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Scalable sub-cycle pulse generation by soliton self-compression in hollow capillary fibers with a decreasing pressure gradient

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Abstract. Advances in the generation of the shortest optical laser pulses down to the sub-cycle regime promise to break new ground in ultrafast science. In this work, we theoretically demonstrate the potential scaling capabilities of soliton self-compression in hollow capillary fibers with a decreasing pressure gradient to generate near-infrared sub-cycle pulses in very different dispersion and nonlinearity landscapes. Independently of input pulse, gas and fiber choices, we present a simple and general route to find the optimal self-compression parameters which result in high-quality pulses. The use of a decreasing pressure gradient naturally favors the self-compression process, resulting in shorter and cleaner sub-cycle pulses, and an improvement in the robustness of the setup when compared to the traditional constant pressure approach.

Keywords: Ultrafast nonlinear optics, Hollow capillary fibers, Soliton-self compression, Sub-cycle pulses.

In a continuous effort to access the briefest and most fundamental phenomena in nature, intense ultrashort laser pulses have become indispensable tools for ultrafast science, breaking new ground in time-resolved spectroscopy and strong-field physics [1, 2]. At present, few-cycle femtosecond pulses in the optical spectral region are routinely generated by nonlinear post-compression in gas-filled hollow capillary fibers (HCFs) [3], which stand out among other compression schemes for their simplicity, high-damage threshold and the possibility of tuning their nonlinearity and dispersion by modifying the filling gas or its pressure. However, although HCF post-compression experiments have been greatly optimized [4], they are currently reaching their limit in terms of the shortest achievable pulse duration due to the complexity of dealing with uncompensated high-order dispersion in octave-spanning spectra. Overcoming this problem, parametric light-field synthesizers have succeeded in generating the shortest optical laser pulses well down into the sub-cycle regime, offering new opportunities for advancing real-time observation and precision control of electron dynamics at the atomic scale [5, 6]. As a promising alternative to these extremely complex systems, high-energy soliton dynamics in HCFs is attracting a great interest as a direct route to extreme pulse self-compression down to the sub-cycle regime [7, 8]. As opposed to more conventional post-compression techniques, soliton self-compression relies on the simultaneous nonlinear spectral broadening and phase compensation arising from the interplay between the negative group-velocity dispersion (GVD) of the waveguide and self-phase modulation (SPM). Still, for a practical implementation, the complexity of this nonlinear interaction calls for theoretical investigations on scaling rules and design guidelines to identify the optimal experimental parameters which result in high-quality self-compression [9]. So far, extreme soliton self-compression in HCFs in the near-infrared (NIR) has only been demonstrated for pre-compressed (~10 fs) pump pulses [7, 10], or otherwise in configurations which ensure a strong anomalous response, i.e., working with longer wavelengths [8, 11], high-order modes [12], or small-core photonic crystal fibers [9, 13]. However, the unexpected applicability of sub-cycle self-compression to standard experimental setups driven by NIR multi-cycle pulses propagating in the fundamental mode of large-core HCFs has been recently demonstrated in negatively pumped fibers, i.e., HFCs filled with a decreasing pressure gradient [14]. Pressure gradients are routinely implemented by sealing the fiber into a gas cell at each end, which can be independently evacuated and filled with gas, yielding a longitudinal pressure distribution with a square root-type profile when the gas flows from highest to lowest pressure [10].

In this communication we further investigate the scalability of soliton self-compression down to the sub-cycle regime in HCFs filled with a decreasing pressure gradient. Varying the filling gas and choosing between atomic (Ne) and molecular (N_2) species, we study the compression

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process in two completely different dispersion and nonlinearity landscapes, which are of critical importance in soliton dynamics. Our results demonstrate that nearly identical self-compression performance can be achieved in very distinct HCF scenarios, and provide a surprisingly simple universal route to find the optimal parameters for generating high-quality NIR sub-cycle pulses.

Our work is based on one-dimensional numerical simulations of nonlinear pulse propagation [15, 16], including the complete linear response of the gas-filled HCF [17], SPM, stimulated Raman scattering modeled in a damped harmonic oscillator approximation [18], and self-steepening. This theorical model accurately describes ultrashort pulse propagation down to the single-cycle limit in a regime of moderate intensities [19].

In order to identify a route towards high-quality self-compression, we have followed the procedure detailed in [14]. In brief, we have systematically simulated the propagation of a transform-limited 30 fs gaussian pump pulse at 800 nm in the fundamental mode of a 3 m long, 100 μm core radius HCF, filled with either Ne or N_2, and with both constant gas pressure and a longitudinal decreasing pressure gradient ending in vacuum. The latter two situations are fairly compared by matching the integrated nonlinear phase shift acquired by the pulse peak during its propagation, which is often referred to as B-integral. Neglecting the fiber losses, a system with a decreasing pressure gradient from p_0 to vacuum can then be compared to that with a constant pressure p_{eq} simply if $p_0 = (3p_{\mathrm{eq}})/2$ [10, 12]. The main difference lies in that non-uniform pressure allows for a dynamic tuning of the dispersion and nonlinearity experienced by the pulse during its self-compression, as the propagation constant and the nonlinear parameter scale linearly with the gas density and, thus, with pressure.

For the parameters considered in our study, Figure 1 shows the GVD and instantaneous nonlinear coefficient (related only to instantaneous Kerr effect or SPM [15]) of a HCF filled with Ne or N_2 at different pressures. As we can see, a Ne-filled HCF has a weaker instantaneous nonlinearity and displays anomalous dispersion (GVD < 0) over a larger pressure range than an identical fiber filled with N_2. An inner fiber radius of 100 μm was chosen because it offers a good balance between acceptable losses at 800 nm and a sufficiently strong anomalous response in N_2. Furthermore, a pulse propagating in N_2 might experience a delayed molecular contribution to the optical Kerr effect which vanishes in noble gases like Ne. Therefore, owing to their very distinct linear and nonlinear nature, the optimal self-compression in either Ne or N_2 is expected to occur for different input pulse and fiber parameters.

Following [14], we have simulated the soliton self-compression of the aforementioned 30 fs pulse while varying its initial energy and the equivalent gas pressure in the HCF. For each energy-pressure pair in the resulting bidimensional parameter space, we have plotted the intensity full width at half-maximum (FWHM) duration and the ratio of output to input peak power of the self-compressed pulses, as shown in Figure 2. In these plots, the optimal region for high-quality self-compression can be readily identified as the intersection between the areas of shortest output pulse duration and largest peak power enhancement. Surprisingly, the results for Ne and N_2 show an identical behavior, which also follows that previously reported for Ar [14], except for the fact that they are displaced to different input energy and gas pressure ranges as mentioned earlier. In both cases, it is clear that, in the whole parameter ranges considered here, the self-compression process is substantially enhanced when the fiber is negatively pumped rather than statically filled, resulting in the generation of self-compressed pulses with extremely short durations well down into the sub-cycle regime (~1 fs) and high peak powers, which in turn implies a clean temporal profile. The most outstanding feature is that there is not just a single pair of input energy and gas pressure values that allow for a high-quality compression, but there is a whole parameter region which yields similar results. This optimal region is found to always appear towards the same corner of the contour line in the energy-pressure map where the fixed fiber length (L) matches an average compression length (L_{av}), that we defined as:

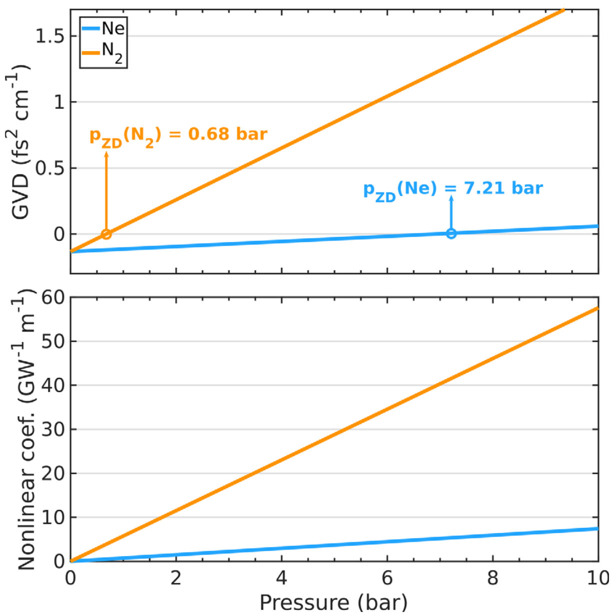

Fig. 1. GVD (top) and instantaneous nonlinear coefficient (bottom) at 800 nm of the fundamental mode of a 100 μm core radius HCF filled with Ne or N_2 as a function of gas pressure. Labels indicate the zero-dispersion pressure (p_{ZD}) in each case.

$$L_{\mathrm{av}} = \frac{L_{\mathrm{sc}} + L_{\mathrm{fiss}}}{2}, \tag{1}$$

where L_{sc} and L_{fiss} represent, respectively, the characteristic self-compression and soliton fission lengths, which are given by [20]:

$$L_{\mathrm{fiss}} = \frac{L_D}{N}; \quad L_{\mathrm{sc}} = \frac{L_{\mathrm{fiss}}}{\sqrt{2}}. \tag{2}$$

$N = (L_D/L_{\mathrm{NL}})^{1/2}$ being the soliton order, and $L_D = T_p^2/(4\ln 2|\beta_2|)$ the dispersion and $L_{\mathrm{NL}} = 1/(\gamma_i P_0) = \sqrt{\pi/(4\ln 2)}\,T_p/(\gamma_i E_0)$ the nonlinear lengths, which describe

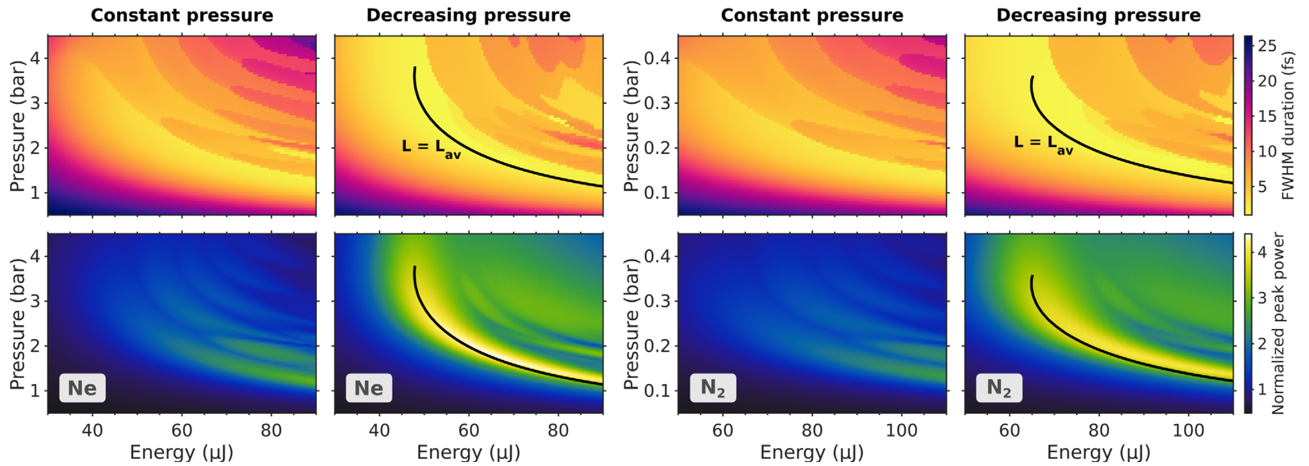

Fig. 2. FWHM duration (top row) and ratio of output to input peak power (bottom row) of the self-compressed pulses as a function of the input energy and the equivalent constant pressure (see text) in both a statically filled or a negatively pumped 3 m long, 100 μm core radius HCF filled with Ne (left) or N_2 (right). The solid black lines represent the contour lines where $L = L_{\rm av}$, which run along the optimal region for self-compression in a decreasing pressure gradient. Note the one order of magnitude change in the pressure range from Ne to N_2 owing to their different dispersion and nonlinearity.

the characteristic length scales of GVD and SPM, respectively. Here T_p represents the intensity FWHM duration of the gaussian pump pulse, P_0 is its input peak power and E_0 its initial energy, β_2 is the GVD coefficient of the HCF, and γ_i is the instantaneous nonlinear parameter as defined elsewhere [15]. The constraint $L = L_{\rm av}$ ensures that $L_{\rm sc} < L < L_{\rm fiss}$ and, therefore, guarantees that the self-compressing pulse reaches its maximum compression without entering in the soliton fission regime. In addition, the soliton order should be kept $N < 15$ to achieve a high-quality compression [13], inevitably setting an upper limit to the achievable pulse energy. Independently of input pulse, gas and fiber parameters, the condition $L = L_{\rm av}$ always describes a contour line in the energy-pressure plane which, when falling inside the space with $N < 15$, can be used to identify the optimal region for high-quality self-compression in a universal way. A detailed inspection of the conditions $L = L_{\rm av}$ and $N < 15$ suggests that upscaling our results towards millijoule pump pulses should become possible, even in practical short fibers (~1 m), by pushing the central wavelength into the mid-infrared spectral region.

As an example of the high-quality sub-cycle waveforms that can be generated from the negatively pumped fiber, in Figure 3 we have plotted the self-compressed pulses obtained for two different pairs of input pulse energy and equivalent gas pressure which lie towards the same area of the optimal self-compression regions in Figure 2, corresponding to Ne and N_2, respectively. When compared to the output pulses in the equivalent constant pressure situations, it is clear that those generated with a decreasing gradient are much better, displaying shorter durations, higher peak powers, a cleaner temporal profile with a higher contrast, and a broader spectrum spanning from the NIR to the mid-ultraviolet. The self-compressed pulses from the negatively pumped HCF reach sub-cycle FWHM durations of 1.1 and 1.2 fs, and output peak powers of 8.8 and

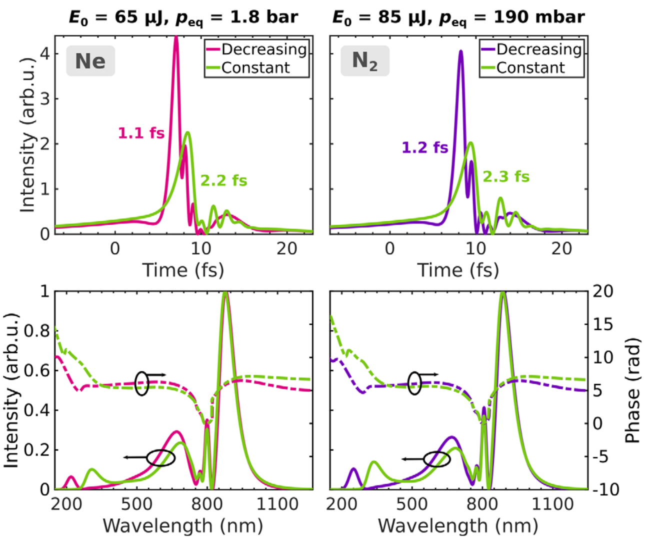

Fig. 3. Temporal intensity profile (top row) and spectrum (bottom row) of the self-compressed sub-cycle pulses obtained after propagation through a HCF filled with Ne (left) or N_2 (right), at both constant or decreasing pressure, for two different pairs of input pulse energy and equivalent gas pressure which lie towards the same area of the optimal self-compression regions in Figure 2.

10.7 GW in Ne and N_2, respectively. However, in the equivalent constant pressure situations, the output pulses where only 2.2 and 2.3 fs in duration, and 4.6 and 5.4 GW in peak power. The improvement with the decreasing pressure gradient has been attributed to an effective suppression of higher-order dispersion and self-steepening in the last stages of the pulse compression, together with a continuous blueshift of the zero-dispersion frequency at the same time as the pulse spectrum broadens by SPM [12, 14]. When propagating in the anomalous dispersion regime, it is straightforward to understand from the trends shown in Figure 1 that a decreasing pressure gradient is the most natural way to emphasize and favor the characteristic dynamics of the self-compression process [11]. In short, at the fiber entrance the higher pressure enhances the accumulation of nonlinear phase shift and the spectral broadening of the input pulse by SPM. In later stages, the larger spectral extent combined with an increase in the magnitude of the anomalous GVD and a reduction of third-order dispersion due to the drop in pressure, assist the phase compensation for pulse self-compression and delay the fission process beyond the maximum compression point that can be reached with constant pressure. Altogether, the decreasing pressure gradient enables unprecedent compression ratios ($\gtrsim 25$) which had remained out of reach due to detrimental high-order effects [13]. Furthermore, the great similarities between the pulses in Figure 3, generated with different gases, energies and pressures, demonstrates the promising scaling capabilities of HCF self-compression down to the sub-cycle regime in different configurations. Another interesting point is that the optimal self-compressed pulse is accompanied by the onset of resonant dispersive wave (RDW) emission, when the strongly nonlinear self-compressing soliton transfers its excess energy to a linear wave propagating in the normal dispersion regime [7, 8, 10, 16]. This is manifested by the isolated peak around 200 nm in the output spectra of the lower panels in Figure 3. At this point, RDW emission has just started and the energy transfer to the ultraviolet is still low, resulting in conversion efficiencies quite below saturation. This fact could be used to experimentally predict the best sub-cycle pulse parameters based on RDW spectral content at the fiber output.

In summary, we have demonstrated that broadly similar high-quality NIR sub-cycle pulses can be generated by extreme soliton self-compression in negatively pumped HCFs in different configurations. Independently of input pulse, gas and fiber choices, the optimal self-compression parameters can always be found by matching the fiber length to and average compression length, providing a simple design guideline for experiments. Furthermore, the decreasing pressure gradient can help to improve the robustness of HCF self-compression and the quality of the generated sub-cycle pulses when compared to the equivalent constant pressure situations, also preventing the onset of undesirable high-order effects. We believe that these findings will pave the way towards a new generation of ultrafast experiments which might benefit from the availably of tailored sub-cycle waveforms, especially those which are carried out in vacuum chambers, like the synthesis of high-frequency isolated attosecond pulses through high-order harmonic generation.

Conflict of interest

The authors declare no conflict of interest.

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