Wavelength scaling of optimal hollow-core fiber compressors in the single-cycle limit

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Abstract: We systematically investigate supercontinuum generation using three-dimensional numerical simulations of nonlinear femtosecond pulse propagation in hollow-core fibers (HCF) at different pump wavelengths ranging from 400 nm to 2 μ m. A general design strategy for HCF compressors is presented, maximizing the spectral broadening while preserving high beam quality for given pump pulse energy, duration and wavelength. We show close fitting of the modeled results with simple analytical formulas, enabling the construction of high-energy pulse compressors at the wavelength range of interest. Based on the presented wavelength scaling study, we propose an orthogonally polarized two-color pumping scheme in a single HCF compressor for the coherent synthesis of the electric fields in the sub-cycle regime with mJ level energies.

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1. Introduction

Optical waveforms at sub-optical-cycle timescale enable fully controlled manipulation of electron dynamics in chemical and atomic processes. Only coherent sources generating pulses with frequency spans over 2 octaves can achieve such control. To date, a number of sources have partially fulfilled such requirements using several novel methods, such as Er-fiber lasers and highly nonlinear fiber technology [1], Raman frequency shifting [2], wavelength multiplexing of optical parametric chirped pulse amplifiers [3], and a HCF compressor with distributed dispersion compensation [4]. Scaling this capability in both energy (beyond the mJ barrier) and spectral bandwidth is of fundamental importance for high-field physics research. Among these techniques, external pulse compression in a rare gas filled HCF pumped by a Ti:sapphire laser at a 800-nm wavelength has been the workhorse for high-energy few-cycle pulse generation and attosecond science. Here, we explore the optimal design criteria of a HCF compressor with different pump wavelengths to extend the capability of HCF compressor to state-of-art ultrafast lasers at various wavelengths. We also propose a HCF compressor design that produces multi-mJ sub-cycle optical pulses in a simple way.

In the HCF pulse compression technique, ultrafast pulses are coupled into a gas-filled HCF in which they undergo spectral broadening via self-phase modulation (SPM) while

preserving beam quality. Using a chirped mirror compressor or prisms, the SPM-induced chirp can be compensated, resulting in ultrafast laser pulses with durations approaching the few-cycle or even single-cycle limit. Durations as short as 5 fs with energies of 5 mJ and 9 fs with 10 mJ at 800 nm have been demonstrated using this compression method [4–8]. For the case of a typical HCF filled at constant gas pressure, the maximum pump pulse energy is limited by self-focusing and plasma formation due to strong ionization of the gas, resulting in significant loss in energy and beam quality. Considerable effort has been invested in improving the maximum pump pulse energy allowed, among them pressure gradient designs, and the use of circular polarization or chirped pump pulses have shown improved output in both quality and energy measures [9, 10], which enabled the use of multi-mJ laser sources.

Stabilization and control of the carrier-envelope phase (CEP) of few-cycle pulses is very important for applications to sub-cycle pulse synthesis as well as attosecond science. Since the CEP is well preserved in the SPM process inside a HCF compressor, CEP-stable few-cycle sources based on HCF [11] have been the most commonly used for the generation of isolated attosecond pulses via high-harmonic generation (HHG). Although most of the CEP stable few-cycle laser sources have been achieved using Ti:sapphire lasers at 800 nm, there is keen interest in generating high-energy CEP stable single-cycle pulses at wavelengths from the UV to the mid-IR. In particular, high-energy ultrafast laser sources in the IR region provide additional advantages when compared to near-IR sources because they extend the cutoff energy of high-harmonic photons, enabling the production of shorter attosecond pulses [12]. Pulse shortening of the CEP-stable mid-IR pulses generated from OPA/OPCPA using HCF compressors can be an efficient source for producing isolated attosecond pulses at photon energies up to the water-window region or even the keV range [13], opening the doorway to important applications in biological imaging and attosecond metrology with unprecedented accuracy [14].

In general, HCF compression has been carried out using a single pump carrier wavelength at visible and IR wavelengths [15–18], since the fiber design parameters typically depend strongly on wavelength and temporal characteristics of the laser source. Pumping with fundamental and second harmonic was studied by Matsubara *et al.* [19], where 3.6 μ J quasi-single cycle pulses of 2.6 fs in duration were presented. Optimization of the fiber parameters to maximize the pulse spectral broadening was reported by Vozzi *et al.* [20]. They discuss general criteria for the design of hollow fiber setups based on a numerical model, and also provide analytical formulas for the calculation of the spectral broadening and minimum fiber radius for the wavelength of 780 nm.

The necessity of optical sources at various wavelengths at the single-cycle limit has motivated the present systematic study on the wavelength scaling of optimal design of HCF compressors. In this paper, we develop and utilize a comprehensive numerical model to investigate the pump wavelength dependence for designing optimal HCF compressors. General criteria for choosing the fiber parameters, such as gas type, pressure, and radius are presented, maximizing the spectral broadening, fiber throughput, and output beam quality. We show that the performance of the HCF compressor can be accurately fitted to simple wavelength-dependent analytic formulas. We also show that for particular cases, the design parameters are similar even at distant pump wavelengths, allowing to utilize a single optimum HCF with pump pulses centered at fundamental and its second-harmonic wavelengths simultaneously, which could be utilized for sub-cycle waveform synthesis. The paper is organized as follows: in Section 2, we present the numerical model utilized for the nonlinear propagation along the fiber. In Section 3, we present numerical results for a variety of pump pulse characteristics, along with general design criteria for the optimization of spectral broadening and beam quality as a function of pump wavelength. We also identify the wavelength-dependent parameters of the fitted analytical formula in the range from 400 nm to 2 µm. In Section 4, we propose a design for producing multi-mJ sub-cycle pulses at 1.5 µm using a single HCF. And finally, Section 5 contains the conclusions and outlook.

2. Numerical model

The nonlinear spatio-temporal dynamics of the pulse propagating inside the HCF is modeled by a quasi-three dimensional generalized nonlinear Schrödinger equation (GNLSE) with radial symmetry. The equation is valid for pulses in the single-cycle regime as long as the wave propagates in one direction. The equation is generally written using a retarded time $\tau \equiv t - z/v_e$ as:

$$\frac{\partial A(r,z,\tau)}{\partial z} = \left(\hat{L} + \hat{N}\right) A(r,z,\tau) \tag{1}$$

where A is the complex envelope function with its amplitude normalized to the intensity I (i.e. $|A|^2 = I$). The operator \hat{L} accounts for the linear propagation effects, which are independent of pulse intensity including diffraction and dispersion, while the operator \hat{N} accounts for the intensity-dependent nonlinear effects including optical Kerr effect, plasma absorption, and plasma defocusing. A detailed description of the operators utilized in this simulator can be found in [21]. For hollow-core waveguides with a diameter that is much larger than the laser wavelength, the transverse laser fields are decomposed into EH_{1m} modes that propagate with different propagation constants $\beta_m(\omega)$ using the Hankel transform [22, 23]. It should be noted that the analytical expression of $\beta_m(\omega)$ takes into account the boundary conditions imposed by the HCF and non-modal dispersion of the filled gases based on the refractive index of the dielectric cladding and gas at specific temperature and pressure [24]. The plasma generation is described by the Ammosov-Delone-Krainov (ADK) formula [25], which gives a good quantitative description of the ionization process in the tunneling regime. In the following simulations the ADK formula is well suited to estimate the plasma density [26], because most of the cases studied here have Keldysh parameters ($\sqrt{I_p / 2U_p}$, where I_p is the ionization potential of the medium, and U_p is the average kinetic energy of the electrons in the laser field) smaller or around one, indicating a tunneling ionization regime. Also, at the intensities of interest, multiphoton ionization can be neglected. For Argon, the ionization potential I_p and nonlinear refractive index n_2 are 15.76 eV and $1.2 \times 10^{-23} \text{ m}^2/\text{W}$, respectively. To perform accurate simulations, self-steepening and space-time focusing effects are included in the model to correct for the frequency dependence of diffraction and nonlinear effects. The splitstep method takes care of the linear propagation operator \hat{L} and nonlinear propagation operator \hat{N} in different steps and the fourth-order Runge-Kutta method in the interaction picture (RK4IP) [27] is used for achieving an accuracy of $O(h^4)$ in the numerical integration. Since the linear step is often treated in the frequency-reciprocal-spacer domain while the nonlinear step is computed in the spatio-temporal domain, Fourier and Hankel transforms are used to convert the field back and forth between the different domains, respectively.

3. Simulation results

In our simulations, we optimized the main fiber parameters for mJ ultrafast laser systems operating at different wavelengths ranging from 400 nm to 2 μ m. In the following, we assume that the fiber length is L = 1 m, since having longer fibers with constant and uniform radius along the length is difficult to achieve and also makes alignment difficult. To compare the different possible designs of HCFs, we use a figure of merit, defined as:

$$FOM = \frac{E_{out}}{E_{in}} Q \frac{\Delta \tau_{SC}}{\Delta \tau_{TI}}$$
(2)

where E_{out} and E_{in} are the output and input pulse energies (throughput), Q is the relative energy in the fundamental mode compared to the total pulse energy (which will be described in more detail later) and $\Delta \tau_{TL}$ and $\Delta \tau_{SC}$ are the Fourier-transform limited pulse duration of the

output pulse and the single-cycle duration at the center wavelength respectively. In the following, we will discuss the designs that maximize the pump energy, broadening, and output beam quality, producing the highest FOM.

Since good focusing quality is beneficial for nonlinear processes such as HHG, minimization of the higher order modes is of fundamental importance. One of the possible approaches to maximize Q is to minimize the hollow fiber inner diameter, since the attenuation constant of higher order modes is inversely proportional to the fiber inner diameter. There is, however, a lower limit in the inner diameter, or core radius, since as the peak intensity is increased, ionization effects start to play an important role in the nonlinear interaction. In general, for proper operation of a HCF-compressor, it is required that the variation of the refractive index generated by the Kerr effect is much larger than the refractive index change produced by gas ionization ($\Delta n_{Kerr} > 10^3 \Delta n_{ioniz}$), setting a lower limit for the core radius [20].

The minimum core radius (a_{min}) , which is independent of gas pressure, can be well fitted by the expression:

$$a_{\min} = C(T_0 / 1.665)^{-\alpha} E_0^{\beta}$$
(3)

where $\alpha \sim 0.45$, $\beta \sim 0.51$ at 780 nm and C is a constant that depends on the gas, T₀ is the FWHM pump pulse duration, and E₀ is the input pulse energy. In the case of Ar, this constant is C_{Ar} ~4.69 × 10⁻⁹ m·s^{\alpha}J^{-\beta} in SI units. The parameters α and β depend on the pump center wavelength λ_0 .



Fig. 1. Calculated minimum fiber radius at different wavelengths for different pump pulse energies: (a) with the pulse duration of 30 fs and (b) 100 fs. Dashed lines are the fitted curves using the expression (3).

We calculated the minimum fiber diameter as a function of pump pulse duration, wavelength and energy using the numerical model described in Section 2. We chose Ar (1 atm) for the simulations in this research although the extension to other gases can be calculated simply using the factors shown in [20]. Figures 1(a) and (b) show the minimum HCF radius curves at the wavelengths of 400 nm, 800 nm, 1 μ m and 2 μ m as a function of pump pulse energy for 30 fs and 100 fs pulse durations. The dotted lines show the fitting of the retrieved values to the analytical formula for a_{min} . For the curve fitting, we calculated the wavelength dependency of $\alpha(\lambda)$ and $\beta(\lambda)$, which is shown in Fig. 2. It is clear from this analysis that the parameters $\alpha(\lambda)$ and $\beta(\lambda)$ have a strong dependence over wavelength for carriers below 1 μ m. However, in the region ranging from 1 μ m to 2 μ m, the curve exhibits a long plateau, which suggests that the same minimum fiber radius could be used for a wide range of pumping wavelengths in the IR.



Fig. 2. Retrieved parameters α and β as a function of wavelength.

Once the radius is set to the minimum value, it is still large enough to avoid ionization effects. We calculate the quality factor (Q) as a function of wavelength, pulse duration and pump pulse energy. Q and the broadening factor depend on gas pressure. We used a typical value for the pressure of 1 bar of Ar, which spectrally broadens the pump pulses close to one octave for most of the cases when using 30 fs pump pulses. An approximation for the spectral broadening can also be calculated using analytical formulas presented in [28]. The amount of energy in the fundamental mode, or quality factor Q, is calculated by decomposing the pulse envelope as a sum of leaky modes using a discrete Hankel transform. For the case of large fiber diameters compared to the driving wavelength, only linear combinations of Bessel functions of the first kind $J_{v}(r)$ are allowed in the core of the waveguide while the energy flow of the leaky modes into the cladding of the capillary is directed outward. For linear polarization, the modes that can be efficiently coupled to a laser beam are the EH_{1m} modes, with the mode EH_{11} having the lowest leakage, and thus we call it the fundamental mode. The mode profile inside the capillary can be approximated analytically. Actually, the intensity of EH_{1m} modes in the radial dimension can be expressed as $I_c(r) = I_{c0}J_0^2(\alpha_m r/a)$, where I_{c0} is the peak intensity, $\alpha_{\rm m}$ is the mth zero of the Bessel function, J_0 , and a is the core radius. Since single-mode operation is highly desirable for pulse compression, excellent mode discrimination is required. This goal is easily achieved in the input by adjusting the waist to 0.65^*a , with a coupling up to 98% to the EH₁₁ mode [29]. During the nonlinear propagation in the fiber, higher-order modes are excited through different nonlinear processes [30], while each mode has different attenuation coefficient depending on the relation between hollow fiber radius and wavelength [22]. We calculate the amount of energy in the fundamental mode using a quasi-discrete Hankel transform as follows. The electric-field amplitude can be decomposed as:

$$E(r,\lambda) = \sum_{m=1}^{\infty} c_m(\lambda) J_0\left(\alpha_m \frac{r}{a}\right)$$
(4)

where the coefficients are calculated from the inverse Hankel transform as:

$$c_m(\lambda) = \frac{1}{a^2 J_1^2(\alpha_m)} \int_0^a E(r,\lambda) J_0\left(\alpha_m \frac{r}{a}\right) r dr$$
(5)

Therefore, the relative energy in the fundamental mode to the total energy of the beam, i.e., the quality factor Q, can be calculated by:

$$Q = \frac{\int |c_1(\lambda)|^2 d\lambda}{\sum_{m=1}^{\infty} \int |c_m(\lambda)|^2 d\lambda}$$
(6)

Figure 3 shows the calculated quality factor Q as a function of pulse energy and wavelength for pump pulses of 30 fs (a) and 100 fs (b) when the core radius is set to a_{min} for each case. In general, high-quality operation of the HCF compressor is achieved when Q > 0.85, which limits the use of commercially available 30-fs chirped-pulse-amplification (CPA) Ti:sapphire lasers to less than 2 mJ (and to the μ J level for the second harmonic at 400 nm) if no further improvements, such as pressure gradient and laser chirp control [7], are implemented in the HCF system.



Fig. 3. Amount of energy in the fundamental mode (Q) as a function of pump pulse energy and center wavelength for (a) 30 fs pump pulses and (b) 100 fs pump pulses in Ar at 1 bar.

For pumping wavelengths ranging from 1 μ m to 2 μ m, it is possible to obtain high output beam quality for pump pulse energies of up to 5 mJ using a simple statically gas-filled HCF. As indicated by Fig. 3(b), by increasing the pump pulse duration to 100 fs, we can increase the pump energy limit by more than a factor of 2 while maintaining the high quality factor Qat virtually any wavelength from 400 nm to 2 μ m. This improvement occurs at the expense of spectral broadening, but it can be mitigated by increasing the gas pressure. Figure 4 shows the spectral intensity and phase of the HCF output pulses at the wavelengths of 400 nm, 800 nm, 1 μ m and 2 μ m for 30 fs and 100 fs pulses, and the corresponding transform-limited pulses achievable in each case, using a_{min} .

As shown in Fig. 4, for pulses with duration of 100 fs centered at 2 μ m the spectral broadening is mainly dominated by SPM, but as the center wavelength is decreased, the effect of self-steepening becomes more apparent. In the case of 800 nm and 400 nm pump pulses, blue-shifting and other non-linear effects start dominating the shape of the spectral intensity and phase, potentially making the output pulses difficult to compress using a standard chirped mirror or prism pair arrangement. For pulse durations of 100 fs the situation is clearly different. In the cases where SPM dominates the spectral broadening process, it is possible to calculate the broadening factor analytically using the approach described in [28]. The broadening factor can be optimized by increasing the fiber length or the gas pressure (which will eventually reduce Q) when the minimum core radius is used.



Fig. 4. Spectral response of the hollow core fiber, Fourier-transform limited pulses and spatiotemporal electric field distribution of the output pulses at different pumping wavelengths for the parameters: L = 1 m, Ar pressure = 1 atm, core radius set to minimum, for input pulses of 30 fs and 100 fs. (a) $\lambda_0 = 400 \text{ nm}$, $E_0 = 1 \text{ mJ}$, (b) $\lambda_0 = 800 \text{ nm}$, $E_0 = 3 \text{ mJ}$, (c) $\lambda_0 = 1 \mu \text{m}$, $E_0 = 5 \text{ mJ}$, (d) $\lambda_0 = 2 \mu \text{m}$, $E_0 = 5 \text{ mJ}$.

It is possible to increase the throughput at a particular pump energy level by chirping the pump pulses. Here, for simplicity, we did not include the laser chirp effect, but it was shown that it is possible to estimate the maximum pulse energy as the chirp is increased by using simple analytical formulas [6].

4. Hollow core fiber pumped at multiple wavelengths for generation of high energy 2octave spanning pulses

Complete tailoring of light fields on electronic timescales requires coherent superposition and manipulation of electromagnetic waveforms with over 2 octaves. Furthermore, energy scaling

is also of fundamental importance for effective control of the electronic processes in the strong-field physics regime. So far, the only energy-scalable coherent pulse synthesis was reported by Huang *et al.* in [3], which was based on coherent wavelength multiplexing of few-cycle OPCPA systems. Otherwise the performance of all other optical waveform synthesizers has been limited to the generation of super-octave μ J level pulses based on HCF compression. Here, we propose a relatively simple approach to the generation of 2-octave spanning pulses for multi-mJ sub-cycle waveform synthesis based on optimum two-color pump HCF compression in the IR, based on simulated results in the previous section.



Fig. 5. (a) Broadened spectrum and phase for fundamental wavelength ($\lambda_0 = 2 \mu m$) and its second harmonic ($\lambda_0 = 1 \mu m$) using the same 250 μm hollow core fiber. Temporal intensity distribution of the output pulses at (a) 1 μm and (b) 2 μm .

This particular approach requires a mJ, 30 fs pulse duration stable laser source at 2 μ m, similar to the one demonstrated in [30], although the concept can be equally implemented at other wavelengths of interest. We took advantage of the fact that at 1 mJ pump pulse energy, the minimum fiber radius at 1 and 2 μ m is very similar (around ~250 μ m). In the same way, the spectral broadening produced by both pulses approaches a full octave for the same gas pressure of 1 atm in Ar. Using orthogonal polarizations for fundamental and second harmonic allows us to manipulate both laser pulses independently when the low ionization level condition is met (for example by using a fiber radius larger than a_{min} at the shorter pumping wavelength). Also, an additional advantage is that a single beam pointing stabilizer can be used for optimizing the coupling into the fiber of both pumping arms. Efficient coupling of both fundamental and second harmonic can be achieved by using a trade-off focusing lens geometry. For example, since the beam waist w₀ is inversely proportional to the driving wavelength λ_0 for the same focusing lens, we can select a ratio for the fundamental of w₀/a ~0.9, which provides a coupling into the mode EH₁₁ of ~90% and the ratio for the second harmonic would be w₀/2a ~0.45, which provides a coupling of ~85% to the mode EH₁₁.

Even though the fiber is not optimum for the two pump pulses, the output beam quality Q for the 1 and 2 μ m beams is found to be 0.94 and 0.99, respectively. Moreover, the calculated total throughput of the fiber is 95% at 1 μ m and 88% at 2 μ m, while the output spectrum is broadened beyond 1 octave for both cases as can be seen in Fig. 5(a). The figure of merit of this HCF setup is FOM (1 μ m) = 0.89 and FOM (2 μ m) = 0.87, making this arrangement an ideal solution to the generation of multi-mJ 2-octave spanning laser pulses, as shown by Figs. 5(b) and (c). In our model, fluctuations of 10% in the input pulse energy were translated to small variations of the output beam quality below 0.1%, throughput variations below 1%, and fluctuations of ~8% for output pulse transform-limited durations.

It is possible to re-compress the output pulses by using two independent compressors at each carrier wavelength. In the case of the 2 μ m arm, compression can be achieved easily in bulk glasses such as a long suprasil block [31], while at 1 μ m a standard chirped mirror arrangement can be effectively used for compressing the pulse down to the few-cycle regime. Coherent combination of the two output pulses would lead to arbitrary electric waveform control over 2-octaves in a relatively simple way.

5. Conclusions

In conclusion, we used numerical simulations of the nonlinear pulse propagation in Ar-filled hollow core fibers to determine the optimum fiber parameters that maximize the output beam quality, broadening and energy. We have also presented a simple analytical model that allows to calculate the optimum fiber core radius for a pump wavelength between 400 nm and 2 μ m, with very good agreement with the model results. Moreover, the regimes where the HCF setup generates a high quality output have been calculated, setting a higher limit in the pump pulse energy across the spectrum when statically-filled HCF compressors are used. We have identified the optimum HCF parameters that enable the efficient use of multi-color pumping schemes. In particular, pumping with both fundamental and second harmonic at 2 μ m provides a number of advantages, such as high beam quality, throughput and spectral broadening beyond 2 octaves. The potential application of such supercontinua is the synthesis of high-energy electric fields in the sub-cycle regime.

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