Cascaded self-compression of femtosecond pulses in filaments

Carsten Brée
1,2,6, Jens Bethge2, Stefan Skupin3,4, Luc Bergé5, Ayhan Demircan1 and Günter Steinmeyer2

1 Weierstraß-Institut für Angewandte Analysis und Stochastik, 10117 Berlin, Germany
2 Max-Born-Institut für Nichtlineare Optik und Kurzzeitspektroskopie, 12489 Berlin, Germany
3 Max-Planck-Institut für Physik komplexer Systeme, 01187 Dresden, Germany
4 Friedrich Schiller University, Institute of Condensed Matter Theory and Optics, 07743 Jena, Germany
5 CEA-DAM, DIF, 91297 Arpajon, France

E-mail: bree@wias-berlin.de

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Abstract. Highly nonlinear wave propagation scenarios hold the potential to serve for energy concentration or pulse duration reduction of the input wave form, provided that a small range of input parameters is maintained. Exploitation of this mechanism for pulse compression is ultimately limited by parameter fluctuations of the input wave. With high compression ratios, it becomes increasingly difficult to maintain control of the waveforms. Here, we suggest an alternative approach to the control of waveforms in a highly nonlinear system. Cascading pulse self-compression cycles at reduced nonlinearity limit the increase of input parameter sensitivity while still enabling an enhanced compression effect. This cascaded method is illustrated by experiments and by numerical simulations of the nonlinear Schrödinger equation, simulating the propagation of short optical pulses in a self-generated plasma.

6 Author to whom any correspondence should be addressed.
The occurrence of modulational instabilities or similar temporal pulse break-up scenarios is a characteristic feature of the nonlinear propagation of waves. Two prototypical examples of such events are the Benjamin–Feir instability [1] of deep-water waves and the azimuthal modulational instability of spatial solitons of the nonlinear Schrödinger equation in optics [2]. Similar phenomena have been reported to occur in Bose–Einstein condensates [3], in plasma physics [4, 5] and in the propagation of short laser pulses [6–9]. In self-generated optical filaments, temporal break-ups serve to actively compress femtosecond laser pulses [10, 11]. Recently, there has been revived interest in such phenomena as they can give rise to an unusual increase of pulse amplitude or concentration of energy and to the appearance of the so-called rogue waves [12, 13]. The probability for the appearance of these rare events rapidly decreases with their amplitude. As the physical systems are deterministic, perfect control of the input wave should, in principle, enable an arbitrary increase of wave amplitude within the system’s limitations. However, exploitation of rogue wave phenomena [14] or other highly nonlinear scenarios for the generation of a desired pulse shape is technically limited by the feasibility of control over the input wave. Noise on the input waveform therefore impedes pulse compression at a certain point. In most systems, a fundamental limitation arises due to quantum noise [8].

In the following, we present a new approach for exploiting rare events in a highly nonlinear system, cascading the process while at the same time limiting the underlying nonlinearity in every step. The latter measure maintains control over the output wave when exploiting such events, e.g., for waveform compression. We illustrate this cascaded waveform control for the propagation of short pulses in a self-generated filament that are suitably described by the nonlinear Schrödinger equation [10, 11]. In this system, pulse compression factors of the order of 3–5 have been previously discussed [15–19] in single-compression cycles. Quite remarkably, the compression factor of each individual process remains nearly conserved in double self-compression, enabling in total nearly twelfold compression.

For the investigation of the double self-compression mechanism, we perform numerical simulations of the generalized nonlinear Schrödinger equation that couples the envelope $\mathcal{E}$ of the electric field to the plasma density $\rho$ of the medium [18] according to

$$\frac{\partial}{\partial t} \mathcal{E} = \frac{i}{2k_0} T^{-1} \Delta_\perp \mathcal{E} + i D \mathcal{E} + \frac{\omega_0}{c} n_2 T |\mathcal{E}|^2 \mathcal{E} - \frac{k_0}{2\rho_c} T^{-1} \rho \mathcal{E} - \frac{\sigma}{2} \rho \mathcal{E} - \frac{U_i W(I)(\rho_{at} - \rho)}{2I} \mathcal{E},$$

(1)

$$\frac{\partial}{\partial t} \rho = W(I)(\rho_{at} - \rho) + \frac{\sigma}{U_i} \rho I.$$  

(2)

Here, $I = |\mathcal{E}|^2$ is the cycle-averaged field intensity. Assuming cylindrical symmetry, the transverse Laplacian may be reduced to $\Delta_\perp = (1/r) \partial_r r \partial_r$, where $r = (x^2 + y^2)^{1/2}$ in cylindrical coordinates. Space–time focusing and self-steepening are introduced by the operator $T = 1 + (i/\omega_0) \partial$, the operator $T^{-1}$ being evaluated in the Fourier domain. Correspondingly, the operator $\tilde{D}$ modeling dispersion in argon is treated in the Fourier domain according to

$$\tilde{D}(\omega) = k(\omega) - k_0 - (\omega - \omega_0) \frac{\partial}{\partial \omega} |_{\omega = \omega_0},$$

(3)

where $k(\omega) = n(\omega) \omega / c$ is the wavenumber at the angular frequency $\omega$, and $n(\omega)$ is the frequency-dependent refractive index in argon according to Dalgarno and Kingston [20]. Note that argon is normally dispersive for all relevant wavelengths and that dispersive shaping plays only a minor role in filamentation. The carrier frequency of the laser field and corresponding wavenumber are denoted by $\omega_0$ and $k_0$, respectively, with a carrier wavelength $\lambda = 800 \text{nm}$. 

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Figure 1. (a) Evolution of on-axis intensity profile along $z$ for pulse self-compression into a sub-diffractive channel in argon, $p = 106$ kPa. (b) Corresponding evolution for the double self-compression scenario at $p = 109$ kPa. Inset: close-up on the pulse break-up in the second focus, accompanied by shock wave formation. (c) Evolution of the on-axis peak intensity for pressure from 106 to 120 kPa.

$n_2$ is the nonlinear refractive index [21] and $U_i$ corresponds to the ionization potential of the medium. Moreover, $\rho_c = 1.73 \times 10^{21}$ cm$^{-3}$ is the critical plasma density at $\omega_0$, and $\rho_{\text{nt}}$ denotes the neutral density of the medium. $\sigma$ is the cross section for collisional ionization. The ionization rate $W(I)$ is modeled according to Perelomov–Popov–Terent’ev (PPT) [22] and adequately describes both multiphoton and tunneling ionization processes.

Based on the experimental parameters discussed below, we assume 2.5 mJ optical input pulses at 800 nm and with initial beam waist $w_0 = 2.5$ mm and pulse duration $t_{\text{FWHM}} = 120$ fs, being focused by an $f = 1.5$ m lens into a noble gas. To identify the small parameter range giving rise to compressed output waveforms on the axis of the filament, a parameter scan is performed by varying the gas pressure in a range from 100 to 120 kPa, at otherwise fixed input parameters, see figure 1. At a pressure $p = 106$ kPa, our simulations predict plasma-dominated dynamics in a relatively short nonlinear focal zone succeeded by a 1 m long self-generated channel, in which plasma formation is virtually absent. In the latter zone, Kerr self-focusing effectively balances linear diffraction, see figure 1(a). This figure clearly reveals how a splitting event at $z = 1.4$ m close to the linear focus position merges into formation of one isolated and shorter pulse. The splitting initially produces two pulses, one at $t = -100$ fs and a second one at $t = +60$ fs. At $z = 1.6$ m, each of these subpulses is roughly 40 fs wide, which is a natural consequence of the split. Upon further propagation ($z = 1.7$ m), the pulse at negative delays...

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dies out quickly, leaving only one isolated and shortened pulse behind. This prototypical split–isolation cycle has already been discussed in \[18, 19, 23\] as the origin of on-axis pulse self-compression \[17\]. After the split–isolation cycle, plasma generation has effectively ceased, such that pulse shaping in the elongated channel at \(z > 1.7\) m is now dominated by an interplay between Kerr-type self-refraction and linear optical effects. Notably, self-focusing compensates for diffractive optical effects, giving rise to a sub-diffractive nature of this final nonlinear propagation stage, as also observed by Faccio \textit{et al.} \[24\]. In comparison to the following example, it is important to understand that the pulse conserves a single-maximum shape after isolation of the trailing pulse. There are faint indications of satellite formation for \(z > 3\) m, which, however, do not evolve to a split–isolation cycle yet.

Increasing the pressure to 109 kPa, we disturb the delicate balance in the sub-diffractive channel behind the strongly ionized zone by a slight increase of Kerr nonlinearity. This increase triggers a refocusing event 0.5 m behind the first nonlinear focus, and a second strongly ionized zone evolves (figures 1(b) and (c)). Here, the pulse experiences a second split–isolation cycle that shows superficially the same behavior as the first one, i.e. the surviving pulse from the first cycle splits into two at \(z = 2.2\) m. In contrast to the first cycle, the trailing pulse dies out at \(t \approx 80\) fs, leaving only one isolated and yet again shortened pulse at \(t \approx 50\) fs behind. In the subsequent nonlinear propagation inside the channel, the pulse reaches a minimum duration of 16.4 fs at \(z = 2.5\) m. Further increasing the pressure to \(p = 120\) kPa, pulses with a minimum duration of 10.9 fs emerge after the second focus. This nearly twelvefold compression chiefly goes back to the two split–isolation cycles. Such a strong compression effect has not been observed in previous experimental \[15\]–\[17\] or theoretical studies \[18, 19\]. The emergence of the refocusing event must not be confused with focusing–defocusing cycles \[25\] that occur on significantly shorter length scales of \(\approx 20\) cm, whereas repetition of the split–isolation cycle is only observed with a distance of \(> 50\) cm between the events. Apart from the different length scales, our simulation indicates a pronounced intensity drop and a resulting cessation of plasma formation between the two cycles (figure 1(c)), which further suggests a conceptual difference to the much milder focusing–defocusing cycles previously reported.

Despite the apparently identical effect on pulse duration, collapse saturation in the two foci is accomplished by different physical effects. In the first nonlinear focus \((z = 1.5\) m), plasma defocusing and related dissipative terms clamp the intensity, whereas temporal effects, in particular dispersion, take over this role in the second focus \((z = 2\) m). To verify this issue, we repeated simulations with neglect of the plasma response for \(z > 1.75\) m. Otherwise, all parameters have been duplicated from the simulation at \(p = 107\) kPa (figure 1(c), blue line). Switching off plasma effects in the second stage of propagation leads to a nearly unchanged dynamical behavior for the second compression stage. Similar plasmaless refocusing events have been discussed in \[26, 27\]. With increasing pressure \((p \geq 1.09\) kPa), however, plasma again becomes essential for preventing spatial wave collapse, while dispersion dominates temporal dynamics by exchanging power between different pulse time slices. The generation of dispersive shock waves in the trailing edge of the pulse (figure 1(b), inset) during the second splitting event further underlines the strong impact of dispersion and self-steepening.

In order to further analyze spectro-temporal signatures of the pulse-shaping action during the refocusing event, we computed cross-correlation frequency-resolved optical gating (XFROG) spectrograms from the simulated on-axis data, see figure 2. XFROG spectrograms are a convenient way to analyze characteristic deviations from a spectrally and temporally homogeneous energy distribution inside the pulse, which are also directly measurable \[32\].

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Figure 2. On-axis XFROG spectrograms (a) of the optical field emerging from the sub-diffractive channel regime of figure 1(a), \( z = 2.5 \) m and \( p = 106 \) kPa. (b) The same during the second pulse breaking at \( p = 109 \) kPa (two-foci regime). Note that the second pulse isolation has not occurred yet, i.e. pulse durations are still nearly identical. (c) Energy of the output pulse at \( z = 2.5 \) m transmitted through an aperture of radius \( r_0 \). (d) Temporal duration (FWHM) of transmitted power profiles.

In highly nonlinear scenarios, these spectrograms have previously elucidated the mechanisms behind supercontinuum generation in photonic crystal fibers [33] and filaments [18]. For the single self-compression regime, the XFROG spectrograms exhibit a characteristic shape that is most suitably described as the mirror image of the Greek letter \( \Gamma \) (figure 2(a)), as has already been discussed in [18]. A short pulse duration is intimately connected to a vanishing slant of the vertical bar of the \( \Gamma \). The extension of this section towards the blue spectral range is a measure of the asymmetric nonlinear spectral broadening effects, mainly caused by self-steepening [28]. The appearance of pronounced horizontal structures along the cap section of the \( \Gamma \), in contrast, is connected with the suppression of the leading pulse in the split–isolation cycle, i.e. pulse contrast. If a second split–isolation cycle appears, the spectrotemporal pattern of the pulses changes in a characteristic way, see figure 2(b). Remnants of the suppressed pulse after the second split–isolation cycle now appear as a blue trailing pedestal of the spectrogram, i.e. the point symmetric to the red leading pedestal appearing after the first cycle, with a shape that we will refer to as Q-shape in the following. The broadening effect appears as a spectral red-shift along the vertical axis in figure 2(b). As filamentary self-compression is typically restricted to a small region around the optical axis, for the simulated output pulses at \( z = 2.5 \) m we calculated the power profile transmitted through an aperture of radius \( r_0 \) defined by \( P_0(t) = 2\pi \int_0^{r_0} I(t, r) dr \), where \( I(t, r) \) defines the spatiotemporal intensity distribution of
Figure 3. (a) Evolution of the XFROG spectogram along $z$ during the propagation through the second focus. (b) The corresponding angularly resolved far-field spectra.

the laser field. Plots of the transmitted energy and averaged pulse duration versus the aperture radius $r_0$ are shown in figures 2(c) and (d), respectively. This clearly shows that, for the chosen initial conditions, double self-compression is superior to the single-focus scenario for two reasons. Firstly, the pulse duration (green curve) increases less rapidly with $r_0$ for the double-compression scenario at 120 kPa. Secondly, only for this scenario, energetic 0.3 mJ output pulses at sub-20 fs duration can be obtained.

Figure 3(a) shows a more detailed view of the transition from inverse $\Gamma$ to Q shape, with a zoomed-in set of spectrograms computed in the range from $z = 195$ to 245 cm. In the initial spectrograms in this sequence, the typical $\Gamma$ shape appears with the vertical bar extending into the blue spectral range. During the approach towards the second focus, however, the spectral extension into the blue decreases, with a red-shift appearing shortly afterwards. The red-shift emerges with the formation of a blue trailing pedestal, which is ultimately a remnant of the blue wing of the inverse $\Gamma$ shape. Figure 3(b) additionally shows a view of the angularly resolved spectra [24] during this transition phase. These structures exhibit a markedly different behavior in the blue and the red wing of the pulse. This behavior is related to the strong influence of self-steepening, which causes the blue-shift in the trailing part of the pulse. In fact, the modulational instability occurring in self-focusing media with normal group-velocity dispersion reshapes this part of the pulse into a characteristic X-shaped spatio-spectral pattern [29]. In the spatio-temporal domain, this instability has also been shown to be responsible for the observed temporal splitting and the emergence of hyperbolic shock waves [30, 31]. Remarkably, those dispersion-dominated dynamics are still observable in the pressure regime above 109 kPa, where plasma defocusing is already essential for wave-collapse arrest. The apparent red-shift of the spectra of the optical fields emerging from the two-foci regime can thus be chiefly ascribed to self-phase modulation in the leading edge of the pulse during the refocusing stage.
Figure 4. (a) XFROG trace of an output pulse after double self-compression in air, obtained from the measured SPIDER data. (b) Numerically obtained Q-shaped XFROG trace at the exit of the air filament after double self-compression, \( z = 3 \text{ m} \).

For experimental verification of the double self-compression, we employed a 45 fs regenerative Ti:sapphire amplifier system with a pulse energy of 5 mJ. The laser pulse energy has been carefully attenuated by means of an adjustable diaphragm and focused with an \( f = 1.5 \text{ m} \) lens to generate a single filament in air. A second diaphragm after the filament served to isolate the core of the filament. After suitable attenuation, the temporal structure in the filament core was analyzed with spectral phase interferometry for direct electric-field reconstruction (SPIDER [34, 35]). The SPIDER method delivers the spectral phase, which can be combined with an independently measured spectrum to reconstruct the complex-valued field envelope in the spectral or temporal domain. Moreover, this information also suffices to directly reconstruct XFROG spectrograms from experimental data.

Except for the fact that no gas cell was necessary, this setup widely resembles the one in [17]. Adjusting the input diaphragm, a regime could be found that displayed a single filament with two clearly separated strongly ionized zones that were separated by about 30–40 cm. With these short input pulses, our simulations indicate that we can at best expect about threefold compression. It may appear intriguing to suggest dispersive stretching of the 45 fs pulses to 120 fs duration in order to demonstrate the full compression potential. Yet, these chirped pulses would already exhibit a much wider bandwidth than Fourier-limited 120 fs pulses, and compression could also stem from linear optical effects. We therefore directly used the short 45 fs pulses delivered by our laser source as the input.

From the measured SPIDER data, the XFROG spectrogram shown in figure 4(a) and the pulse shape in figure 5(c) were reconstructed. The spectrogram shows features previously discussed for the single-focus and the double-focus regime in argon, cf figures 2(a) and (b), respectively. From the former, a temporally stretched leading pedestal is discernible, which is generally quite typical for self-compression [18]. In addition to previous experimental findings, however, a clearly visible trailing blue pedestal appears. A Q-shaped spectrogram thus forms, which characterizes a second split–isolation cycle. We repeated simulation runs with identical parameters as in the experiments, see figures 4(b) and 5(c). These simulations indicate nearly identical pulse shape and duration on a linear scale. Agreement in the pedestal structures on a logarithmic scale is certainly less ideal. Yet, the simulation shows similar indications for the leading red and trailing blue pedestals as in the experiments, i.e. there is qualitative
Figure 5. (a) Evolution of on-axis temporal intensity along $z$ in a numerically simulated filament in air, exhibiting a refocusing stage and double splitting events. (b) On-axis spectra numerical simulation and experiment. (c) On-axis temporal intensity profile from SPIDER measurement (blue curve) versus on-axis profile at $z = 3.5$ m obtained from numerical simulations (black curve).

agreement in the pedestal structure. To the best of our knowledge, such a structure has not been reported in the literature yet. It is striking that this feature appears temporally less stretched than the leading red pedestal from the first split–isolation cycle, which corroborates less exposure to linear and nonlinear pulse-shaping effects. Therefore, the experimental findings appear to be highly compatible with the causal sequence of events predicted by numerical simulation. This finding also suggests that the second split–isolation cycle is caused by a different mechanism than the first one, causing pedestal formation at opposing spectral and temporal edges of the main pulse. To compare our experimental findings with theoretical predictions, we included a delayed Kerr nonlinearity in the model equations for pulse propagation in molecular air and performed additional numerical simulations, with 2.5 mJ Gaussian input pulses $w_0 = 3.5$ mm, $t_{\text{FWHM}} = 45$ fs. These initial conditions match the experimental input pulse parameters as closely as possible. The numerical simulation shows two distinct ionization zones and a characteristic Q-shaped XFROG spectrogram (figure 4(b)) emerging after the refocusing stage and the corresponding split–isolation cycle (figure 5(a)). Thus, the numerical data reproduce the characteristic features of the measured pulses, including red-shifted leading and blue-shifted trailing pedestals, as also observed in numerical simulations of double self-compression in argon. In addition, figure 5(b) shows spectra from experiment and theory. Both simulated and experimentally recorded spectra exhibit a pronounced red-shift, which, according to our previous discussion, emerges due to spatio-spectral reshaping of the pulse during the refocusing stage. In figure 5(c), a comparison is shown between on-axis temporal profiles of measured and simulated pulses. The measured pulse exhibits a duration $t_{\text{FWHM}} = 22$ fs, whereas the simulated pulse has $t_{\text{FWHM}} = 14$ fs on-axis.
Figure 6. Pulse compression ratio in a krypton filament for various initial conditions in the energy versus $P/P_{\text{crit}}$ plane. The solid lines correspond to lines of equal pressure. The dashed line separates the subdiffractive channel regime (below) from the double self-compression regime (above).

The cascaded compression scenario is not an isolated phenomenon, but can be obtained for a range of input pulse parameters and gas species, which sets it apart from a highly optimized single-compression scenario. Assuming a different gas, e.g. krypton, as the nonlinear medium [20, 21], for a demonstration of the universality of this mechanism we have scanned the parameter range of input pulse energy and peak power in numerical simulations for the appearance of this phenomenon. Beam waist and temporal duration were fixed at $w_0 = 5$ mm and $t_{\text{FWHM}} = 120$ fs, respectively. The observed pulse shortening as a function of input energy and system nonlinearity (peak power normalized to $P_{\text{crit}}$) is depicted in figure 6, with iso-pressure lines shown in white. The dashed line, roughly collinear with the 100 kPa pressure line, marks the lower limit of double self-compression. From this picture, the capability of the cascaded self-compression immediately becomes clear, giving rise to up to twelvefold compression. Compression ratios above 10 are localized in the region of double self-compression and can already be observed at powers exceeding the critical power by a factor of only three. Our scan also reveals examples of threefold cascading of the split–isolation cycle, yet with imperfect isolation in the last cycle. Generally, for pressures exceeding 160 kPa, we observe an increased tendency for such undesired multiple temporal splits. Importantly, cascaded self-compression fills a considerable fraction of the parameter space mapped out in figure 6. This sets it apart from sparsely represented rogue-wave-like events [13].

Our numerical investigations and experimental studies pinpoint an alternative approach toward efficient exploitation and control of highly nonlinear wave-shaping mechanisms. Rather than trying to confine input parameters in an increasingly narrow range, it appears much more
promising to relax these constraints in order to avoid input noise strongly affecting the output waveform. We demonstrated that physical systems exist that allow for cascaded application of the waveform-shaping effect, e.g. in order to compress optical pulses or to concentrate energy. While this effect certainly also narrows the input parameter space, this narrowing is minor by comparison to the immediate rogue wave control that exhibits a rapidly imploding parameter space with increasing amplitude [12]. Our cascaded compression method, therefore, opens up a perspective not only for optical pulse compression but also for exploitation of waveform control in a wide range of similar highly nonlinear physical scenarios.

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