Generation of supercontinuum compressible to single-cycle pulse widths in an ionizing gas

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Abstract. Analysis of the evolution of submillijoule few-cycle light pulses in an ionizing gas reveals physical mechanisms and scenarios enabling the generation of high-intensity supercontinuum radiation compressible to single-cycle pulse widths. The main properties of supercontinuum generation in this regime are analyzed by numerically solving a spatiotemporal evolution equation for a few-cycle light field adapted to include ultrafast ionization phenomena in a gas medium. The enhancement of ionization-induced phase shifts and shock-wave effects, which is inherent in few-cycle fields, is shown to be the key factor for a substantial intensification of the high-frequency wing of the spectrum, giving rise to broadband radiation with appropriate spectral phase profiles that can be compressed to single-cycle pulses upon an accurate temporal and spatial chirp compensation. The dynamics of few-cycle light fields in an ionizing gas is shown to be sensitive to the initial pulse width, pulse shape and ionization regime.

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1. Introduction

Ultrashort high-intensity light pulses propagating through an ionizing gas undergo cascades of spectral, temporal and spatial nonlinear–optical transformations, often leading to a dramatic broadening of the spectrum of the laser field [1, 2]. This phenomenon, usually referred to as supercontinuum generation, involves an intricate combination of strongly coupled nonlinear–optical effects, making it an interesting and challenging object for fundamental studies in ultrafast nonlinear–optical physics [3, 4]. From the standpoint of advanced optical technologies, supercontinuum generation in the gas phase is viewed as a promising strategy of pulse compression, compatible with high field intensities, which suggests a way toward shortening high-intensity laser fields to few- and even single-cycle pulse widths. Hollow waveguides have been shown to substantially enhance spectral broadening of high-intensity ultrashort laser pulses due to the Kerr [5]–[7] and Raman [8] effects, thus offering a powerful technology for the synthesis of few- to single-cycle field waveforms [9]. Freely propagating high-intensity laser beams, on the other hand, tend to form filaments [10] as a result of Kerr-lens self-focusing balanced by an ionization-induced self-defocusing. The length of nonlinear interaction in the filamentation regime is often sufficient for a dramatic spectral broadening of a laser field [11]–[13], while the joint action of dispersion and nonlinearity in a filament can provide an efficient self-compression of a laser field to a few-cycle pulse width [14, 15]. In certain situations, this makes filamentation-assisted pulse compression a competitive alternative to the pulse-compression technique based on hollow waveguides.

In this work, we show that the spectral broadening of high-intensity few-cycle pulses in an ionizing gas can be tailored, with an appropriate choice of gas and input-pulse parameters, toward the generation of even shorter field waveforms, allowing the enhancement of carrier–envelope-phase control and offering attractive solutions for the synthesis of powerful sub-cycle field waveform shaping. Based on the analysis of the evolution of submillijoule few-cycle light pulses in an ionizing gas, we demonstrate physical mechanisms and scenarios...
enabling the generation of high-intensity supercontinua that can be compressed to single-cycle pulse widths upon an accurate compensation of the temporal and spatial chirp of the light field. We show that the enhancement of ionization-induced phase shifts and shock-wave effects for few-cycle fields is the key factor that substantially intensifies the high-frequency wing of the field spectrum, thus allowing the generation of broadband spectra with appropriate spectral phase profiles supporting compression to single-cycle pulse widths. Dynamics of few-cycle light fields in an ionizing gas will be shown to be highly sensitive to the initial pulse width, pulse shape and regime of ionization.

2. Theoretical framework

To analyze the evolution of high-intensity few-cycle light pulses in an ionizing gas, we employ the framework of slowly evolving wave approximation (SEWA) [16], adapted to include ionization effects [17, 18]. Our SEWA-based model of few-cycle pulse propagation involves a numerical solution of the following equation for the evolution of the field $E$:

$$\frac{\partial E(z, r, \omega)}{\partial z} = i \left( k(\omega) - \frac{\omega}{V_g} \right) E(z, r, \omega) + \frac{i \omega}{2c} \chi^{(3)} \hat{F}(E^3(z, r, t))$$

$$- \frac{1}{2} \hat{F} \left[ \frac{I_p}{I(z, r, t)} \frac{\partial n_e}{\partial t} E(z, r, t) \right] - i \frac{1}{2\omega c} \hat{F} \left( \omega_p^2(z, r, t) E(z, r, t) \right)$$

$$+ i \frac{1}{2k(\omega)} \left[ \frac{\partial^2}{\partial r^2} + \frac{\partial}{r \partial r} \right] \hat{F}(E(z, r, t)).$$

(1)

Here, $E(z, r, \omega) = \hat{F}^{-1}(\sqrt{I_0} A(z, r, t) \exp(i\omega_0 t) + \text{c.c.})$, $\hat{F}(\bullet)$ is the Fourier transform operator, $I_0$ is the peak field intensity, $A(z, r, t)$ is the field envelope, $z$ and $r$ are the longitudinal and radial coordinates, respectively, $\omega$ is the radiation frequency, $V_g$ is the group velocity, $\chi^{(3)}$ is the third-order susceptibility, $I_p$ is the ionization potential, $\omega_p(z, r) = [4\pi^2 e^2 n_e(z, r)/m_e]^{1/2}$ is the plasma frequency, $e$ and $m_e$ are the electron charge and mass, respectively, and $n_e$ is the electron density. Dispersion of the gas medium is plugged into equation (1) through the frequency profile of the wave number $k = k(\omega)$, which automatically includes high-order dispersion effects. The electron density $n_e$ created in the gas by a laser field was calculated by solving the kinetic equation $\partial n_e/\partial t = w(n_0 - n_e)$ with the ionization rate $w(t)$ calculated using the Popov–Perelomov–Terentyev modification [19] of the Keldysh formula [20] for the ionization rate.

Analysis based on equation (1) includes effects related to gas and plasma dispersion, beam diffraction, Kerr-effect-induced spatial and temporal self-action of the laser field, as well as ionization-induced loss and nonlinear–optical phenomena. One of the main goals of the analysis presented in this paper is to identify the key mechanisms leading to an efficient spectral broadening of few-cycle laser pulses in an ionizing gas medium and to isolate, wherever possible, the impact of each of these mechanisms on pulse-propagation dynamics. To explain the procedure enabling us to accomplish this goal, it is instructive to represent the field evolution
equation in the form of an equation for the temporal envelope $A(z,r,t)$:

$$\frac{\partial A(z,r,t)}{\partial z} = -\frac{1}{2} \left[ \frac{I_p}{I(z,r,t)} \frac{\partial n_e(z,r,t)}{\partial \tau} \right] A(z,r,t) + i \sum_{m=2}^{\infty} \frac{i^m}{m!} \frac{\partial^m k}{\partial \omega^m} A(z,r,t) \times \frac{\partial^m A(z,r,t)}{\partial \tau^m} + i \frac{c}{2 \omega_0} \left[ \frac{\partial^2}{\partial \tau^2} + \frac{\partial}{\partial r} \frac{\partial}{\partial r} \right] \left[ 1 + \frac{i}{\omega_0} \frac{\partial}{\partial \tau} \right]^{-1} A(z,r,t)$$

$$+ in_2 I_0 \frac{\omega_0}{c} \left( 1 + \frac{i}{\omega_0} \frac{\partial}{\partial \tau} \right) |A|^2 A - i \frac{1}{2 \omega_0 c} \left[ 1 + \frac{i}{\omega_0} \frac{\partial}{\partial \tau} \right]^{-1} \left[ \omega_p^2(z,r,t) A(z,r,t) \right],$$

(2)

where $\tau$ is the retarded time, $n_2$ is the nonlinear refractive index of the gas, $\omega_0$ is the central frequency of the laser field, and the influence of third-harmonic generation is neglected.

With the pulse-propagation equation written in the form of equation (2), effects related to conventional self-phase modulation (SPM) can be easily isolated by dropping the $n_e$ and $\omega_p$ plasma terms and omitting the $(i/\omega_0)(\partial/\partial t)$ shock-wave part of the Kerr-nonlinearity term. The influence of shock-wave effects can also be readily assessed by simply switching on and off the $(i/\omega_0)(\partial/\partial t)$ terms in equation (2). By contrast, the purely ionization-induced part of spectral broadening is quite hard to isolate from spectral broadening due to other nonlinear–optical processes, since there is no simple way to exclude ionization terms from equations (1) and (2) without losing a balance between the Kerr-lens self-focusing and self-defocusing due to plasma electrons. In view of this difficulty, a special procedure has been implemented allowing the ionization-related part of spectral broadening to be artificially switched on and off without perturbing the balance between the Kerr- and ionization-induced lenses. This technique involves an artificial suppression of the time dependence of the electron density $n_e$ in equation (2) by setting $n_e$ at each step in $z$ equal to the value that the electron density would acquire for a given $z$ by the end of the light pulse.

The numerical procedure used in this work to solve equation (1) was based on the split-step Fourier technique [21]. Simulations were performed on a computation domain with a radius of 1 cm and a time window of 1 ps, which included 1024 and 4096 sampling points for the radial coordinate and time, respectively. The longitudinal coordinate $z$ was discretized with a step of 10 $\mu$m. Simulations were performed for laser pulses focused by a 50 cm-focal-length lens or mirror into a 30 cm chamber filled with 25 atm of helium ($n_2 \approx 3 \times 10^{-21}$ cm$^2$ W$^{-1}$). These parameters have been chosen in such a way as to model recent experiments [22], demonstrating a highly efficient generation of supercontinuum radiation with a spectrum spanning the range 270–1000 nm by a few-cycle laser pulse focused in a high-pressure helium chamber. The high-ionization potential of helium, as shown by simulations presented below in this paper, as well as recent experiments [22], helps to increase the field intensity in a focused beam relative to lower $I_p$ gases. This conclusion is consistent with the recent theoretical comparison of filamentation in low and high $I_p$, presented by Couairon et al [23]. A high gas pressure has been chosen in order to compensate for the low optical nonlinearity of helium.

3. Tailoring supercontinuum spectra

3.1. Pulse-width effects

We start our analysis with the discussion of the influence of laser-pulse parameters, such as pulse duration, pulse shape, and chirp, on the spectral properties of supercontinuum generated as
Figure 1. Spectra of laser pulses transmitted through a 30 cm gas chamber filled with helium at a pressure of 25 atm. The input laser pulse has a Gaussian envelope, a peak power of 44 GW, and a pulse width of 3 fs (1), 4 fs (2), 5 fs (3), 10 fs (4), 30 fs (5) and 50 fs (6). The input laser beam is focused with a 50 cm-focal-length lens or mirror. The inset shows the same spectra, but presented on a linear scale.

A result of nonlinear-optical pulse transformation in an ionizing gas medium. We first examine pulse-width-sensitive features in supercontinuum generation. To this end, we solve the field-evolution equation (1) for input laser pulses with a Gaussian shape of temporal envelope. For a fair comparison of supercontinua generated by laser pulses with different initial pulse widths, we fix the input peak power at a level of $P_0 = 44$ GW (corresponding to approximately $4P_{cr}$, where $P_{cr}$ is the critical power of self-focusing) and vary the initial temporal duration of laser pulses from 3 to 50 fs. As a beam carrying a peak power exceeding $P_{cr}$ becomes unstable with respect to multiple filamentation, the initial conditions were chosen in such a way as to minimize the influence of multiple filamentation within the interaction length through a small beam width and a short beam waist in the focal region. The spectra of supercontinua generated by such pulses in a gas chamber filled with 25 atm of helium are presented in figure 1. It can be seen from the results of these simulations that shorter input pulses tend to generate spectra with more pronounced asymmetry and enhanced high-frequency wings. These features reflect the growing significance of shock-wave effects and ionization-induced phase shifts for very short light pulses. Indeed, the mathematical smallness of the shock-wave term in equation (1) is controlled by the ratio of the field cycle period $T_0$ to the pulse width $\tau_p$. As a result, the significance of this term rapidly increases as the temporal envelope of the light field becomes shorter.

With the pulse width $\tau_p$ approaching the field cycle period $T_0$, the high-frequency wing in the spectrum of output radiation in figure 1 is enhanced to such an extent that a plateau develops within the frequency range stretching from approximately $1.5\omega_0$ to $2.7\omega_0$ (curves 1–3) in place of an exponentially decaying short-wavelength tail of the spectrum, which is observed for longer pulses (curves 5 and 6). This plateau is readily seen in the spectrum of the output field presented.
on a linear (see the inset to figure 1) rather than logarithmic scale, which would be necessary to visualize an exponentially decaying blue wing in the spectra of longer light pulses.

3.2. Pulse-shaping control of supercontinuum generation

Figure 2 illustrates the influence of the initial pulse shape and initial chirp on the spectrum of supercontinuum radiation generated in an ionizing gas. Here, we compare the spectra of supercontinua generated by a 5 fs transform-limited Gaussian light pulse and a pulse with the initial spectrum and spectral phase defined as shown in the inset to figure 2. This type of input field mimics the conditions of experiments in [22]. In the time domain, the field defined by the spectrum and the spectral phase shown in the inset to figure 2 is represented by a pulse with a duration of about 5 fs. The two pulses are assumed to have the same peak power, $P_0 = 60$ GW. As can be seen from figure 2, a moderate chirp imposed on an input few-cycle light field does not lead to a dramatic change in the total bandwidth of supercontinuum generated in an ionizing gas. Both the high- and low-frequency wings of the supercontinua generated by two different types of input light pulses are seen to closely follow the same pattern, with the output spectra stretching from roughly 270 nm in the high-frequency tail to wavelengths exceeding 1000 nm in the low-frequency part of the spectrum. While the total bandwidth of supercontinuum radiation does not exhibit a high sensitivity to a moderate chirp of the input field, the central part of the output spectrum keeps a memory of the spectral structure of the input field. For an input pulse defined by the spectrum and spectral phase in the inset to figure 2, the central part of the output spectrum is seen to feature a much more complicated behavior compared with the spectrum of supercontinuum radiation generated by the transform-limited Gaussian pulse.
3.3. Shock-wave- and ionization-enhanced blue shifting

To assess the significance of the key physical mechanisms of spectral broadening for the enhancement of spectral broadening in the regime of few-cycle light pulses, we performed modeling using an artificially modified field-evolution equation as explained in section 2. The results of this analysis are presented in figure 3. The dash–dotted line in this figure shows the spectrum broadened by pure SPM, which was calculated with the right-hand side of equations (1) and (2) reduced to SPM and linear dispersion terms. Spectral broadening due to SPM and shock waves is presented by the dotted line. The joint effect of SPM and ionization is illustrated by the dashed curve. As can be seen from the comparison of these results, shock-wave pulse reshaping and ionization-induced phase shifts substantially enhance the high-frequency part of the spectrum and can noticeably suppress the long-wavelength spectral wing relative to purely SPM-broadened spectra. For the chosen parameters of the input pulse and the gas medium, the joint action of the shock-wave and ionization effects shifts the high-frequency cutoff of the supercontinuum spectrum by at least 350 nm and leads to an order-of-magnitude enhancement of the 300–700 nm spectral region, giving rise to a broad plateau in this range of the supercontinuum spectrum. As demonstrated in earlier work, ionization-induced blue shifting plays an important role in the dynamics of high-intensity ultrashort pulses in gas media, as well as in filamentation-assisted pulse compression [24]–[26]. This effect may also help to optimize ultrashort light pulses for the generation of isolated attosecond field waveforms [23, 27].
3.4. Sensitivity to the regime of ionization

To examine the sensitivity of spectral properties of supercontinuum radiation generated by few-cycle pulses to the regime of ionization, we performed simulations using two models of ionization for the calculation of the ionization rate \( w(t) \). The first model was based on the Popov–Perelomov–Terentyev modification \([19]\) of the Keldysh formula \([20]\). The second model assumed a simple \( \propto \sigma_n I^n \) scaling of the ionization rate as a function of the field intensity \( I \), with \( \sigma_n \approx 1.93 \times 10^{-10^4} \) s\(^{-1}\) cm\(^2\) W\(^{-n}\) \([13, 28]\). Although, rigorously speaking, this latter scaling law is typical of the regime of multiphoton ionization (MPI), with \( n \) understood as the number of photons required to ionize an atom, it has been shown in earlier work \([28]\) that, with a properly chosen noninteger effective exponent \( n (n = 8.22 \) for the considered regime of pulse evolution \([13, 28]\)), this ionization model may provide a reasonable fit for the ion yield within a rather broad range of field intensities, not necessarily limited to the MPI regime.

The effective MPI ionization model with \( w = \sigma_n I^n \) generally tends to overestimate the electron density in the regime of high field intensities \( I \), where the inequality \( \gamma \gg 1 \) is no longer satisfied for the Keldysh parameter \( \gamma = \omega_0 (2m_e I_p)^{1/2} (e E)^{-1} \), where \( E \) is the field amplitude, \( I_p \) is the ionization potential, and \( e \) and \( m_e \) are the electron charge and mass, respectively, indicating the intermediate or tunneling regime of ionization. As a result, the effective MPI ionization model tends to somewhat overestimate the spectral broadening of an ultrashort laser pulse. The discrepancy between the predictions of this model and the Keldysh-formula-based model is especially noticeable in the short-wavelength wing of the supercontinuum spectrum (figure 4), where the ionization-induced broadening, as pointed out in section 3.3, is most significant. It can be also seen from figure 4 that the Keldysh-formula-based model provides better agreement with experimental results. However, the use of the effective MPI ionization model can, of course, be often justified, as this model correctly predicts the main tendencies in the behavior of the electron density as a function of time and allows a substantial simplification of computations.

4. Supercontinuum generation and decisive moments of spatiotemporal evolution of few-cycle pulses in an ionizing gas

The main aim of the analysis presented in this section is to understand the main properties of supercontinuum generation by a few-cycle high-intensity laser field in terms of spatiotemporal dynamics of this field in an ionizing gas. In figure 5, we present the results of our numerical simulations in the form of snapshots of light field intensity \( I \) in the \( \tau-r \)-plane (left) and the \( \lambda-r \)-plane (right) for different distances \( \xi \) from the entrance window of the gas cell in the decisive moments of field evolution around the waist of the laser beam, where the influence of nonlinear–optical phenomena is most dramatic. Similar to the previous sections, the initial conditions for the light field mimic experiments by Goulielmakis et al \([22]\), with the input pulse energy taken to be 0.3 mJ and the initial spectrum and spectral phase defined as shown in the inset to figure 2. The laser beam focused with a 50 cm-focal-length lens or mirror is set to propagate through a 30 cm gas chamber filled with helium at a pressure of 25 atm.

One of the most striking features in the field dynamics right before the beam-waist region, seen in the frames corresponding to \( \xi \) ranging from 13.5 to 14.2 cm in figure 5 (where positive times correspond to the leading edge of the pulse), is related to a self-steepening of the laser pulse, caused by an intensity-dependent change in the group index, which makes the peak of the laser pulse travel slower than the pulse edges. This effect intensifies the trailing edge...
Figure 4. Spectral broadening of a few-cycle laser pulse in an ionizing gas medium simulated with the use of (dotted line) the effective MPI model with $w = \sigma_n I^n$, $n = 8.22$, and (solid line) the Keldysh-type formalism. Input pulses with an energy of 0.2 mJ and a pulse width of 6 fs are focused into a gas chamber with a 1 m-focal-length mirror or lens. The dashed line presents the experimental spectrum from [12].

...of the pulse and leads to a more efficient focusing of this part of the pulse, as can be seen from the 13.5–14.2 cm frames in figure 5. In the frequency domain, this effect translates into a noticeable blue shifting, which is also clearly visible for $\xi$ ranging from 13.5 to 14.2 cm in figure 5 along with the SPM-induced broadening, observed in both high- and low-frequency parts of the spectrum.

Around $\xi = 14.2$ cm, the pulse develops a very steep trailing edge, which efficiently ionizes the gas medium, thus inducing a negative lens due to plasma electrons. Defocusing of the light beam by this lens then starts to dominate beam dynamics around the trailing edge of the laser pulse, giving rise to a delta-shaped profile of light field intensity in the $\tau - r$-plane (starting with $\xi = 15.5$ cm in figure 5). In the spectral domain, strong blue shifting of the trailing edge of the pulse generates intense spectral components in the visible, which tend to form a halo because of beam defocusing. This halo is readily seen in experimental beam patterns of supercontinuum radiation generated by few-cycle laser pulses [22]. The total energy of the light pulse reaching the output window of the gas chamber is estimated as 250 $\mu$J, with 17% of the input laser energy spent on ionization of the gas.

In the course of pulse propagation through a gas chamber from the beam-waist region to the output chamber window, the new spectral components generated through the above-specified nonlinear–optical processes tend to interfere with the high-frequency components that were initially present in the spectrum of the laser field (see the inset to figure 2), giving rise to well-resolved oscillatory features on the trailing edge of the pulse arriving at the output window of the gas chamber (figure 6(a)). An artificial attenuation of the high-frequency part of the input spectrum in our simulations resulted in a complete suppression of the oscillatory structure in $I(\tau, r)$ at the output of the gas chamber.

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Figure 5. Decisive moments of few-cycle field evolution in an ionizing gas: snapshots of the (left) spatiotemporal ($\tau$–$r$-plane) and (right) spatio spectral ($\lambda$–$r$-plane) structure of the field propagating through a 30 cm gas chamber filled with helium at a pressure of 25 atm at different distances from the entrance window of the gas chamber, ranging from 11 to 17 cm. The input spectrum and spectral phase of the laser field are shown in the inset to figure 2. The input pulse energy is 0.3 mJ. The laser beam is focused with a 50 cm-focal-length lens or mirror.
Figure 5. Continued.
Figure 5. Continued.
5. Pulse compressibility of supercontinuum radiation

In spite of the large spectral bandwidth of supercontinuum generated by few-cycle pulses under the above-specified conditions, compression of this field to single-cycle pulse widths is impossible without a careful compensation for a complicated spatially nonuniform spectral phase profile acquired by the light field at the output of the gas chamber (figure 6(a)) as a result of nonlinear–optical interactions leading to supercontinuum generation, as well as involved spatial beam dynamics. To assess the compressibility of a few-cycle pulse transformed in a filament, we fit the spectral phase \( \varphi(\omega, r) \) of the output field calculated as a function of the frequency \( \omega \) and the radial coordinate \( r \) by a polynomial \( \varphi(\omega, r) \approx \psi(r) + \phi(\omega, r) \). Here, \( \psi(r) = a_1 r^2 + a_2 r^4 \) and \( \phi(\omega, r) = \alpha_1 (1 + \xi r^2) \Delta \omega + \sum_{k=2}^{5} \alpha_k (\Delta \omega)^k / k! \), where \( \Delta \omega = \omega - \omega_0 \) is the detuning of a current frequency \( \omega \) from the central frequency \( \omega_0 \), and \( a_1, a_2, \alpha_k \) and \( \xi \) are the fitting parameters. For the conditions of experiments considered here, the best fit for the simulated phase \( \varphi(\omega, r) \) at the output of the gas chamber is achieved with \( a_1 = 2.26 \times 10^3 \) cm\(^{-2} \), \( a_2 = -3.2 \times 10^3 \) cm\(^{-4} \), \( \alpha_1 = 10 \) fs, \( \alpha_2 = 8.5 \) fs\(^2 \), \( \alpha_3 = 3.4 \) fs\(^3 \), \( \alpha_2 = 0.17 \) fs\(^4 \), \( \alpha_2 = 0.05 \) fs\(^5 \) and \( \xi = 90 \) cm\(^{-2} \).

Figures 6(a)–(d) illustrate pulse compression of a field with such a spectral phase profile through a compensation of different parts of \( \varphi(\omega, r) \). While an ideal compensation for the phase \( \varphi(\omega, r) \) to a perfectly flat, spatially uniform phase would allow pulse compression of supercontinuum radiation to a pulse width of about 1 fs (figure 6(b)), any imbalance in the compensation for the spatial chirp or high-order dispersion terms in \( \phi(\omega, r) \) dramatically complicates the spatiotemporal field structure (figures 6(c) and (d)), lengthening the pulse and resulting in the formation of a rather long pedestal, which typically carries a substantial fraction of energy of the compressed pulse. In particular, for supercontinuum radiation compressed through a compensation of the spatial chirp, as well as dispersion terms proportional to \( (\Delta \omega)^k \), \( k = 1, 2, 3, 4 \) and \( 5 \), resulting in a field structure shown in figure 6(c), the fraction of radiation energy confined to the most intense peak inside the white rectangle in figure 6(c) (corresponding to a pulse width of about 1.5 fs) is estimated as 10% of the total energy of the output supercontinuum field (the cost of supercontinuum generation under the above-specified conditions is 17% of the input radiation energy, lost for the ionization of the gas medium).
Figure 6. The field intensity $I$ calculated as a function of time $\tau$ and the radial coordinate $r$ for (a) the field at the output of the 30 cm chamber filled with 25 atm of helium and (b–d) the field after a compressor compensating for (b) both the spatial and spectral parts of the full phase $\phi(\omega, r)$, (c) dispersion terms proportional to $(\Delta \omega)^k$, $k = 1, 2, 3, 4, 5$, and the spatial chirp, and (d) full spectral phase with no spatial phase compensation. The input spectrum and spectral phase of the laser field are shown in the inset to figure 2. The input pulse energy is 0.3 mJ.

For the field waveform produced behind a pulse compressor providing an ideal compensation for the spectral phase, but leaving the spatial phase profile uncompensated (figure 6(d)), a single-cycle or even a sub-cycle pulse can be filtered out by using a small-aperture pinhole aligned with the beam axis. Such a pinhole will, of course, introduce an additional energy loss, as the energy carried by the peripheral part of the laser beam will be blocked. In particular, for the field with a spatiotemporal structure $I(\tau, r)$ defined by figure 6(d), integration $\tilde{I}(\tau) = 2\pi \int_0^R I(\tau, r)r\, dr$ within an aperture of a pinhole with a radius $R = 0.5\, \text{mm}$, aligned to transmit the central, most intense part of the beam, yields a pulse with a full width half maximum (FWHM) pulse width of 2.5 fs (the solid line in figure 7) carrying 15% of the field energy. Integration within an aperture of a pinhole with a radius $R = 0.3\, \text{mm}$ gives a 1 fs pulse (the dashed line in figure 7) that carries 7% of the field energy.

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Pulse-front distortions occur when ultrashort bursts of light propagate through lenses [29] or lens-based systems. Indeed, on the basis of a geometrical representation, a paraxial ray traversing a lens is temporally retarded with respect to a marginal one. This retardation depends solely on the geometrical characteristics of the lens and the dispersion of its material. When light pulses are considered, the above considerations translate into a variable group delay and therefore a distortion of the spatial pulse front, with the central parts to be delayed with respect to the marginal ones. In contrast, the nonlinear interaction of a few-cycle pulse with a gas results in distortions with an opposite sign. As can been seen in figure 6(d), the marginal parts of the pulse front are now delayed with respect to the center. Therefore propagation of the supercontinuum pulses through a lens provides a potential basis for dealing with such a distortion in an effective way without compromising its energy. To explore the potential of this idea, we have designed a meniscus lens and we have studied its capability to counteract the spatial pulse-front distortion of the supercontinuum beam emerging from the gas chamber. In particular, the spatial phase profile of the field, defined by figure 6(d), can be satisfactorily corrected by a silica meniscus with a curvature radius of 53.6 cm and an aperture of 3 mm. Along with the compensation of the group delay across the supercontinuum beam, such a meniscus will, of course, introduce an additional spatially nonuniform phase \( \theta(\omega, r) \) across the supercontinuum beam due to high-order dispersion of silica. As a result, the supercontinuum pulse transmitted through such a meniscus has a larger pulse width and a more complicated shape (the dotted line in figure 8) than a pulse with an ideally compensated, flat phase (figure 6(a) and the dashed line in figure 8). While it is quite difficult to completely compensate for the phase \( \theta(\omega, r) \), as it depends on \( r \), changing from the beam center to its periphery, the compressed pulse can be further shortened and its temporal aberrations can be reduced by compensating for only the spectral part of the phase \( \theta(\omega, r) \). In the inset to figure 8, we present the spatiotemporal map of the supercontinuum field transmitted through the meniscus assuming that high-order dispersion introduced by this component has been fully corrected at the center of the supercontinuum.
Figure 8. Light pulses produced through a compensation of the spatial chirp across the supercontinuum beam with the spatiotemporal field structure as defined by figure 6(d) using a meniscus with parameters as specified in the text: (solid line) pulses representing the field with the phase $\theta(\omega, r) - \theta(\omega, 0)$, corresponding to a partial compensation of high-order dispersion introduced by the meniscus, and (dotted line) pulses representing the field with the phase $\theta(\omega, r)$ with no compensation for high-order dispersion introduced by the meniscus. The dashed line shows supercontinuum pulses after an ideal spatial and spectral phase compensation to a perfectly flat phase. The inset presents the spatiotemporal map of the supercontinuum field transmitted through the meniscus assuming a partial compensation of high-order dispersion introduced by the meniscus with the residual phase given by $\theta(\omega, r) - \theta(\omega, 0)$.

With such a phase correction, the supercontinuum field can be compressed to a pulse width of about 1.8 fs (the solid line in figure 8). The peak intensity of this compressed pulse ($7 \times 10^{12}$ W cm$^{-2}$) is an order of magnitude higher than the peak intensity of the pulse at the output of the gas chamber before pulse compression ($6 \times 10^{11}$ W cm$^{-2}$). From the instrumental standpoint, this phase compensation procedure does not complicate an optical setup, as this operation can be combined with the compensation for the phase $\varphi(\omega, r)$, acquired by the supercontinuum field in the gas chamber, and can be implemented by a single accurately designed optical component.

Simulations of spatiotemporal dynamics of a few-cycle pulse in an ionizing gas presented in figure 5 clearly indicate significant plasma-induced defocusing on the trailing edge of the pulse—a phenomenon highlighted in an earlier work by Zaïr et al [30]. Moreover, the spatiotemporal structure of the field in our simulations is similar to the field structure reported in [30]. It is important to note, in this context, that the meniscus-lens compression technique presented here suggests an attractive alternative to the chirped-mirror compression technique beautifully demonstrated by Zaïr et al [30]. In particular, the presented meniscus-lens compression scheme allows the energy conversion from the input field to the compressed pulse to be improved relative to the chirp-mirror compression technique. In our simulations, the 2.5 fs
pulse compressed by the meniscus lens (the dotted line in figure 8) contains 60% of the input laser pulse energy.

Notably, many of the key tendencies in the evolution of a high-intensity ultrashort light pulse presented in this paper agree very well with the results reported in the existing literature (see [11, 12, 31, 32] for a review). In particular, it has been demonstrated in earlier work that single-cycle pulses can be generated at a specific location in a filament [31] and can be extracted by arresting filamentation after an appropriate propagation distance [33]. The spatiotemporal structure of few-cycle pulses simulated by Couairon et al [31, 33] is fully consistent with the results of our calculations, implying that the pulse compression technique using a properly designed meniscus lens can be considered as a versatile tool for the compensation of the space–time chirp of ultrashort pulses generated in the filamentation regime in various gases under different conditions. The meniscus-lens design presented in this paper is intended for the maximum temporal compression of supercontinuum pulses through chirp compensation and is not optimized for chirp tailoring aimed at efficient generation of isolated attosecond pulses by plasma-blue-shifted fields, as proposed by Couairon et al [23, 27].

6. Conclusion

Analysis of the evolution of submillijoule few-cycle light pulses in an ionizing gas presented in this paper reveals physical mechanisms and scenarios, enabling the generation of high-intensity supercontinuum radiation compressible to single-cycle pulse widths. We have shown that the enhancement of ionization-induced phase shifts and shock-wave effects for few-cycle fields is the key factor that substantially intensifies the high-frequency wing of the field spectrum, thus allowing the generation of broadband spectra with appropriate spectral phase profiles supporting compression to single-cycle pulse widths. The analysis presented in this paper suggests that compression to minimum pulse widths supported by the bandwidth of supercontinuum radiation is hindered by a spatially nonuniform group delay across the supercontinuum beam. A partial compensation of this spatial chirp with a suitably designed meniscus has been demonstrated, allowing efficient compression of supercontinuum radiation to single-cycle pulse widths along with suppression of temporal aberrations on a compressed pulse.

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