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Self-compression of 120 fs pulses in a white-light filament

Jens Bethge¹, Carsten Brée^{1,2}, Harald Redlin³, Gero Stibenz⁴,
Peter Staudt⁴, Günter Steinmeyer¹, Ayhan Demircan² and
Stefan Düstener³

¹ Max-Born-Institut für Nichtlineare Optik und Ultrakurzzeitspektroskopie,
Max-Born-Straße 2a, D-12489 Berlin, Germany

² Weierstraß-Institut für Angewandte Analysis und Stochastik (WIAS), Mohrenstraße 39,
D-10117 Berlin, Germany

³ Deutsches Elektronen-Synchrotron (DESY), Notkestraße 85, D-22607 Hamburg, Germany

⁴ Angewandte Physik und Elektronik GmbH (APE), Plauener Straße 163-165,
D-13053 Berlin, Germany

E-mail: bethge@mbi-berlin.de

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Abstract

Self-compression of pulses with more than 100 fs input pulse duration from a 10 Hz laser system is experimentally demonstrated, with a compression factor of 3.3 resulting in output pulse durations of 35 fs. This measurement substantially widens the range of applicability of this compression method, enabling self-compression of pulsed laser sources that neither exhibit extremely low pulse-to-pulse energy fluctuations nor a particularly clean beam profile. The experimental demonstration is numerically modeled, revealing the exact same mechanisms at work as at shorter input pulse duration. Additionally, the role of controlled beam clipping with an adjustable aperture is numerically substantiated.

Keywords: pulse compression, filamentation, ultrashort pulses

(Some figures in this article are in colour only in the electronic version)

1. Introduction

Immediately after its first description by Hauri *et al.*, femtosecond filamentation found widespread application for compressing laser pulses with energies in the range from a hundred microjoules up to several millijoules [1]. Compared to other pulse compression techniques, such as the hollow fiber compressor [2], self-guiding filaments are intriguing as they completely eliminate the potential damage issues of a guiding structure. While early demonstrations of filament compression still required chirped mirrors for dispersion compensation [1], more recent demonstrations of filament compression achieved spectral broadening and dispersion compensation in the filament channel [3]. The self-compression of laser pulses further simplifies the generation of intense short laser pulses and increases the attractiveness of filament compression schemes.

Despite numerous experimental demonstrations of both self-compression [3–9] and chirped mirror based filament compression [1, 10–12], however, optimized filament compression is still a very challenging task as it can only be successfully exploited in a small parameter window at the verge of two competing collapse scenarios. While increasing the intensity or gas pressure will enhance the spectral broadening and thereby the potential shortness of a compressed pulse, such simple parameter scaling is ultimately limited by the break-up of the beam profile into multiple filaments [13, 14], as originally suggested by Bepalov and Talanov [15]. This spatial break-up is induced by an azimuthal modulational instability, seeded by initial spatial beam distortions. Additionally, a second break-up scenario can easily occur along the propagation axis, giving rise to temporal fragmentation of the pulse into an irregular train of two or more pulses [3, 16]. Ultimately, neither type of fragmentation, spatial or temporal, can be tolerated in

Figure 1. Schematic diagram of the experimental setup. M: mirror; W: wedge; A1–A3: apertures.

applications of compressed pulses. Yet, optimum performance of the self-compression scheme is often found at the verge of instability.

The proximity to break-up scenarios becomes a problem when lasers with strong energy fluctuations are being used for self-compression. Consequently, most demonstrations of filament compression have nearly exclusively utilized kilohertz femtosecond amplifier systems with their comparatively relatively weak pulse-to-pulse fluctuations. Given the currently available pump power and cooling technology, such lasers are typically limited to about 5 mJ pulse energy. Compared to their few-hertz counterparts, kilohertz lasers typically exhibit superior beam quality and pulse durations below 100 fs, which are considered to delay multifilament and temporal break-up, respectively. Given that there is little alternative for scaling pulse energies above 10 mJ, however, it is the few-hertz class of lasers that could benefit most from filamentation as there are essentially no working schemes for pulse compression in this regime. For 10 Hz laser systems filament self-compression has been demonstrated [17], however, only for pulse durations smaller than 50 fs. In the following, we will demonstrate filament self-compression with a 10 Hz laser system that neither exhibits exceptional beam quality nor sub-100 fs pulse duration. We show that it is possible to exploit filament self-compression with such a laser, and indicate a less stringent set of criteria that needs to be fulfilled.

2. Experimental setup

Figure 1 shows the setup used in the experiments reported. A commercial system consisting of a regenerative Ti:sapphire amplifier (Hydra-25 F, Coherent Inc.) in combination with a two-pass booster amplifier system that delivers up to 25 mJ pulses at a repetition rate of 10 Hz is used. The amplifier is pumped with a flash lamp pumped laser (Surelite II, Continuum Inc.). However, for our self-compression experiments only a 5 mJ fraction of this energy was used, which was further attenuated down to about 1 mJ by an iris. For a complete characterization of the laser pulses we use spectral phase interferometry for direct electric-field reconstruction (SPIDER) [18]. For our measurements, we use commercial apparatus that employs asymmetric group velocity matching in a long nonlinear optical crystal, i.e., a variant that is also known as LX-SPIDER [19]. This technique enables

Figure 2. (a) Spectral power density of a representative input pulse (red line), an experimentally measured output pulse after filamentary propagation (black line), and a theoretically predicted output pulse (green line, see section 4). (b) Spectral phases of the measured input pulse, the output pulse, and the theoretically predicted output pulse. (c) The resulting temporal structure of the input pulse, the output pulse, and the calculated output pulse.

reliable and immediate pulse characterization of the >100 fs input pulses as well as of the compressed output pulses in the sub-40 fs regime. Like all other SPIDER techniques, LX-SPIDER measures the spectral phase of the pulses under test, allowing for reconstruction of temporal pulse profile and chirp with additional information on the spectrum of the pulses. Figures 2(a) and (b) show the measured spectral power density of the input pulses (red line) and the spectral phase from the LX-SPIDER measurements (red line). The resulting temporal profile of the pulse is indicated by the red line in figure 2(c). The profile of the input pulses exhibits only negligible deviations from the ideal profile of a bandwidth-limited pulse (dashed line) at negative delay positions, as shown by the green line in the same graph. Thus, the pulse duration of 115 fs cannot be significantly reduced without using nonlinear optical effects for expanding the bandwidth of the pulses.

In our experiment, self-generated filamentation serves for spectral broadening. The filament is generated within a Brewster windowed gas cell as previously employed for various compression methods in the millijoule range [1–3]. The main advantage of such a cell over simply using atmospheric air [12] is the adjustability of the nonlinearity via the pressure and the gas fill, for which we used krypton

at atmospheric pressure throughout. It should be noted that the unavoidable 0.5 mm thick silica windows of the cell have been discussed to have an important impact on pulse formation [20]. As our laser system delivers relatively long > 100 fs output pulses, we can replace the focusing mirror by a simple lens, avoiding potential issues with astigmatism, which allows for some simplification. This measure may also to some extent delay spatial break-up of the filament [21]. For the measurements reported in this paper, a lens with a focal length of $f = 75$ cm is used at a distance of 25 cm to the Brewster-oriented input window. This lens exhibits half of the focal length used in previous experiments on self-compression [3]. This rather tight focusing is a necessary compromise for a compact in-line setup. Our setup additionally contains three adjustable apertures before and after the focusing lens and after the gas cell. Adjusting the diameter of the first diaphragm clips the outer rim of the spatial beam profile and is particularly important for obtaining a spatially stable single filament. The second aperture is helpful for fine tuning of the beam profile and the pulse energy, providing an additional degree of freedom to produce a single filament and to prevent multifilamentation. The third aperture, finally, only serves to limit the amount of light entering the LX-SPIDER apparatus and to restrict the measurement to the core of the filament.

For the application of pulse self-compression, the laser system used is not ideal in several aspects. One apparent problem of high-pulse energy systems is deviation from a Gaussian beam profile, in particular small-scale inhomogeneities. An added problem is the poor pointing stability of such systems. Figure 3 shows the color-coded intensity beam profile of the laser, with lighter colors indicating higher energies. The beam profile shows an inhomogeneous mode structure with a vertical width of $\Delta Y = 25$ mm and a horizontal width of $\Delta X = 20$ mm. This profile exhibits several hot spots, which may compete in seeding independent filaments provided sufficient input energy. We measured a series of 1000 successive beam profiles and computed the center of gravity of each frame, as indicated by the red dots in figure 3. The rms spread of these dots amounts to $168 \mu\text{m}$, i.e., just 1% of the beam dimensions. To stabilize the filament, we spatially filtered the input beam with a diaphragm, rejecting all light outside a 1.5 mm diameter around the center of gravity. This portion of the beam also shows the weakest pulse-to-pulse energy fluctuations, amounting to about 10% rms for 1000 recorded profiles. The remaining pulse energy amounts to about 1 mJ. The resulting beam is more stable in time and shows a much cleaner beam profile, which may further improve due to the self-cleaning effect during filamentary propagation [22].

3. Experimental results

Under the described experimental conditions inside the gas cell, we observe formation of a single filament that complies with the criteria in [23]. In particular, the filament exhibits a strongly ionized region at the location of the classical focus, followed by a post-ionization regime. In kilohertz experiments both these zones are typically clearly distinguishable and

Figure 3. A characteristic beam profile of the laser, showing different hot spot areas and one prominent hotspot (lighter colors indicate higher energies). The red dots show the position of the center of gravity for 1000 successive pulses. The inset shows the whole beam profile with a diameter of about 1 inch.

exhibit a characteristic fluorescence and scattered white-light, respectively. In the current experimental setup, it is not possible to clearly measure the total length of the filament, since the intensity of the light scattered from the post-ionization channel is an order of magnitude lower, due to the low repetition rate of the laser system used. However, filament formation is evident from the beam profile after the cell, indicating beam confinement over more than the Rayleigh length of the focal geometry employed here. Additionally, strong nonlinear interaction can be deduced from the output spectrum after the cell. By increasing the aperture diameter, i.e., also increasing pulse energy, one first observes a small amount of blue frequencies created in the clearly visible plasma flame forming at the linear focus of the beam. A further increase of pulse energy leads to a sudden appearance of a significant white-light spot in the far field behind the gas cell, showing divergence behavior different from the original beam. Still further increasing the energy, multiple filaments start to form, as indicated by two or more spots in the far field. In contrast to the behavior of the single filament these multiple filaments are not stable in position and show unpredictable dynamics. The broadest spectrum and best compression ratio have been observed for an energy slightly below the onset of multiple filamentation, similar to the observations in [3].

Figures 2(a) and (b) show the measured spectral amplitude and phase of the pulse after the filamentary propagation, respectively. The measured data are shown as a black curve, which is to be compared with the numerically simulated data (green curves) that will be further discussed in section 4. The initial conditions used in the numerical simulations are adopted from the measured experimental input pulse parameters. Inspecting the output spectrum shown in figure 2 ((a), black line) and comparing it to the input pulse (red line), one finds prevalent spectral broadening towards blue

frequencies (380–405 THz) by about 25 THz. This is contrasted by a marginal 4 THz broadening towards the red. This asymmetric broadening is another indication for a strong influence of plasma effects in the broadening process. The characteristic asymmetry has been previously explained by self-steepening [24], and is typical for filament compression. Compared to the ≈ 8 THz width (FWHM) of the input spectrum, the spectrum from the filament encompasses a 4–5 times wider range. The Fourier-limited pulse duration of the filament output amounts to 22 fs, indicating a possible 4.7-fold temporal compression in the filament process.

The spectral phase as measured with the LX-SPIDER apparatus is plotted in figure 2 ((b), black curve). The spectral phase is relatively flat up to 390 THz and starts to roll off in the extreme blue range of the newly generated spectral content. For the new frequencies the phase profile remains linear, i.e. almost no GDD is observed. Numerical simulations indicate a similar behavior with a flat spectral phase in the central part of the spectrum, however starting to roll off some 10 THz lower than in the experiments.

Figure 2(c) shows the resulting temporal profile of the self-compressed pulse (red line). Comparing the pulse duration one finds a compression by a factor of 3.3 from 115 to 35 fs. The pulses were characterized after a third aperture, which is placed after the gas cell and which separates the filament light from light that was scattered into the reservoir region. This isolates the white-light core of the filament. The core has a highly improved spatial profile compared to the initial profile of the beam and is spatially stable, with an rms deviation of pulse energy of about 13%, an instability only 3% higher than that of the input pulses.

4. Numerical simulations

Filamentary self-compression has been previously discussed as mainly resulting from spatial effects [1, 5, 16, 25]. As the pulse is spatially confined in traditional compression schemes [26], energy transport can only occur along the longitudinal axis. Filament scenarios do not obey any such constraints on the beam radius, and energy transport in the radial direction may also occur. In particular, this may give rise to a time-dependent beam diameter along the pulse. Such spatial mechanisms have been recently confirmed as the dominant mechanism behind filament self-compression of sub-50 fs input pulses. This serves as a motivation for our numerical studies, which we conducted to see whether the now widely recognized explanation for self-compression also applies for longer pulse durations.

For this purpose, we performed numerical simulations of the generalized nonlinear Schrödinger equation [5], modeling filamentary propagation of the envelope of the light electric field. To match the experimental conditions, special emphasis is put on proper modeling of the initial conditions, for which we used experimentally measured SPIDER data. In view of its important role in the experiments, we included the second aperture in our numerical modeling. The envelope of the initial

electric-field distribution used in the numerics is described by

$$\mathcal{E}(r, t) = \sqrt{I_{\max}} \exp \left(-2 \frac{r^2}{w_0^2} - 2 \frac{r^{16}}{d_{\text{ap}}^{16}} \right) \times \int_{-\infty}^{\infty} d\omega \exp \left(i \frac{r^2}{2k(\omega)f} + i\phi(\omega) + i\omega t \right) \sqrt{\tilde{I}(\omega)}. \quad (1)$$

Here, the initial beam waist is given by $w_0 = 2.5$ mm, a value which appears reasonable for the hot spot selected by the first aperture. $I(\omega)$ and $\phi(\omega)$ are the measured spectrum and spectral phase of the input pulse, respectively. $I_{\max}$ is the peak temporal intensity, assuming a proper normalization of $\tilde{I}(\omega)$. The peak intensity of the input pulse can then be estimated according to

$$I_{\max} \approx \frac{2E_{\text{in}}}{\pi w_0^2 \int dt I(t)}, \quad (2)$$

where $I(t)$ is the temporal intensity profile reconstructed from measured SPIDER data, normalized to unity. The diameter of the second aperture is given by d_{ap} , and $E_{\text{in}} \approx 1$ mJ is the laser pulse energy transmitted through the first aperture. A super-Gaussian was used to model the aperture, to avoid numerical artifacts. As our numerical model includes space-time focusing effects that arise because different frequency components diffract into different cone angles we utilized a frequency-dependent factor $\exp(ir^2/2k(\omega)f)$, with $f = 75$ cm in the Fourier-domain for proper modeling of the lens [27]. Here $k(\omega) = n(\omega)\omega/c$ is the wavenumber in krypton, with dispersion modeled according to [28].

We analyze the propagation of the apertured input pulse with $d_{\text{ap}} = 5$ mm, where the experimental SPIDER data of the 115 fs pulse are used for the initial temporal profile. The chosen aperture size amounts to a transmitted energy through the second aperture of ≈ 0.8 mJ. In fact, it turns out that the pulse duration of the numerically simulated, self-compressed pulse (34 fs) matches that of the measured pulse within experimental precision, as is obvious from the representation of the on-axis temporal intensity profile shown in figure 2.

The simulations allow for a calculation of XFROG spectrograms, which are a proven tool to analyze the spatio-temporal structure in various supercontinuum scenarios. These spectrograms can also be computed from experimental data, provided that a complete characterization method as SPIDER is used. Such spectrograms are shown in figure 4, with excellent quantitative and qualitative agreement of the experimental and simulated spectrograms. These spectrograms indicate a characteristic inverse Γ shape which emerges due to asymmetric spectral broadening in the self-steepened trailing edge of the pulse. This feature has been independently confirmed in experiments and numerical simulations [5] for shorter pulses and clearly confirms that one and the same mechanism is at work in both pulse duration regimes. This analysis also rules out spatio-temporal solitons [29] of any kind, as these would lead to rather featureless round or elliptic spectrogram representations. The evolution of the on-axis temporal profiles leading to the characteristic Γ -shaped spectrograms can be visualized by the color-coded representation of the evolution of the on-axis temporal intensity profile along the propagation direction z shown in figure 5(a).

Figure 4. (a) XFROG trace reconstructed from experimental SPIDER data. (b) XFROG spectrogram of an apertured output pulse, numerically simulated, with initial temporal profile taken from experiment. (c) Corresponding XFROG spectrogram for unapertured input pulse.

Figure 5. (a) Evolution of on-axis temporal profile, apertured input pulse. (b) Evolution of on-axis temporal profile, unapertured input pulse. (c) Evolution of peak electron density along z for apertured (solid line) and unapertured (dashed line) input pulses.

This shows temporal break-up in the vicinity of the focal region, followed by a subsequent isolation of a self-compressed trailing subpulse experiencing further temporal compression in a weakly ionized channel for $z > 0.8$ m. This behavior clearly confirms that the self-compression evolves according to the spatially dominated self-pinching mechanism of [25]. In fact, the remnants of the suppressed leading subpulse can be observed in both the measured and simulated XFROG traces, figures 4(a) and (b).

To demonstrate the impact of the aperture, we performed a simulation with unchanged initial conditions, but neglecting the aperture ($d_{\text{ap}} = \infty$). In this case, the full 1 mJ energy of the unapertured input pulse is available for filamentary propagation. The spectrogram representation of the emerging output pulse shown in figure 4(c) reveals that the pulse has experienced less spectral broadening in the trailing edge as compared to the apertured pulse. Also, we observe an increased pulse duration (FWHM) of ≈ 36 fs. Thus, we observe that aperturing the input pulse enhances the spectral broadening. Indeed, a comparison of the evolution of apertured and unapertured input pulses in the numerical simulations shown in figures 5(a) and (b) and the evolution

of the peak electron densities for both cases as shown in figure 5(c) reveal the origins of this surprising effect. In fact, the dynamical behavior for the unapertured case figure 5(b) agrees qualitatively with the evolution of the apertured pulse figure 5(a), yet with reduced longitudinal dimension of the nonlinear focus and a reduction of the intensity in the weakly ionized channel, as is obvious from comparing the evolution of the peak electron densities for the two cases as shown in figure 5(c). We thus observe that this reduces the efficiency of the spectral broadening for the unapertured case and the given aperture and input pulse parameters. Finally, the enhanced nonlinearity encountered by the apertured input pulse increases its spectral broadening and gives rise to a reduced pulse duration for the output pulse.

In conclusion, our results clearly emphasize the importance of proper modeling of initial conditions, leading to a remarkable qualitative agreement between the experimentally measured pulse durations and numerical simulations. In fact, whether self-compression can be observed in our simulations or not depends most crucially on the diameter of the second aperture, as has also been experimentally verified in [30]. With appropriately chosen aperture diameter, the simulations correctly predict an approximate fourfold temporal compression of the initial 115 fs input pulse as well as the characteristic Γ shaped XFROG traces.

5. Discussion

As the experiments clearly reveal, filament self-compression can be exploited even with laser systems that appear to be prone to inducing either spatial or temporal break-up in the compression process. In particular, at first sight, it seems surprising that a beam profile with several hot spots (cf figure 3) does not automatically and unavoidably induce multiple filamentation. Let us recall, however, that filament formation is described by a deterministic set of equations, which lead to the same spatial and temporal characteristics when the same input parameters are fed into the system. While the presence of hot spots in the beam profile will certainly favor initialization of filaments at these locations, only strong pulse-to-pulse fluctuations of either beam profile or temporal pulse shape are expected to turn out prohibitive for meeting

Figure 6. Comparison of power levels required to meet the condition for self-compression. The ordinate is normalized to the critical power P_{cr} . The circles refer to kilohertz repetition rate experiments (Hauri *et al* [4], Gallmann *et al* [8], Stibenz *et al* [3], Schulz *et al* [7] and Uryupina *et al* [17]), and the squares to 10 Hz systems (Kosareva *et al* [9] and current work).

the small parameter window where self-compression can take place. Therefore, low pulse-to-pulse fluctuations are expected to be the key parameters for successful self-compression schemes at a few-hertz repetition rate. In fact, such conditions could not be met in an earlier set of experiments with a 10 Hz laser, cf section 2 in [5]. In these experiments, single filamentation and a short compressed pulse could only be seen for a small percentage of the laser shots. In addition, our numerical analysis clarifies the role of aperturing the input pulses. This measure not only helps to stabilize the filament, preventing spatial break-up and multiple filamentation, but it also increases the longitudinal extension of the nonlinear interaction region in the vicinity of the focus, giving rise to spectrally broader and temporally shorter output pulses.

For further analysis of our results, we have compiled a comparison of input peak powers required to meet the conditions for filament self-compression in various experiments, see figure 6. These data comprise experiments at 10 Hz and kHz repetition rates as well as in different noble gases. Quite clearly, the experiments at 3 mJ input pulse energy or below used input powers that were only about a factor three above the critical power $P_{\text{crit}} = \lambda^2/2\pi n_2$. All experiments at higher pulse energies, however, also reported significantly higher values of up to 20 critical input powers, which is somewhat surprising as multiple filamentation appears hard to avoid at such elevated power levels. In our current experiments, using krypton as the medium we could reach self-compression at only eight critical powers, which is significantly lower than in the only other 10 Hz experiment published so far. This may pave a way towards further increase of pulse energies without suffering from spatial break-up. Given that the peak powers in our experiment are quite high in comparison to P_{crit} , it may also be interesting to explore the role of higher-order nonlinearities [31] in this particular parameter range. Even though it has been found impossible so far to explain self-compression in the absence of plasma formation by numerical simulations, higher-order Kerr nonlinearities may

nevertheless have an influence on the pulse shaping process for our experimental conditions.

6. Conclusion

Among the different criteria that have been brought up for deciding the suitability of an amplifier system for self-compression, our experiments clearly indicate a surprisingly low importance of pulse duration and spatial beam quality. Starting with pulses of 115 fs duration, a compression factor larger than three could be obtained. This extends the applicability of self-compression schemes to durations that are about three times as long as used in most previous experiments. We numerically verified the role of the input aperture and demonstrated its importance for effectiveness of the self-compression mechanism. However, our experiments also emphasize the importance of small pulse-to-pulse fluctuations for successful application of this compression method. Obviously, this criterion not only includes fluctuations of the pulse energy but also requires small pulse-to-pulse variations in the beam profile and temporal structure of the pulse, which are, however, difficult to monitor. Given that these criteria can be fulfilled at higher pulse energies and given that the nonlinearity is adapted by a suitable choice of medium and pressure, we do not see any systematic limitation for application of self-compression schemes. Using gases with lower nonlinearity, self-compression may therefore open up a perspective at pulse energies where practically no active compression schemes are available to date, allowing for the concentration of energy to a volume encompassing only a few wavelengths laterally as well as longitudinally.

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