Sub-2 fs pulses generated by self-channeling in the deep ultraviolet

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The generation of sub-2 fs light pulses in the UV is numerically demonstrated, using frequency conversion in filamentation regime. Few-cycle pulses emitted at 266 nm keep their temporal shape over several tens of centimeters. Self-compression results from the interplay between Kerr self-focusing and a low-density plasma, which continuously defocuses the pulse over extended propagation ranges. © 2008 Optical Society of America

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Ultrashort, deep-UV (200–300 nm) optical pulses are often required for time-domain studies in fundamental chemistry and biology [1]. A current method for generating such pulses relies on the frequency conversion of visible/infrared beams in atom gases. By exploiting the four-wave mixing (4WM) of the second harmonic (400 nm) with the fundamental mode (800 nm) of Ti:Sa laser sources (2 × 2ω−ω−3ω), Durfee and co-workers demonstrated efficient generation of 270 nm pulses in gas-filled capillaries, where phase matching was controlled by tuning the gas pressure [2]. Because self- and cross-phase modulations result in broad spectra, 8 fs UV pulses have been produced in argon [3]. Recently, Fuji et al. [4] used the same 4WM scheme in the self-guiding regime to achieve higher UV energies with an easier alignment of the pump in neon. Self-guiding results from the dynamical balance between the Kerr self-focusing of the two-color beam and the defocusing plasma created by ionization of the gas [5]. Interplay of these nonlinear effects enhances the interaction length and broadens the spectra [6], so that pulse widths of ~12 fs were reported [4]. In [3–5], the shortest pulse durations were obtained after compensating the output pulse phase through, e.g., grating compressors.

In the present work, we reexamine this scheme and numerically demonstrate that UV pulses can be produced while keeping FWHM durations down to 2 fs and below, stably over at least 30 cm. This is made possible by optimizing the very slow diffraction stage of a femtosecond filament, which continuously shrinks the pulse in time. Similar propagation regimes were recently tested with infrared beams [7]. In the deep UV, light structures with ~1 fs durations now become accessible along extended optical paths. They exhibit flat spectral phases and can be produced in various experimental setups.

Numerical simulations have been performed by means of two propagation models. To investigate the 400 nm pump pulse alone (Fig. 1), we employed a nonlinear envelope equation, which describes the forward pump component \( U(r, t, z) \) at frequency \( \omega_0 \) (see [8] for review):

\[
\frac{\partial}{\partial z} U = \frac{i}{2k_0} T^{-1} - \frac{1}{r} \frac{\partial}{\partial r} r \frac{\partial}{\partial r} U + iD_U + i \frac{\omega_0}{c} n_2 T|U|^2 U
\]

\[
- \frac{k_0}{2n_0^2 \rho_c} U + \frac{\sigma}{2} \rho U - (\rho_{at} - \rho) \frac{U_I W(I)}{2|U|^2} U,
\]

where \( z \) is the propagation distance and \( t \) the time in the frame moving at the group velocity \( k_0 \). The dispersion operator has the Fourier transform \( \hat{D} = k(\omega) - k_0 - (\omega - \omega_0) k_0' \), where \( k(\omega) \) is the wavenumber of the optical field. The operator \( T = (1 + i \omega_0 \frac{\partial}{\partial t}) \) stands for space-time focusing \((\sim T^{-1})\) and self-steepening \((\sim T)\); \( n_0, n_2 \) and \( \rho_c \) are linear refraction index, Kerr index, and critical plasma density, respectively. To describe beam couplings in large frequency windows (Figs. 2

![Fig. 1](image-url)

Fig. 1. (Color online) (a) Maximal intensity (solid curve) and peak electron density (dashed curve) of a 30 fs, 400 nm Gaussian pulse in argon at ~0.1 bar pressure. (b) On-axis temporal evolution versus propagation distance. (c) Same without space–time focusing, self-steepening, and dispersion effects. White arrows specify FWHM durations.
and 3), we instead used a unidirectional propagation model expressing the high-frequency field in the Fourier domain [9]:

\[ \hat{E} = \frac{i}{2k(\omega)} \nabla^2 \hat{E} + ik(\omega) \hat{E} + \frac{i\mu_0\omega^2}{2k(\omega)} \hat{J}_{NL}, \] (2)

such that \( E \sim U e^{i(k(\omega)z-i\omega t)} + c.c. \) and \( \hat{J}_{NL} = \hat{P}_{NL} + i\hat{J}/\omega \) contains the nonlinear polarization \( P_{NL} \) and current density \( J \) [8], corresponding to the last four terms of Eq. (1) for a single frequency \( \omega_0 \). The density of free electrons, \( \rho(r,z,t) \), is described by

\[ \partial_t \rho = W(I)(\rho_{nt} - \rho) + \frac{\sigma}{U_i} \rho I, \quad I = |U|^2. \] (3)

Similarly to [3], we consider the dispersion curve for argon established in [10] for the local pressure \( p = 100 \) mbar and select \( n_p = 2 \times 10^{-18} \) cm\(^{-2}\)W, inspired from [11]. Plasma sources are driven by the pump.

They include the photoionization rate \( W(I) \) derived in [12] for atoms; \( \rho_{nt} = 2.6 \times 10^{18} \) cm\(^{-3}\) and \( U_i = 16 \) eV are density of neutral species and Ar ionization potential. Collisonal ionization has the cross section \( \sigma = 2.4 \times 10^{-21} \) cm\(^2\). The incident beam is defined by \( \omega_0 = 0.8 \) mJ, \( w_0 = 150 \) μm in collimated geometry \( (f = + \infty) \). Selecting near-critical peak powers, \( P_{in}^{cr} = 1.62 P_{cr} \), yields an input intensity close to the ionization threshold, \( I_{th} \sim 80 \) TW/cm\(^2\) (\( E_{in}^{cr} = 0.8 \) mJ). Figure 1(a) shows that in this case the pulse intensity increases up to saturation, attained at the nonlinear focus \( z_c = 16.5 \) cm [13]. Figure 1(b) specifies that the early electron burst defocuses the front pulse. The rear pulse then slowly re-focuses, clamped with an almost flat electron density. Its temporal profile shrinks more and more along the \( z \) axis. The highest compression rate is not reached at maximal intensity but when the pulse slowly diffracts upon the next 50 cm. The plasma density, although reaching low levels, still competes with the Kerr response, which continuously shortens the on-axis pulse duration. To figure out the underlying compression mechanism, we performed simulations discarding steepening and dispersion effects. The pulse, when it undergoes only optical self-focusing versus plasma generation and related losses, develops similar dynamics, as illustrated in Fig. 1(c). Its back part reduces to a thin time slice propagating over \( \sim 1 \) m and behaves like a “gas-induced soliton” [14]. A smooth plasma defocusing depletes the pulse in time, whereas its transverse extent remains confined to \( \sim 100 \) μm diameters. In this simplified model, pulse shortening is overestimated, as the pulse can even reach subcycle durations. In the realistic model including gas dispersion and steepening effects, pulse compression is relaxed to some extent. Nevertheless, \( \sim 5 \) fs pulses can be produced at 400 nm with no external dispersion-compensation systems.

We now couple the 400 nm pump beam with a seed pulse at 800 nm (\( E_{in}^{cr} = 0.5 \) μJ). 4WM produces a UV pulse at 266 nm following the scheme \( 2\omega_0 - \omega_p = \omega_{4WM} \). The pump beam has an input waist increased to \( w_0 = 250 \) μm and power equal to five times critical (\( E_{in}^{cr} = 2.5 \) mJ). Figure 2(a) shows the same scenario as in Fig. 1(a). Plasma slowly turns off from \( z = 1.25 \) m to \( z = 2 \) m, along which it actively defocuses the pulse time slices. Figure 2(b) repeats the tempo-

![Figure 2](image-url)
eral evolution of Fig. 1(b), apart from interferences at \( \omega_0 - \omega_t \) between the pulse and seed pulses. Figure 2(c) shows the evolution of the pulse spectrum along the optical path. Near the first focus, upconversion takes place and creates the 4WM pulse at frequencies \( \omega_{4WM} = 7.1 \) THz. Higher-order frequencies are also produced around \( 2\omega_0 \) (\( \approx 9.4 \) THz) due to cascaded 4WM processes [4]. The spectral evolution is dictated by two different dynamics. The first one concerns the conversion process, for which new frequencies are generated upon coherence lengths \( L_{4WM} \approx 15 \) cm. The linear phase mismatch \( \Delta k_{lin} = k(2\omega_0 - \omega_p) + k_p - 2k_0 = 0.25 \) cm\(^{-1} \) and the one resulting from the interplay of nonlinearities, \( \Delta k_{NL} = 2 \omega_0 [n_J - \rho(I)/2n_0 \rho_c]/c \approx -0.019 \) cm\(^{-1} \), are both small owing to the low pressure, yielding the estimate \( L_{4WM} = \pi(\Delta k_{4WM} + \Delta k_{NL})^{-1} \approx 13 \) cm during early propagation. The second one is caused by the tremendous blueshift in the pump and 4WM pulses, which makes their spectra overlap at relatively high intensity levels. This spectacular supercontinuum is initiated by steepening effects for pump powers above critical [8]. Achieving the broadest spectrum allows the production of very short pulses. By selecting the frequency window around the 4WM component, i.e., \( 6 \leq \omega \leq 10 \) THz, sub-2 fs UV pulses can be generated, as demonstrated by Fig. 2(d). These optical structures, with FWHM durations comprised between 1.46 and 1.9 fs, hold over 30 cm as seen in Fig. 3(a). Figure 3(b) details two temporal profiles of the generated pulse along this stage; the inset specifies the corresponding spectral window. The 4WM peak can be isolated, and the surrounding spectral distribution is broad enough to produce optical structures with high compression rates. These keep a stable duration \( \approx 2.2 \) fs between \( z = 2 \) m and \( z = 2.4 \) m, i.e., over 40 cm of propagation. Plasma and intensity are maintained at sufficiently low levels to be detectable by direct inspection and imaging of the exiting pulses. Energy in the selected bandwidth is 1.55 \( \mu J \) at \( z = 2.4 \) m, and XFROG traces were again found to be flat, indicating unchirped phases.

In summary, we have demonstrated that deep UV pulses with sub-2 fs durations could be created by frequency conversion over several tens of centimeters in argon, within specific propagation regime. This regime is characterized by a slow deflection of the beam along which the interplay between the Kerr nonlinearity and the generated plasma, although of weak density, actively reduces the pulse extent in time. This property is generic and can be exploited both in parallel and focused geometries.

References