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ScienceDirect
Optics Communications 280 (2007) 173–182
OPTICS COMMUNICATIONS
www.elsevier.com/locate/optcom

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

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Received 5 March 2007; received in revised form 27 April 2007; accepted 13 June 2007

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.

PACS: 42.65.Tg; 52.38.Hb; 42.65.Jx; 42.68.Ay

Keywords: Supercontinuum generation; Femtosecond filaments; Nonlinear schrödinger equation; Self-phase modulation

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 $P_{\rm cr} \simeq \lambda_0^2/(2\pi n_0 n_2)$. Here, λ_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

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0030-4018/$ - see front matter © 2007 Published by Elsevier B.V.
doi:10.1016/j.optcom.2007.06.067

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/cm^2 [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 this 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 λ_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 $\Delta\omega_j/\omega_j \ll 1$ $(j = \omega, 3\omega)$ 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, $I_{\max}$: 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 λ_0 (248, 800, 1550 nm). We discuss spectral modifications versus the height of the clamped intensity $I_{\max}$, 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 $I_{\max}$ 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]

$$\partial_z \widehat{E} = \frac{\mathrm{i}}{2k(\omega)} \nabla_\perp^2 \widehat{E} + \mathrm{i}k(\omega)\widehat{E} + \frac{\mathrm{i}\mu_0\omega^2}{2k(\omega)} \widehat{F}_{\mathrm{NL}}, \quad (1)$$

where $\widehat{E}(r,z,\omega) = (2\pi)^{-1}\int E(r,z,t)\mathrm{e}^{\mathrm{i}\omega t}\,\mathrm{d}t$ is the Fourier transform of the forward electric field component, z is the propagation variable, $\nabla_\perp^2 = r^{-1}\partial_r r \partial_r$ $(r \equiv \sqrt{x^2+y^2})$ is the diffraction operator, $\mu_0\epsilon_0 = 1/c^2$, $k(\omega) = \sqrt{1+\chi^{(1)}(\omega)}\omega/c$ is the wavenumber of the optical field