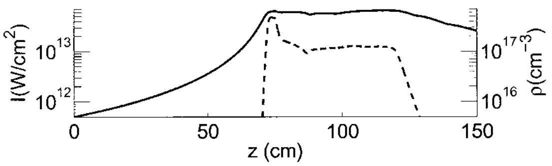

Fig. 2. Peak intensity (continuous curve, left-hand axis) and electron density (dashed curve, scale at the right) as functions of the propagation distance.

limit of a single cycle[12] and includes the effects of diffraction, the optical Kerr effect, self-phase modulation, self-steepening, and multiphoton ionization and absorption. We have concentrated on the nonlinear propagation of a femtosecond laser pulse at 800 nm for a large number of configurations of gas density gradients, with a practical design as a guideline. The parameters correspond to those of argon gas with potential $U_i = 15.76$ eV, nonlinear index $n_2 = 4.9 \times 10^{-19} p(\mathrm{cm}^2/\mathrm{W})$ (see Ref. 13), where p denotes the pressure in bars, and multiphoton ionization rate σ_{11}, in agreement with Keldysh's formulation. For simplicity we have always chosen the same incident laser characteristics, which correspond to commercially available kilohertz laser systems, and adopted a converging laser geometry. The incident laser pulse has a duration of 30 fs and an energy of 1 mJ. The beam, of waist $w_0 = 2$ mm, is focused with an $f = 80$ cm lens. The input pulse is assumed to have a Fourier limited bandwidth. Several particularly favorable cases emerge from computations performed with a large number of probed gradient configurations.

To illustrate these cases we consider the argon gas density shown by the dashed curve in Fig. 1. It has a steep front edge, followed by a smoother decreasing density gradient. The maximum gas density is at $z = 85$ cm. The computed width (FWHM) of the propagating beam is shown by solid curves in the same figure. The high intensity at the front edge leads to ionization and beam defocusing, immediately followed by a refocusing stage. Figure 2 shows the peak intensity and electron density produced in argon gas as a function of the propagation distance. Ionization, through plasma defocusing that saturates Kerr self-focusing, fixes the upper limit of the pulse intensity to a value of $I = 6 \times 10^{13}$ W/cm^2. The electron density reaches 4×10^{17} cm^{-3} at the focus, which produces a defocusing effect sufficient to slow the refocusing dynamics that occur at constant pressure filamentation.[4]

The efficiency of the pulse shortening can be seen from Fig. 3, which shows the computed pulse duration as a function of the propagation distance when the beam intensity is integrated over 100 μm. The shortening process is clearly visible for $80 \leqslant z \leqslant 120$ cm. The core of the beam (i.e., the partial power contained in a cylinder of radius $r < 100$ μm) reaches a single-cycle pulse duration during the descending part of the pressure gradient.

Figure 4 shows the temporal profile of the pulse intensity integrated over a radius of 100 μm, obtained at $z = 115$ cm. For comparison, the input pulse is shown by a dashed curve. The compression ratio in this case is ~ 12. The short pulse generated in the pressure gradient persists on further propagation into vacuum, where its peak intensity starts decreasing slightly because of diffraction ($z > 130$ cm). This single-cycle pulse can be used easily because it is long lived over a large distance, therefore avoiding any critical positioning of an extraction setup. As the pressure gradient along z ensures a smooth transition between gas and vacuum, an interaction chamber can be achieved simply by addition of a transverse gas jet at any location $z > 85$ cm.

Interestingly, integration of the pulse intensity over larger radii revealed that the partial power contained in cylinders of radii 300 or 500 μm also exhibits a decreasing duration for increasing distances. Even over 500 μm, the pulse duration is below 3 fs.

To obtain a better grasp of the self-compression process, it is instructive to examine the pulse shape at different distances in the cell. The dynamics is shown in Fig. 5. Because of the converging laser beam geometry, with the geometric focus set at the rising edge of the gas gradient, ionization occurs immediately, leading to superluminal propagation. The ionization front accelerates the pulse on axis, giving it a fishbone structure ($z = 80$ cm).[14] Part of the trailing edge of the pulse is refocused by the optical Kerr effect, the importance of which is enhanced by the increasing gas pressure. The slope of the pressure gradient, however, does not permit a complete refocusing cycle. The pressure decreases again before the trailing part of the pulse is refocused. The light, distributed in a V shape, is progressively diffracted during the decreasing edge of the pressure gradient ($85 < z < 120$ cm). Eventually, a clean, isolated pulse is formed, with a duration of 1.8 fs ($z = 130$ cm). It results from the erosion of the input pulse by the electron plasma created in its wake and by control of the diffraction process by the inhomogeneous pressure. In fact, the nonlinear effects are extinguished after the single-cycle structure is generated. By contrast, a constant gas pressure would lead to a multiple pulse structure.[2,4] The gradient of the plasma density along z therefore washes out the axial part of the light located in the trail of the input pulse. This reorganization from a fishbone to a pancake shape permits a slower decrease of the peak intensity than that which corresponds to Gaussian diffracting beams.

Although these results have been illustrated for a specific pressure gradient, they are quite generic in the sense that it is possible to obtain single-cycle pulses with similar features by varying the shape of the pressure gradient or the parameters of the input

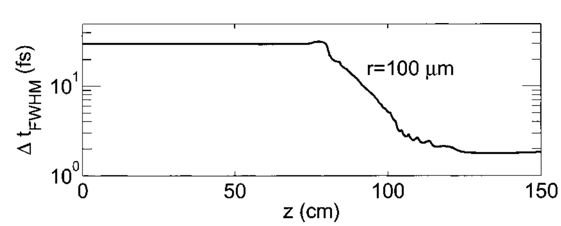

Fig. 3. Evolution of the pulse duration (FWHM) as a function of the propagation distance. The duration is computed after radial integration of the beam intensity over a beam radius of 100 μm.

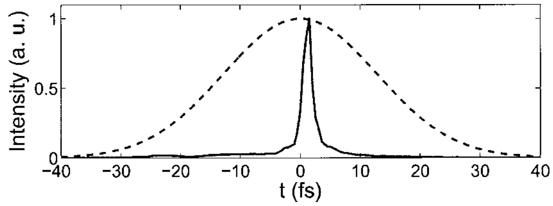

Fig. 4. Normalized intensity of the single-cycle pulse at $z=115$ cm. The dashed curve indicates the input pulse duration. The pulse has been averaged over a radius of 100 μm.

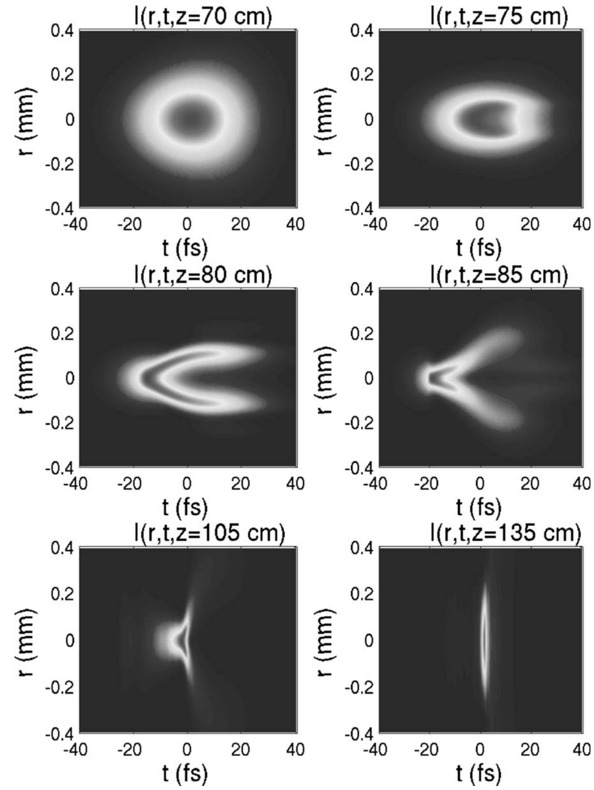

Fig. 5. Pulse dynamics during propagation through a pressure gradient.

pulse. Sending the same pulse (1 mJ, 30 fs, focus at 80 cm) on a parabolic (triangular) pressure gradient with a maximum $p=1$ ($p=0.5$) bar at 85 cm and a length of 24 cm results in a short pulse of ~2 fs that can be extracted anywhere from $z=100$ to $z=150$ cm. If the triangular pressure gradient is used and the input duration and energy are decreased to 60 fs and 2 mJ, respectively, a pulse of less than 3 fs in duration is formed at the end of the filament with a good compression ratio of 23. If the same triangular pressure gradient is used and the input pulse duration and energy are increased to 60 fs and 2 mJ, the simulation shows that a pulse of less than 3 fs in duration is formed at the end of the filament with a good compression ratio of 23. This suggests that these pressure gradients could be achieved by an argon gas flow entering an open tube from the middle, with differential pumping along the tube.

For applications, a crucial issue is the available energy in the short pulse. The simulation shows that the energy contained within a cylinder of radius of 100 μm is 60 μJ at $z=115$ cm.[15] Approximately 10% of this energy is contained within the short pulse of ~3 fs. Another important characteristic is the intensity contrast, especially at negative times. Our calculations show that the contrast is better than 10^{-2} at $t=\pm 20$ fs and better than 10^{-5} at $t=\pm 50$ fs. Calculations with a parabolic gradient show a deterioration of the contrast with 5×10^{-2} at $t=\pm 100$ fs.

In conclusion, we have described an efficient self-compression scheme by filamentation based on the presence of suitable pressure gradients. The process yields single-cycle optical pulses. Enhancement of the energy in the single-cycle pulse could be achieved by coupling of this self-compression process with the organization of multiple filaments recently shown in Ref. 9. Input pulses with larger energy lead to multiple filaments that eventually coalesce into a single filament, provided that controlled intensity gradients are introduced in the input beam. Another approach could be scaling to lower peak gas pressures, which should increase the size of the filament and its energy. The corresponding numerical treatment would require inclusion of multiple ionization.

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