Supercontinuum generation of ultrashort laser pulses in air at different central wavelengths

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Abstract

Supercontinuum generation by femtosecond filaments in air is investigated for different laser wavelengths ranging from ultraviolet to infrared. Particular attention is paid on the role of third-harmonic generation and temporal steepening effects, which enlarge the blue part of the spectrum. A unidirectional pulse propagation model and nonlinear evolution equations are numerically integrated and their results are compared. Apart from the choice of the central wavelength, we emphasize the importance of the clamped intensity reached by self-guided pulses, together with their temporal duration and propagation length as key players acting on both supercontinuum generation of the pump wave and emergence of the third harmonic. Maximal broadening is observed for large wavelengths and long filamentation ranges.

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1. Introduction

Third-harmonic (TH) generation and supercontinuum (SC) emission are two phenomena which have attracted broad interest in the past years [1–5]. An evident reason is their direct application in atmospheric remote sensing measurements based on LIDAR (LIght Detection And Ranging) femtosecond laser setups [6]. In this context, spectral broadening originates from complex mechanisms that drive the long-range propagation of ultrashort pulses, when they form narrow filaments in optically-transparent media.

The physics of isolated femtosecond filaments in air is nowadays rather well understood (see, e.g., [7] and references therein). It involves the competition between Kerr self-focusing and plasma defocusing, triggered whenever the input pulse power exceeds the critical power for self-focusing \( P_c \approx \lambda_0^2/(2n_0n_2) \). Here, \( \lambda_0 \) is the central laser wavelength, \( n_0 = 1 \) and \( n_2 \) are the linear and nonlinear refraction indices in air, respectively. For high enough powers, multiple filaments nucleated after an early stage of modulational instability have also been widely investigated [7–9]. They produce spectral patterns mostly analogous to those generated by a single filament, as filamentary cells emerge in phase from the background field and possess the same phase link [10]. By comparing Terawatt (TW) multifilamented beams with Gigawatt (GW) single filaments in air, this property was again verified in the UV–visible region (230–500 nm), where femtosecond self-focusing pulses centered at 800 nm generically produce a tremendous plateau of wavelengths [11–13].

This latter phenomenon has recently become a subject of inspiration for several researchers. Two scenarios have been proposed for justifying the build-up of new wavelengths in the UV–visible range. On the one hand, temporal steepening phenomena undergone by the pump were
shown to deeply modify the filament spectrum [14]. Full chromatic dispersion included in the optical field wave number $k(\omega)$ affects both the diffraction operator and the nonlinearities. This induces shock-like dynamics at the back edge of the pulse through space–time focusing and self-steepening effects, which strongly “blueshift” the spectra. On the other hand, spectral broadening becomes enhanced by harmonic generation. The coupling of TH with an infrared (IR) pump produces a “two-colored” filament from pump intensities above 10 TW/cm² [15–17]. The amount of pump energy transferred into TH radiation not only depends on the pump intensity, but also on the focusing geometry and on the linear wave-vector mismatch parameter $\Delta k = [3k(\omega) - k(3\omega)]^{-1}$ fixing the coherence length $L_c = \pi/|\Delta k|$. In particular, the smaller the coherence length, the weaker TH fields. Along meter–range distances, the TH component can limit the clamped intensity of the pump wave within about 0.5% conversion efficiency [13]. Experimental and numerical data reported ring structures embarking most of the TH energy and having a half-divergence angle of about 0.5 mrad [18]. This process contributes to create a continuous spectral band of UV–visible wavelengths [11,13,19].

Resembling spectral dynamics have also been reported from 1-mJ infrared pulses propagating in argon at atmospheric pressure, after subsequent compression by chirped mirrors [20]. Simulations of these experiments [21], discarding TH emission, revealed that temporal gradients inherent to the steepening operators are sufficient to amplify UV shifts and cover the TH bandwidth down to 250 and 210 nm for initial pulse durations of 10 and 6 fs, respectively. Very recently, numerical simulations [22] refound the tendency for atmospheric propagation, i.e., TH generation, while it affects the pump dynamics to some extent over long ranges, does not change significantly SC spectra, whose variations are mostly induced by the fundamental field in air.

Despite these last results, we are still missing a detailed understanding of the key parameters which are supposed to drive SC generation. A first important parameter is, of course, the laser wavelength itself: How does the supercontinuum evolve when $\lambda_0$ is varied? This question was addressed in Ref. [11] for various laser wavelengths, at which some spectral components were seen to merge. However, the model used a two-envelope approximation (for the pump and TH fields, separately). As emphasized in [22], splitting into TH and SC pump within envelopes becomes problematic when their respective spectra overlap inside a wide frequency interval where the basic validity condition $\Delta \omega/\omega_0 \ll 1$ ($j = \omega, 3\omega$) may no longer be fulfilled. Actually, TH radiation produced through the nonlinear polarization needs to be described self-consistently from a single equation governing the total real optical field. This model was missing in Refs. [13,19], which made the role of TH and cross-phase modulation overestimated compared with the broadening of the pump in the UV–visible domain. Another important parameter is the length of the self-guiding range: Successive cycles of focusing and defocusing events promote the creation of shorter peaks in the pulse temporal profile and lead to a maximal extension of the spectrum. A third potential player is the input pulse duration. In [21], this was shown to affect the spectra in noble gases for pulses containing a few optical cycles mainly. Clearing this aspect requires several simulations using distinct pulse durations and exploiting different propagation ranges. In connection, we demonstrate that spectral enlargements are directly linked to the level of maximum (or clamped) intensity, $I_{\text{max}}$. Steepening operators as well as TH radiation broaden all the more the spectra as the intensity in the filament is high.

The paper is organized as follows: Section 2 presents the model equations, namely, a unidirectional propagation equation for the total electric field that generates higher-order harmonics (mostly TH) through Kerr nonlinearities. Results from this equation will be compared with those inferred from the “standard” nonlinear evolution equation (NEE) for the pump wave. The major difference between these two models lies in the production of the TH field and its coupling with the pump wave. Section 3 is devoted to the long-range propagation of 127-fs pulses in air described by the previous models. Emphasis is put on the influence of the central wavelength $\lambda_0$ (248, 800, 1550 nm). We discuss spectral modifications versus the height of the clamped intensity $I_{\text{max}}$, the input duration, together with the temporal steepening dynamics and merging between TH and pump spectral bands. Section 4 revisits SC for short-range (focused) propagations. It is shown that $I_{\text{max}}$ becomes closer to analytical evaluations when the beam develops few focusing/defocusing cycles. In this configuration, a lesser broadening may be achieved. Section 5 finally summarizes the generic features resulting from our analysis.

2. Models for pulse propagation and underlying physics

Our unidirectional pulse propagation equation (UPPE) assumes scalar and radially-symmetric approximations. It also supposes negligible backscattering. These hypotheses hold as long as the beam keeps transverse extensions larger than the central laser wavelength and as the nonlinear responses (together with their longitudinal variations) are small compared with the linear refraction index. Straightforward manipulations of Maxwell equations allow us to establish the equation for the spectral amplitude of the optical electric field in the forward direction as [7]

$$\hat{\varepsilon} \hat{E} = \frac{i}{2k(\omega)} \nabla_\perp^2 \hat{E} + ik(\omega) \hat{E} + \frac{i \mu_0 \omega^2}{2k(\omega)} \hat{F}_{\text{NL}},$$

where $\hat{E}(r,z,\omega) = (2\pi)^{-1} \int E(r,z,t)e^{i\omega t} \, dt$ is the Fourier transform of the forward electric field component, $z$ is the propagation variable, $\nabla_\perp^2 = r^{-1} \partial_r r \partial_r$ ($r = \sqrt{x^2 + y^2}$) is the diffraction operator, $\mu_0 = 1/c^2$, $k(\omega) = \sqrt{1 + \delta(\omega)/\omega_0^2}/c$ is the wavenumber of the optical field.
depending on the linear susceptibility tensor \( \chi^{(1)}(\omega) \) defined at frequency \( \omega \). In Eq. (1), \( \hat{P}_{NL} \equiv \hat{P}_0 + i \hat{j} / \omega \) is the Fourier transform of the nonlinearities that include the nonlinear optical polarization \( P_{NL} \) and the current density \( J \) created by charged particles. Eq. (1) restores the earlier UPPE formulation proposed by Kolesik et al. [23] in the limit \( k_{2}^{\perp} / k_{2}^{(\omega)} \ll 1 \) \( (k_{2}^{\perp} = k_{2}^{(\omega)} + k_{2}^{(\omega)}) \).

For practical use, it is convenient to introduce the complex version of the electric field

\[
E = \sqrt{c_1}(\vec{\varepsilon} + \vec{\varepsilon}^*), \quad \vec{\varepsilon} = \frac{1}{\sqrt{c_1}} \int \Theta(\omega) \hat{E} e^{-i\omega t} \, d\omega,
\]

where \( c_1 = \omega_0 e_0 / 2 k_0 \) employs the central wavenumber and frequency of the pump wave \((k_0 = n_0 \omega_0 / c)\) and \( \Theta(x) \) denotes the Heaviside function. Because \( \vec{\varepsilon} \) satisfies \( \vec{\varepsilon}^*(\omega) = \vec{\varepsilon}(-\omega)^* \) (\( * \) means complex conjugate), it is then sufficient to treat the UPPE model (1) in the frequency domain \( \omega > 0 \) only. The field intensity can be defined by \( E^2 \) averaged over an optical period at least. Expressed in \( \text{W/cm}^2 \), it is simply given by the classical relation \( I = |E|^2 \).

Concerning the nonlinearities, we assume a linearly polarized field. We consider a cubic susceptibility tensor \( \chi^{(3)} \) keeping a constant value around \( \omega_0 \), so that \( P_{NL} \) contains the instantaneous cubic polarization expressed as \( \chi^{(3)}(\vec{r}, t) = \epsilon_0 \chi^{(3)}(\vec{E}) \). In addition, the phenomenon of Raman scattering comes into play when the laser field interacts with anisotropic molecules, in which vibrational and rotational states are excited. Depending on the transition frequency \( \Omega_{13} \) between ground (level 1) and translational (level 3) states in three-level molecular systems and related dipole matrix element \( \mu [24] \), the Raman response takes the form

\[
P_{\text{Raman}} = \frac{2 \chi^{(1)}(\mu)}{\Omega_{13}^2 \hbar^2} \int_{-\infty}^{\infty} e^{it'} \sin \left( \frac{2\pi}{t_1} - t' \right) E^2(t') \, dt' \times E,
\]

where \( t_1 = 1 / \omega_R \) is the inverse of the fundamental rotational frequency and \( t_2 \) is the dipole dephasing time. Expressed in terms of the rescaled complex field \( \vec{\varepsilon} \) (Eq. (2)) and with appropriate normalizations [25], Eq. (3) completes the cubic polarization as

\[
P_{NL} = 2 n_0 n_2 \epsilon_0 \sqrt{c_1} \int_{-\infty}^{\infty} R(t - t') |\vec{\varepsilon}(t')|^2 \, dt' \vec{\varepsilon}
+ 2 n_0 n_2 \epsilon_0 \sqrt{c_1} (1 - x_K) \delta/(3 + c.c.),
\]

\[
R(t) = (1 - x_K) \delta(t) + x_K \theta(t) \delta(t),
\]

\[
h(t) = \frac{t_1}{\tau_1} + \frac{t_2}{\tau_2} \epsilon^{-t_2/\tau_2} \sin(t / \tau_1),
\]

where \( n_2 = 3 \chi^{(3)}(3) / (4 \epsilon_0^2 c_0^2 \epsilon_0) \) is the Kerr nonlinear index. Expression (4a) possesses both retarded and instantaneous components in the ratio \( x_K \). The instantaneous part \( \approx \delta(t) \) of Eq. (4b) describes the response from the bound electrons. The retarded part \( \approx h(t) \) accounts for the Raman contribution, in which fast oscillations in \( E^2 \) give negligible contributions, as \( \tau_1 \sim \tau_2 \sim 70 \text{ fs} \) are assumed to exceed the optical period \( \sim \omega_0^{-1} \).

When free electrons are created, they induce a current density \( J = q_e \rho v_c \), which depends on the electron charge \( q_e \), the electron density \( \rho \) and the electron velocity \( v_c \). \( J \) is computed from fluid equations involving external plasma sources and the electron collision frequency \( v_c \). At moderate intensities (<\( 10^{15} \text{ W/cm}^2 \)), the current density obeys

\[
\partial_t J + v_c \rho J = \frac{q_e^2}{m_e} E.
\]

Assuming electrons born at rest, the growth of the electron density is only governed by external source terms, i.e.,

\[
\partial_t \rho = W(I)(\rho - \rho_\text{sat}) + \frac{\sigma}{U_c} \rho I,
\]

that include photo-ionization processes with rate \( W(I) \) and collisional ionization with cross-section

\[
\sigma(\omega) = \frac{q_e^2}{m_e \epsilon_0 \epsilon_0 \epsilon_0 (1 + \omega^2 / \omega_c^2)}.
\]

Here, \( \rho_\text{sat} \) and \( U_c \) are the density of neutral species and the ionization potential, respectively. Electron recombination in gases is efficient over long (ns) time scales, and therefore we omit it. In Eq. (6), the rate for photo-ionization \( W(I) \) follows from the Perelomov, Popov and Terent’ev (PPT)’s theory [26] (see also Ref. [27] for more detail). Ionization rates stress two major limits bounded by the Keldysh parameter

\[
\gamma = \omega_0 \sqrt{2 m_e U_i / |q_e| E_p},
\]

namely, the limit for Multi-Photon Ionization (MPI, \( \gamma \gg 1 \)) concerned with rather low intensities and the tunnel limit (\( \gamma \ll 1 \)) concerned with high intensities, from which the Coulomb barrier becomes low enough to let the electron tunnel out. Here, \( E_p \) denotes the peak optical amplitude. For laser intensities \( I = |\vec{\varepsilon}|^2 \leq 10^{15} \text{ W/cm}^2 \), MPI characterized by the limit

\[
W(I) \sim W_{\text{MPI}} = \sigma_k \Gamma \sim K
\]
dominates, where \( K = \text{mod}(U_i / \hbar \omega_0) + 1 \) is the number of photons necessary to liberate one electron. The level of clamped intensity, \( I_{\text{max}} \), depends on the selected ionization rate.

Energy lost by the pulse through single ionization processes is determined by a local version of the Poynting theorem, yielding the loss current \( J_{\text{loss}} \), such that

\[
J_{\text{loss}} : E = U_i W(I)(\rho - \rho_\text{sat} - \rho). \text{As a result, our UPPE model reads in Fourier space as}
\]

\[
\frac{\partial}{\partial \omega} \vec{\varepsilon} = \left[ \frac{i}{2k(\omega)} \nabla_\omega + i k(\omega) \right] \vec{\varepsilon} + \frac{i \mu_0 \rho_0^2}{2k(\omega)\sqrt{c_1}} \theta(\omega) \hat{P}_{NL}
- \frac{ik(\omega) \theta(\omega)}{2 \epsilon(\omega_0) k(\omega)(1 + \frac{\gamma_0}{\omega_c})} \rho \frac{\rho_0}{\rho_c} - \frac{\theta(\omega)}{2} \sqrt{\epsilon(\omega)} \varphi(\omega),
\]

\[\text{NL = q_{|E|}}|E|^{2}\]
where \( \epsilon(\omega) \) is the dielectric function and
\[
\mathcal{L}(\omega) = \frac{U_i}{2\pi} \int \delta \left[ \frac{W(I)}{I} \left( \rho_m - \rho \right) + \frac{\sigma(\omega)}{U_i} \right] e^{i\omega t} dt.
\] (11)

In Eq. (10), \( P_{NL} \) and the expression containing the electric density \( \rho(\vec{x}, t) \) (Eq. (6)) must be transformed to Fourier space, from which we retain only positive frequencies for the symmetry reasons given above.

Alternatively, when a central frequency \( \omega_0 \) is imposed, Eq. (1) restitutes the Nonlinear Envelope Equation (NEE), earlier derived by Brabec and Krausz [28]. We can make use of the Taylor expansion
\[
k(\omega) = k_0 + k' \omega + \delta, \quad \delta \equiv \sum_{n=2}^{+\infty} \frac{k^{(n)}}{n!} \omega^n,
\] (12)
where \( \omega = \omega - \omega_0, k' = \partial k/\partial \omega|_{\omega=\omega_0}, k^{(n)} = \partial^n k/\partial \omega^n|_{\omega=\omega_0}, \) and take the inverse Fourier transform of Eq. (1) in which terms with \( k(\omega) \) in their denominator are expanded up to first order in \( \omega \). After introducing the complex-field representation \( \tilde{E} = U e^{i(k_0 \omega + \omega_0 t)} \), the new time variable \( t \to t - z/v_F \) can be utilized to replace the pulse into the frame moving with the group velocity \( v_F \). Further more assuming \( \omega^2/\omega_0^2 \ll 1, \sqrt{\epsilon(\omega_0)/\epsilon(\omega)} \approx 1 \) and ignoring the TH component, the nonlinear envelope equation for the forward pump envelope \( U \) expands as follows:
\[
\frac{\partial}{\partial z} U = \frac{i}{2k_0} T^{-1} \nabla_\perp^2 U + i \delta U + i \frac{\omega_0}{c} n_2 T \times [(1 - x_k)]|U|^2
\]
\[
+ x_k \int_{-\infty}^{t'} h(t - t')[U(t')|^2 dt' U - i \frac{k_0}{2n_0 \rho_e} T^{-1} \rho U
\]
\[
- \frac{\sigma}{2} \rho U - (\rho_m - \rho) \frac{U_W(I)}{2|U|^2} U,
\] (13)
where \( \sigma = \sigma(\omega_0), \delta \equiv \sum_{n=2}^{+\infty} (k^{(n)}/n!)(\omega_0)^n \) and \( T = (1 + z/v_F) \). The first term of the operator \( \delta \) corresponds to group-velocity dispersion with coefficient \( k' = \partial k/\partial \omega|_{\omega=\omega_0} \). Eq. (13) describes wave diffraction, Kerr focusing response, plasma generation, chromatic dispersion with self-consistent deviations from the classical slowly-varying envelope approximation through the space–time focusing and self-steeping operators \([T^{-1} \nabla_\perp^2 \delta]\) and \([T \delta^* \delta]\), respectively. This model will be integrated numerically by using initially Gaussian pulses
\[
U(x, y, z = 0, t) = \sqrt{\frac{2P_{in} e^{-z^2/\omega_0^2 - i k_0 x \sigma}}{\pi w_0^2}},
\] (14)
which involves the input power \( P_{in} \), the beam waist \( w_0 \) and \( 1/e^2 \) pulse half-width \( t_p \). Input pulses can be focused through a lens of focal length \( f \) and they linearly diffract over the distance
\[
z_f = (f^2 / \omega_0^2) / (1 + f^2 / \omega_0^2),
\] (15)
where \( \omega_0 = \pi w_0^2 / \lambda_0 \) is the Rayleigh range of the collimated beam \((f = +\infty)\).

In the coming analysis, we shall employ the nonlinear refractive indices \( n_2 = 8 \times 10^{-19}, 4 \times 10^{-19}, \) and \( 1 \times 10^{-19} \) cm²/W for the wavelengths \( \lambda_0 = 248, 800 \) and 1550 nm, respectively [29,30]. At 800 nm, we consider a fit ted MPI formulation for the ionization rate, \( W(I) = \sigma(\lambda) K^2 \), where \( K = 8 \) and \( \sigma(\lambda) = 2.88 \times 10^{-19} \) s cm²/W⁶. This approximation is known to reproduce experimental data at \( \lambda_0 = 800 \) nm rather faithfully, in particular \( I_{max} \approx 50 \) TW/cm² [27,31,32]. For the two other wavelengths, we lack well established formulations, so we employ PPT ionization rates. Because nitrogen molecules have an ionization potential \( U_i \) higher than that of O₂ molecules, we only consider the latter species as undergoing ionization with \( U_i = 12.1 \) eV and an effective residual charge \( Z_{eff} = 0.53 \) [33]. Nitrogen molecules are indeed expected to deliver much weaker electron densities, which was numerically verified at UV and IR wavelengths [34,35]. All ionization rates used in the present paper are illustrated in Fig. 1. They yield a clamped intensity below the threshold of 100 TW/cm² currently claimed in the literature [7,11]. The dispersion relation for air has been parameterized as in Ref. [36].

Since our outlook is to understand spectral variations versus the propagation dynamics at different wavelengths, we find it instructive to fix the same ratio of input power over critical, e.g., \( P_{in} = 4 \times P_{crit} \), at all wavelengths. To locate the Kerr-driven filamentation onset upon comparable \( z \) scales, we also adjust the ratio of the nonlinear focus length \( z_c \) and Rayleigh range \( z_0 \) between 2 and 4, by adapting suitably the beam waist \( w_0 \) between 1 and 4 mm.

### 2.1. Self-phase modulation and SC generation

In air, dispersion is weak with \( k'' \approx 1.2 \) fs²/cm in the UV domain and \( k'' \approx 0.2 \) fs²/cm in the mid-IR. Self-channeling then mainly relies on the dynamical balance between Kerr self-focusing and plasma defocusing, so that estimates for peak intensities \( (I_{max}) \), electron densities \( (\rho_{max}) \) and filament radius \( (L_{min}) \) can be deduced from equating diffraction, Kerr and ionization responses in Eq. (13). This yields the simple relations
\[
I_{max} \approx \frac{\rho_{max}}{2 \rho_e n_0 n_2}, \quad \rho_{max} \approx t_p \rho_{max} W(I_{max}),
\] (16a)
\[
L_{min} \approx \pi (2k_0^2 n_2 I_{max}/\rho_0)^{-1/2},
\] (16b)

![Fig. 1. Ionization rates for the different wavelengths used throughout this paper: \( \lambda_0 = 800 \) nm (solid line), \( \lambda_0 = 248 \) nm (dotted line), and \( \lambda_0 = 1550 \) nm (dashed line). The dashed-dotted line shows the overestimated ionization rate chosen in Section 3.2 for \( \lambda_0 = 1550 \) nm.](image)
where
\[
\tilde{n}_2 = n_2(1 - x_K) + n_2 x_{K\text{max}}, \int_{-\infty}^{t} h(t - t') e^{-\frac{2\pi}{\lambda}} dt',
\] (17)

represents the maximal effective Kerr index over the initial pulse profile. For practical use, \(W(I_{\text{max}})\) can be simplified to \(\sigma_{K} I_{\text{max}}^{2}\) in MPI-like formulation.

The magnitude of \(I_{\text{max}}\) directly impacts spectral broadening, which is basically driven by self-phase modulation (SPM). Because the frequency spectrum is expanded by the nonlinearity, SPM leads to SC, as the wave intensity strongly increases through the self-focusing process. Noting by \(\varphi(t)\) the phase of the field envelope, frequency variations are dictated in the limit \(T, T^{-1} \rightarrow 1\) by
\[
\Delta \omega = -\tilde{n}_2 \varphi \sim -k_0 \Delta z \tilde{n}_2 (\tilde{n}_2 I - \rho / 2 n_0 \rho_c),
\] (18)

which varies with the superimposed actions of the Kerr and plasma responses. Near the focus point \(z_c\), only the front edge of the pulse survives from this interplay and a redshift is enhanced by plasma generation. At later distances, second focusing/defocusing sequences attenuate this first tendency. In contrast, when accounting for temporal steepening \((T, T^{-1} \neq 1)\), shock edges in the back of the pulse are created and a “blue shoulder” appears in the spectrum, to the detriment of the early redshift [7,14,37,38].

In addition, the cubic polarization generates third-order harmonic, modeled by the last term of Eq. (4a). In self-focusing regimes, the third-harmonic intensity usually contributes by a little percentage to the overall beam fluence [16]. Despite its smallness, this component may act as a saturable nonlinearity for the carrier wave. It lowers the peak intensity of the pump and contributes to enhance the blue side of the spectrum after the TH and pump bandwidths increase and overlap [11,13].

3. Long-range propagation

We numerically analyze supercontinuum generation for the three laser wavelengths of 248 nm, 800 nm and 1550 nm. Results of the nonlinear Schrödinger-like equation (13) for the pump envelope involving or not space-time focusing and self-steepening operators are compared with those of the unidirectional propagation equation (10) avoiding any Taylor expansion in the dispersion relation.

Special attention is given here to the long-range propagation, which rather favors several cycles of focusing-defocusing events. Before proceeding with the above parameters specifically, we perform three different series of simulations showing SC at 800 nm, whose results are summarized in Fig. 2. The first one concerns direct integrations of Eq. (10); the second refers to the same pulse described by Eq. (13), which eludes TH production; the third approach relies on Eq. (13), in which temporal steepening is omitted, i.e., \(T = T^{-1} = 1\).

The insets in Fig. 2 show the on-axis spectra when SC is maximal. Following the UPPE description, TH, which emerges from \(z_c \sim 6\, \text{m}\), develops a limited redshift, whereas SC of the fundamental is widely extending towards the blue/UV wavelengths (Fig. 2a). Note that, although a broad plateau occurs in this domain, the TH bandwidth still appears separated from the pump spectrum. Following the NEE description, there is no TH generation. However, SC is so amplified in the blue region by temporal steepening effects, that it overlaps the TH zone and simply hides it (Fig. 2b). Finally, when neglecting temporal steepening, the pump instead develops a wide redshift (overestimated by plasma coupling) and a much narrower blueshift (Fig. 2c). A first observation can be drawn from Fig. 2: Since TH is responsible for lowering the clamped intensity of the pump [13], \(I_{\text{max}}\) reached in the UPPE model is lower and the frequency variations \(\Delta \omega \sim I_{\text{max}} \Delta z / \Delta t\) are diminished compared with NEE spectra for the pump wave alone.
Apart from this difference, no significant other change was detected between both these models, so that NEE seems to be nothing else but the UPPE description subtracted by the self-generated harmonics. This similarity was already reported in Ref. [7] and it was refound for the present laser parameters when we compared results from UPPE without harmonic generation and the NEE model (not shown here). Importantly, omitting temporal derivatives of the operators $T, T^{-1}$ imply more serious discrepancies, as can be seen from Fig. 2. By comparing Fig. 2b and c, it is clear that these operators are responsible for creating an impressive blueshift, which is not caused by the balance between Kerr and plasma responses.

3.1. Influence of $\lambda_0$

We now examine Gaussian pulses at different wavelengths (248, 800 and 1550 nm) with $\tau_p = 127$ fs and $P_{\text{in}}/P_{\text{cr}} = 4$ as initial conditions for the UPPE model in parallel geometry ($f = +\infty$). Figs. 2a and 3 show snapshots of spectra at maximal extent, together with associated peak intensities and electron densities. We here specify that no TH generation was included for $\lambda_0 = 248$ nm, because no reliable data of the dispersion relation was available for this wavelength. We believe, instead, that spectral components below 90 nm should be rapidly absorbed by the medium.

By comparing Figs. 2a and 3, it is seen right away that supercontinuum generation increases with the wavelength. To quantify this observation we introduce $\Delta \lambda_{\text{SC}}$ as the total extension of the on-axis spectra over wavelengths at $10^{-3}$ times the maximal spectral intensity. Then a measurement for the effective broadening is the ratio $\Delta \lambda_{\text{SC}}/\lambda_0$, which we find close to $0.5$ at 248 nm, $\sim 1$ at 800 nm and $\sim 1.5$ at 1550 nm. A look at the propagation dynamics reveals that the self-guiding range is noticeably augmented at longer wavelengths. This can be explained by the transverse size of the filament. Eq. (16b) gives an estimate for the beam waist in filamentation regime. If we assume comparable $I_{\text{max}}$ for all wavelengths, we deduce that the filament diameter at 1550 nm is about one order of magnitude larger than that at 248 nm, which is compatible with our numerical data. Hence, the larger the wavelength, the slower the filament is expected to diffract. Moreover, by virtue of the formula for the critical power $P_{\text{cr}} \propto \lambda_0^2/n_2$ and since a filament conveys a few $P_{\text{cr}}$ [7], it is obvious that IR filaments contain much more energy than their UV counterparts. Thus, nonlinear losses along the filamentation range are less dramatic in the IR domain. Due to the longer propagation range, more focusing/defocusing events participate in enlarging the spectra at longer wavelengths. We should also mention that coefficients in the temporal derivatives ($i\omega_0^3\partial_\zeta$) of the steepening operators increase with $\lambda_0$, which can contribute to enhance SC of the pump at large wavelengths. Fig. 4 details the evolution of the filaments in the plane $(t, z)$. It is seen that the time window in which the pulses disperse occupies the length of the input pulse duration. Although shorter temporal peaks arise through self-focusing/defocusing events, multi-peaked profiles mostly develop patterns having a whole extent close to $t_p$.

Before going on, we find it worth investigating SC at 1550 nm more thoroughly, in relationship with third-harmonic generation. Fig. 5 plots three of the SC development stages, first when TH and pump components are clearly separated $(z = 10 \text{ m})$, second when they start to merge $(z = 15 \text{ m})$. At the last propagation distance $(z = 25 \text{ m})$, we can observe that, unlike Fig. 2, TH and pump spectra overlap and make the TH bandwidth not distinguishable. For comparison, results from the NEE model for the pump wave alone have also been plotted.

A few features with the TH generation described by two coupled envelope equations as used in Ref. [11] can be retrieved at this stage. First, the two continua owing to the fundamental and TH bandwidths are separated by a minimum that seems all the deeper as the central wavelength is short. Over long enough propagation distances, this dip disappears for 1550 nm. Second, TH conversion efficiency can be enhanced at large wavelengths during the early focusing event (see, e.g., Fig. 5a). As noticed in [11], this property follows from the initial wave-vector mismatch which is considerably smaller at 1550 nm than at 800 nm. At further distances, differences with the numerical findings of this reference become more pronounced, mainly
because of the SC of the pump promoted by the steepening operators in the short wavelength domain.

Besides, Kolesik et al. [39,40] proposed that chromatic dispersion plays an important role in spectral broadening, especially on fixing the UV cut-off frequency. Actually, our simulations corroborate this statement, since dispersion becomes stronger at shorter wavelengths. For comparable clamped intensities, the spectral extent increases with $k_0$, while the absolute UV cut-off diminishes as the laser wavelength is decreased.

### 3.2. Influence of $I_{\text{max}}$

The impact of the clamped intensity onto SC is investigated by simply changing the ionization model: By decreasing the photo-ionization rate artificially it is possible to increase $I_{\text{max}}$ and the maximal plasma level $q_{\text{max}}$ accordingly. Reversely, increasing $W(I)$ reduces these two quantities, which can have a direct influence on the spectral broadening, as inferred from Eq. (18). To study this point, we concentrate on the wavelength of 1550 nm only, because it yields the broadest spectra explored so far. Fig. 6 shows the maximal intensity and peak electron density at this wavelength, when the ionization rate is artificially increased (see Fig. 1). All other parameters are unchanged, compared with the simulation shown in Fig. 3b. With the original ionization rate, $I_{\text{max}}$ reaches the value of 80 TW/cm$^2$; with the artificial one, $I_{\text{max}}$ stays below 13 TW/cm$^2$. The inset in Fig. 6 details the corresponding spectrum at $z_{\text{max}} = 40$ m. With low $I_{\text{max}}$, the TH component is reduced to some extent as the pump intensity barely exceeds the TH conversion threshold [15]. Meanwhile, SC of the pump driven by the $T, T^{-1}$ operators in the blue side decreases, i.e., a lower $I_{\text{max}}$ for analogous pulse compression implies less sharp temporal gradients and smoother optical shocks, which weakens blueshifted frequency variations. These features are visible in Fig. 6, where TH and pump broadbands remain separated. In Figs. 3b and 5b, in contrast, SC extends beyond the TH

![Fig. 4. Temporal evolutions of the pulses shown in Figs. 2a and 3 in the $(t, z)$ plane: (a) $\lambda_0 = 248$ nm; (b) $\lambda_0 = 800$ nm; (c) $\lambda_0 = 1550$ nm.](image)

![Fig. 5. On-axis spectra for the pulse used in Fig. 3b ($\lambda_0 = 1550$ nm) for the propagation distances (a) $z = 10$ m (solid line) and $z = 15$ m (dash-dotted line) and (b) $z = 25$ m (solid line). The dashed line in (b) refers to a spectrum at $z = 25$ m computed from the NEE model.](image)

![Fig. 6. Peak intensity (solid curve, left-hand side scale) and peak electron density (dashed curve, right-hand side scale) for the same pulse as in Fig. 3b ($\lambda_0 = 1550$ nm) computed from the UPPE model with an overestimated ionization rate (see Fig. 1). The inset shows maximal spectral broadening attained at $z_{\text{max}} = 40$ m.](image)
wavelength and increases more the pulse spectrum. Similar features were observed at the two other wavelengths, when the ionization rate was changed.

3.3. Influence of $t_p$

We investigate the influence of the initial pulse duration on the propagation dynamics and SC generation. Since we consider transform-limited pulses, the value of $t_p$ is directly linked to the initial spectral width. Moreover, $I_{\text{max}}$ comes into play in SC generation and is expected to scale as $\sim(1/t_p)^{1/4K-1}$ (see Eq. (16a)). Thus, the initial pulse duration should play a significant role in spectral broadening. To check this assessment, we performed several simulations using the UPPE model, by varying $t_p$ from 20 fs up to 500 fs. Because group-velocity dispersion becomes very efficient at short pulse durations and may even stop the Kerr self-focusing at powers too close to critical [41,42], we increased the input power up to $20P_{\text{cr}}$ for $t_p = 20$ fs. With this, we ensure to trigger a filamentation regime even for this short input duration.

To illustrate the dependency of $I_{\text{max}}$ upon $t_p$, short wavelengths are preferable because the number of photons for ionization is small. Fig. 7 shows maximum intensity, peak electron density and maximal spectral extent of a 20-fs pulse at 248 nm. At this wavelength, $K = 3$ and, following Eq. (16a), $I_{\text{max}}$ and $\rho_{\text{max}}$ should increase by a factor $\sim 2.5$ compared to the 127-fs pulse shown in Fig. 3a. Indeed, both quantities are increased by a factor of $\sim 2$ in the simulation. Spectral broadening is augmented from 0.5 to 0.8 in terms of $\Delta \lambda_{\text{SC}}/\lambda_0$, especially to the blue side. As explained in Section 3.2, this results from the action of the steepening operators. The overall propagation dynamics, characterized by the filamentation length and number of focusing/defocusing cycles are, however, comparable for both the 20-fs and the 127-fs pulses.

On the other hand, if we increase the initial duration $t_p$ towards the ps time scale, the propagation dynamics changes drastically. As an example, Fig. 8 shows the temporal evolution of a 500-fs pulse at 800 nm. Compared with Fig. 4b employing $t_p = 127$ fs, the obvious difference is the huge number of focusing/defocusing cycles. The action of the generated plasma breaks the pulse profile into a larger number of shorter peaks. With a longer pulse duration, more “time slices” are available for feeding successive focusing events. The filamentation range is increased and $I_{\text{max}}$ is maintained over several meters. Inspection of the simulations, however, reveals maximal spectral extent comparable with that displayed in Fig. 2a.

At 1550 nm the influence of $t_p$ on the maximal intensity is much less pronounced, since we have $K = 15$ (see Figs. 3b and 9). For all pulse durations, we indeed observe $I_{\text{max}} \sim 80$ TW/cm$^2$. This can explain why the maximal

Fig. 7. Peak intensity (solid curve, left-hand side scale) and peak electron density (dashed curve, right-hand side scale) of a 20-fs pulse with ratio of input power over critical equal to 20, $\lambda_0 = 248$ nm, $w_0 = 1$ mm. The inset shows on-axis spectra: the dotted curve represents the initial spectrum; the solid curve the spectrum at the distance $z_{\text{max}} = 3.5$ m where maximal broadening is observed.

Fig. 8. Temporal evolution of a 500-fs pulse at 800 nm with $4P_{\text{cr}}$ and waist $w_0 = 2$ mm in the $(t,z)$ plane.

Fig. 9. Peak intensities (solid curves, left-hand side scales) and peak electron densities (dashed curves, right-hand side scales) of 4 mm waisted pulses with different durations and ratios $P_{\text{in}}/P_{\text{cr}}$ at $\lambda_0 = 1550$ nm. The insets show on-axis spectra: dotted curves represent the initial spectra; the solid curves the spectra at the propagation distance $z_{\text{max}}$ where maximal broadening is observed: (a) $t_p = 20$ fs, $P_{\text{in}} = 15 \times P_{\text{cr}}$, $z_{\text{max}} = 20$ m; (b) $t_p = 500$ fs, $P_{\text{in}} = 4 \times P_{\text{cr}}$, $z_{\text{max}} = 40$ m.
are shown in Fig. 11 for line, parameters used in Fig. 3 b); in Fig. 9 b). The on-axis temporal profiles shown in Fig. 10 all exhibit rable, we might also find similar temporal patterns. Indeed, profiles upon propagation. If the spectral extent is compa-

tduration. So, there is no significant change in SC genera-
tion range, which increases with the initial pulse

ural pulse length $t_p$ just determines how many of these peaks appear, or, in other words, how many focusing/defocusing cycles the pulse is able to develop upon propagation. Another indication for the change in the effective pulse duration upon propagation is provided by the curve of the intensity maximum in Fig. 9b: The first focusing cycle is halted at slightly lower intensities $\sim 60$ TW/cm$^2$, because in this early stage the peak duration remains of the order of $t_p = 500$ fs. At later stages, $I_{\text{max}}$ increases as the pulse undergoes temporal compression.

4. Short-range propagation

So far, we have analyzed free propagation dynamics where long filaments achieve temporal gradients and SC extents similar to those produced by initially much shorter pulses. Now, we force all pulses to cover the same short fila-

lextents similar to those produced by initially much shorter pulses. Now, we force all pulses to cover the same short fila-

mentsation range mainly, which is consistent with the previous statements [19], based on a propagation model in the UPPE model). Thus, the initial pulse duration strongly influences spectral broadening in configurations of short filamentation range mainly, which is consistent with the numerical results of Ref. [21].

5. Conclusion

In summary, we have revisited recent works on SC generation versus third-harmonic emission, by showing from a complete UPPE model that spectral enlargements of femto-

second pulses in self-guiding regime are mostly driven by space–time focusing and self-steepening. TH generation, although changing the pump dynamics, affects the spectra to a limited extent only. This conclusion corrects some previous statements [19], based on a propagation model in
which temporal steepening terms were analyzed separately from an envelope description for TH generation. As it appears from our simulations, the TH component mainly broadens around its central wavelength, but additional broadening of the pump caused, e.g., by cross-phase modulation looks limited. Going one step beyond, we have demonstrated the important role of the clamped intensity in the frequency variations enlarging both TH and pump broadbands. The input pulse duration becomes a significant player in the spectral extents as long as pulses do not propagate too far, i.e., they do not let the temporal profiles of the pulse fluctuate so much that many peaks and sharp gradients can develop along the optical path. This property is mainly enlightened in short-range focused geometry and lost in long-range parallel geometry. Finally, the role of the central wavelength is preeminent: Our numerical simulations displayed evidence that SC clearly augments with the laser wavelength.

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