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Generation of Few Cycle Femtosecond Pulses via Supercontinuum in a Gas-Filled Hollow-Core Fiber

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1. Introduction

The interaction of intense short nano- and picosecond laser pulses with plasma leads to reach variety of important applications, including time-resolved laser induced breakdown spectroscopy LIBS (Walid Tawfik et al., 2007), soft x-ray lasers (X. F. Li et al., 1989), and laser-driven accelerators (W. P. Leemans et al., 1989). In most cases the useful output - whether it be photons or accelerated particles - increases with the distance over which the laser-plasma interaction occurs, at least up to some upper limit which might, for example, be set by the onset of phase mismatching. The strength of any such interaction will depend on the intensity of the driving radiation, which for the short pulse lengths reached higher power densities and started the race for ever shorter laser pulses as a unique tool for time-resolved measurements of ultrashort processes (R. A. Cairns et al., 2009).

The progress in generating of femtosecond down to sub-10 fs optical pulses has opened a door for scientists with an essential tool in many ultrafast phenomena, such as femtochemistry (Zhong et al., 1998), high field physics (Li Y T et al., 2006) and high harmonic generation HHG (John et al., 2000). The advent of high-energy laser pulses with durations of few optical cycles provided scientists with very high electric fields, sufficient to suppress the Coulomb potential in atoms, accelerate electrons up to kilo-electron-volt energies, and produce coherent intense UV and extreme ultraviolet (XUV) radiation with durations of attoseconds (10\textsuperscript{-18} second) (Hentschel et al., 2001, Christov et al., 1997) which allow for the investigation of molecular dynamics with 100-as resolution (Baker et al., 2006). Ultrashort pulsed laser amplifiers can easily provide pulse energies in millijoule and even joule levels but with pulse durations longer than 20 fs. Amplification of pulses with shorter durations even to the millijoule level is a difficult task requiring elaborate setups (Ishii et al., 2005). Durations down to the few-cycle regime can be achieved through pulse compression schemes. Typically, such methods start in the picosecond or femtosecond region, i.e. already in the regime of ultrashort pulses. These methods can be grouped into two categories: linear pulse compression and nonlinear pulse compression (Shank et al., 1982).
In linear pulse compression; when pulses are chirped, their duration can be reduced by removing (or at least reducing) this chirp, i.e. by flattening the spectral phase (Treacy et al., 1969). Dechirping can be accomplished by sending the pulses through an optical element with a suitable amount of chromatic dispersion, such as a pair of diffraction gratings, a prism pair, an optical fiber, a chirped mirror, a chirped fiber Bragg grating or a volume Bragg grating (Martinez et al., 1984, Fork et al., 1984). The smallest possible pulse duration is then set by the optical bandwidth of the pulses, which is not modified by dispersive compression. In the ideal case, bandwidth-limited pulses are obtained. On the other hand, in the nonlinear pulse compression; in a first step, the optical bandwidth is increased, typically with a nonlinear interaction such as a supercontinuum generated by self-phase modulation (SPM) in nonlinear medium (Tomlinson et al., 1984). In most cases, this leads to chirped pulses, often with a longer duration which can be strongly reduced by linear compression (Fork et al., 1984).

The main challenge in pulse compression using supercontinuum generation lies in effective dispersion compensation and hence compression of the generated bandwidths to yield an isolated ultrashort optical pulse (Schenkel et al., 2003). In recent years significant progress has been made in the generation of supercontinuum in the visible and near-infrared spectral region from hollow and microstructured fibers. Recently, a high-energy supercontinuum extending a bandwidth exceeding 500 THz was generated with two gas-filled hollow fibers in a cascading configuration (Nisoli et al., 2002). The supercontinuum at the output of a single gas-filled hollow fiber has been compressed to 4.5 fs by use of a combination of chirped mirrors and thin prisms (Nisoli et al., 1997).

Hollow fiber pulse compression is currently the most widespread method for generating high-power, few-cycle pulses, with pulse durations as short as 2.8 fs and pulse energies up to 5.5 mJ being achieved in this manner (Nisoli et al., 1996), with the energy throughput limited by damage and plasma formation at the fiber entrance and self-focusing in the waveguide. In this technique, laser pulses are spectrally broadened in a gas contained within a long (~1 m, 50–500 μm inner diameter) hollow, glass fiber, that acts as a dielectric waveguide for the light, allowing significantly longer interaction lengths than possible in an unguided interaction. When an intense laser pulse propagates through a gas in the fiber it induces continuum generation due primarily to self-phase modulation (SPM). To maximize energy throughput, it is important to efficiently couple the pulse into the fiber. This is achieved by carefully matching the focal spot of the laser to the guided mode of the fiber. In a typical setup, the fiber is entirely contained within a single chamber which is statically filled (SF) with gas. In a differentially pumped (DP) fiber, gas is injected into the exit end of the fiber, and pumped away at the fiber entrance. The broadened pulses can be temporally compressed to durations shorter than the input pulses by using appropriate dispersive optical elements, e.g. prism compressors or chirped mirrors (Nisoli et al., 1996, Sartania et al., 1997).

In this chapter we represent a review on the generation of few-cycle fs light pulses using a gas-filled hollow-core fiber. Also, we report our experimental results of reaching 3.8-fs light pulses with energies up to 0.5 mJ using a laser of 0.9 mJ energy at repetition rate of 3 kHz and pulse duration of 28 fs. The pulse compressing achieved by the supercontinuum produced in neon gas-filled hollow fibers while the dispersion compensation is achieved by five pairs of chirped mirrors. In these experiments, we demonstrated the broadening variation with static pressures of neon gas. The applied technique allows for a straightforward tuning of the pulse duration via the gas. Since the focusing into the fiber is
no longer dependent on pressure and intensity, consistent coupling conditions are achieved for a broader range of pressures and input pulse parameters.

2. Supercontinuum generation and ultrashort pulses

Supercontinuum (SC) generation or white-light generation, is a complex physical phenomenon where the interplay between different nonlinear processes and dispersion leads to a significant spectral broadening of laser pulses propagating in a nonlinear medium. White light generation by laser radiation was first reported by Alfano and Shapiro who observed a spectral broadening of a 5 mJ picosecond second-harmonic output pulses of a Nd:YAG laser using a bulk of borosilicate glass (Gersten et al., 1980). The major nonlinear phenomena participating in SC generation are self-phase modulation (SPM), cross phase modulation (XPM), four wave mixing (FWM), intrapulse Raman scattering (IPRS) and soliton self-frequency shift (SSFS) (Alfano et al., 1987). If an intense laser pulse propagates through a medium, it changes the refractive index, which in turn changes the phase, amplitude, and frequency of the incident laser pulse. Changing the phase can cause a frequency sweep within the pulse envelope which called self-phase modulation (SPM).

Nondegenerate four-photon parametric generation (FPPG) usually occurs simultaneously with the SPM process. Photons at the laser frequency parametrically generate photons to be emitted at Stokes and anti-Stokes frequencies in an angular pattern due to the required phase-matching condition. When a coherent vibrational mode is excited by a laser, stimulated Raman scattering (SRS) occurs which is an important process that competes and couples with SPM. The interference between SRS and SPM causes a change in the emission spectrum resulting in stimulated Raman scattering cross-phase modulation (SRSXPM) (Gersten et al., 1980). A process similar to SRS-XPM occurs when an intense laser pulse propagates through a medium possessing a large second-order $\chi^2$ and third-order $\chi^3$ susceptibility as a nonlinear effect. Sometimes both of second harmonic generation SHG and SPM occur simultaneously and can be coupled together thus the interference between them alters the emission spectrum that is called second harmonic generation cross-phase modulation (SHG-XPM) (Alfano et al., 1987). When a weak pulse at a different frequency propagates through a disrupted medium whose index of refraction is changed by an intense laser pulse according to another nonlinear process called induced phase modulation (IPM) at which the phase of the weak optical field can be modulated by the time variation of the index of refraction originating from the primary intense pulse (Alfano et al., 1986). Recent development of ultra-short femtosecond lasers has opened the way to the investigation of ultrafast processes in many fields of science. An important milestone in the generation of femtosecond pulses was posed in 1981, with the development of the colliding pulse mode-locked (CPM) dye laser (Alfano et al., 2007). The first 27 fs pulses were generated in 1984 using a prism-controlled CPM laser (Agrawal, 1980). By using chirped mirrors to control the intracavity dispersion of Ti:sapphire oscillator; pulses as short as 7.5 fs have been generated (Fork et al., 2007) and by with the additional use of broadband semiconductor saturable absorber mirror; sub-6-fs pulses were generated (Valdmanis et al., 1985). In 1985, the first chirped-pulse amplification technique (CPA) was found which introduce amplification of ultrashort pulses as short as 20 fs to become available with extremely high power levels up to terawatt peak power at kilohertz rates (Donna Strickland et al., 1985, Xu et al., 1996, Sutter et al., 1999). Compression techniques represent an alternative way to generate ultrashort pulses. Nakatsuka and co-workers
introduced an optical compression technique based on the interplay between self-phase modulation (SPM) and group velocity dispersion (GVD) occurring in the propagation of short light pulses in single-mode optical fibers (Nakatsuoka et al., 2007). It is found that nonlinear propagation induces both of spectral broadening and chirping of the laser pulses then a subsequent propagation in an appropriate optical dispersive delay line can provides compression of the chirped pulse. Increasing the spectral bandwidth of the output pulse leads to the generation of a compressed pulse shorter in duration than the input one. Using this technique with a prism chirped-mirror Gires–Tournois interferometer compressor, 4.6 fs compressed pulses were obtained from a 13-fs cavity dumped Ti:sapphire laser but with a few with only nanojoules due to the low-intensity threshold for optical damaging of the single-mode optical fibers (Backus et al., 2007). By introducing a novel technique, based on spectral broadening in a hollow fiber filled with noble gases, the pulse compression extended to millijoule energy range which presents the advantage of a guiding element with a large diameter mode and of a fast nonlinear medium with high damage threshold (Nakatsuoka et al., 1997). By controlling of group-delay dispersion (GDD) in the compressor stage over an ultrabroadband (650–950 nm) spectral range, the hollow fiber technique could delivered a high-throughput up to 4.5-fs with multigigawatt (Nakatsuoka et al., 2007). Advances in the design of chirped multilayer coatings led to the adequate dispersion control without the need for prisms which opened the way to scaling sub-10-fs hollow fiber-based compressors to short pulses of 5 fs with substantially higher peak power up to 0.11 TW were generated at 1 kHz repetition rate (Nisoli et al., 1996).

3. Nonlinear pulse propagation in optical fibers
The Kerr-nonlinearity-induced intensity-dependent additive to the refractive index is one of the key physical factors in supercontinuum generation. This nonlinearity is due to the anharmonic motion of bound electrons under the influence of an applied field that causes also the induced polarization $P$ from the electric dipoles to be nonlinear with the electric field $E$, and follow the nonlinear relation (Shen et al., 1984)

$$P = \varepsilon_0 \left[ \chi^{(1)} E + \chi^{(2)} E^2 + \chi^{(3)} E^3 + ... \right]$$

where $\chi^{(i)}$ (i = 1, 2, ...) is susceptibility for the order the i-th and $\varepsilon_0$ is the vacuum permittivity. Here, the dominant contribution to $P$ is the linear susceptibility $\chi^{(1)}$ while the second-order susceptibility $\chi^{(2)}$ is responsible for nonlinear processes such as sum-frequency generation (SFG) and second-harmonic generation (SHG) (Shen et al., 1984). However, for media exhibiting an inversion symmetry at the molecular level, $\chi^{(2)}$ is zero. Therefore, optical fibers do not normally show second-order nonlinear effects since $\chi^{(2)}$ vanishes for silica glasses (SiO$_2$ is isotrop). So that the lowest order nonlinear effects in optical fibers originate from the third-order susceptibility $\chi^{(3)}$ which is responsible for phenomena such as third-harmonic generation (THG), four-wave mixing (FWM), and nonlinear refraction. However, the processes are not efficient in optical fibers unless special efforts are made to achieve phase matching. Therefore, most of the nonlinear effects in optical fibers originate from nonlinear refraction due to the intensity dependence of the refractive index resulting from the contribution of $\chi^{(3)}$. That means, the refractive index of a medium with a Kerr nonlinearity is written as (Shen et al., 1984)
where \( n_0 \) is the field-free nonperturbed refractive index of the medium, and \( I(t) \) is the laser radiation intensity, and \( n_2 = (2\pi/n_0)^2 \chi^{(3)}(\omega, \omega, -\omega) \) is the nonlinear refractive index at the frequency \( \omega, \chi^{(3)}(\omega, \omega, -\omega) \) is the third-order nonlinear-optical susceptibility.

For the case of short laser pulses, the intensity-dependent additive to the refractive index gives rise to a physically significant phase modulation of the laser field, the so-called self-phase modulation (SPM). We used formula (2) to represent the nonlinear phase incursion acquired by a laser pulse over a distance \( L \) in a medium with a Kerr nonlinearity in the form

\[
\Phi(t) = \frac{\omega}{c} n_2 I(t)L
\]

It can be seen from expression (3) that the intensity dependence of the refractive index of the medium maps the temporal profile of the field intensity in a laser pulse on the time dependence of the nonlinear phase shift, which, in turn, gives rise to a time-dependent frequency deviation across the laser pulse:

\[
\Delta \omega(t) = \frac{\omega}{c} n_2 L \frac{\partial I}{\partial t}
\]

The maximum SPM-induced spectral broadening of the laser pulse can then be estimated as

\[
\Delta \omega(t) = \frac{\omega}{c} n_2 L \frac{I_0}{\tau}
\]

where \( I_0 \) is the peak intensity of the laser pulse, and \( \tau \) is the pulse width. SPM is responsible for spectral broadening of ultrashort pulses and the existence of optical solitons in the anomalous-dispersion regime of fibers (Hasegawa et al., 1973). The third-order susceptibility controls the nonlinear effects which seem to be elastic in the sense that no energy is exchanged between the electromagnetic field and the dielectric medium. Another class of nonlinear effects results from stimulated inelastic scattering due to the optical field transfers part of its energy to the nonlinear medium.

Stimulated Brillouin scattering (SBS) and stimulated Raman scattering (SRS) represent two important nonlinear effects in optical fibers which are related to vibrational excitation modes of silica (Stolen et al., 1972, Ippen et al., 1972). The main difference between the two is that acoustic phonons participate in SBS while optical phonons participate in SRS. In a simple quantum-mechanical picture applicable to both SBS and SRS, a photon of the incident field (the pump) is annihilated to create a phonon with the right energy and momentum to conserve the energy and the momentum and a photon at the downshifted Stokes frequency. Of course, if a phonon of right energy and momentum is available it can creates a higher-energy photon at the so-called anti-Stokes frequency via the inverse process. The FWM generates an anti-Stokes photon, where two pump photons annihilate themselves to produce Stokes and anti-Stokes photons provided the conservation of the total momentum. The conservation of momentum requirement leads to a phase-matching condition, that must be satisfied for FWM to occur. In single-mode fibers, that phase-matching condition is not easily satisfied, thus the anti-Stokes wave is rarely observed during SRS. Even though SBS and SRS are very similar in their origin, different dispersion relations for optical and acoustic phonons lead to some basic differences between the two. A
fundamental difference is that in optical fibers SRS dominates in the forward direction whereas SBS occurs only in the backward direction. Although a complete description of SBS and SRS in optical fibers is quite involved, the initial growth of the Stokes wave can be described by a simple relation for SRS, as follows:

$$\frac{dI_s}{dx} = g_r I_p I_s$$ (6a)

where $g_r$ is the Raman-gain coefficient and $I_p$ is the pump intensity and $I_s$ is the Stokes intensity. Since the growth of the anti-Stokes wave is negligible, it is not discussed here. A similar relation holds for SBS as follows:

$$\frac{dI_s}{dx} = g_B I_p I_s$$ (6b)

where $g_B$ is the Brillouin-gain coefficient and $I_p$ is the pump intensity and $I_s$ is the Stokes intensity. The Raman-gain spectrum is measured to be very broad extending up to ~30 THz while the Brillouin-gain spectrum is extremely narrow with a bandwidth of only ~10 MHz (Stolen et al., 1972, Ippen et al., 1972). The maximum value of Brillouin-gain decreases by a factor of $\Delta v_p/\Delta v_B$ for a broad-bandwidth pump, where $\Delta v_p$ is the pump bandwidth and $\Delta v_B$ is the Brillouin-gain bandwidth. So that for short pump pulses (< 50 ns), SBS is negligible. An important feature for both SBS and SRS is that they exhibit a threshold-like behavior. The later means that a significant conversion of pump energy to Stokes energy occurs only when the pump intensity exceeds a certain threshold level. For example, for SRS the threshold pump intensity typically is ~ 10 MW/cm$^2$ (Hasegawa et al., 1973).

4. Supercontinuum generation in hollow fibers

Ultrashort pulses can be obtained not only by linear pulse compression, but also by nonlinear pulse compression techniques as described earlier. In nonlinear pulse compression, the pulses are spectrally broadened by propagation through a suitable nonlinear medium and subsequently compressed in a dispersive delay line. In 1981, a new method based on spectral broadening by SPM was introduced for optical pulse compression. With this technique 6 fs pulses at 620 nm have been generated due to the propagation of short pulses in single-mode optical fibers and by using a prism-pair for external dispersion compensation (Nakatsuka et al., 1981). Furthermore, a 4.5 fs pulses at 800 nm have been achieved using an improved ultrabroad-band dispersion compensation scheme broadened by propagation through a suitable nonlinear medium (Baltuska et al., 1997). However, the use of single-mode optical fibers limits the input pulse energy to a few nanojoules range. Therefore, the need for new spectral broadening techniques was born with the availability of high-energy (mJ) femtosecond pulses from solid-state laser amplifiers. One possibility is to achieve spectral broadening in bulk materials but due to the very short interaction length, limited by Rayleigh lengths, high intensities are needed to achieve the necessary nonlinearity for spectral broadening (Rolland et al., 1988). These high intensities can lead to damage and spatial beam quality problems due to multiphoton ionization. In 1996, another particularly suitable technique for high-energy ultrashort pulses was introduced. This technique depends on spectral broadening by SPM in a hollow cylindrical fused silica fiber filled with a noble gas under constant pressure (Nisoli et al., 1996). That technique enables pulses shorter than 5 fs to be generated at multigigawatt peak powers (Nisoli et al., 1996).
Hollow fiber is suitable for large pulse energies since it provides a guiding element with a large-diameter single mode. Furthermore, the use of noble gases as nonlinear medium offers several important advantages compared to optical fibers.

Light propagation in a hollow waveguide is a well-studied topic, which was developed when long-distance communication in standard optical fibers was still inaccessible (Marcatili et al., 1964). Electromagnetic radiation propagates in hollow fibers by grazing incidence reflections; only leaky modes are supported because of power losses through the fiber walls. Three propagation modes can be excited: transverse circular electric (TM$_{pm}$) modes, with the electric field directed radially; TE$_{pm}$ modes, in which the electric field lines are transverse concentric circles and centered on the propagation axis; hybrid modes (EH$_{pm}$, with $p \geq 1$) at which all field components are present, but axial components are so small that such modes can be thought as transverse. For fiber diameters sufficiently larger than the optical wavelength, EH$_{1m}$ modes appear linearly polarized and can be efficiently coupled to a laser beam. The radial intensity profile of EH$_{1m}$ modes is given by $I_r(r) = I_0 j_0^2(v_m r/a)$, where $j_0$ is the zero-order Bessel function, $a$ is the capillary radius, $v_m$ is the $m$th zero of $j_0(r)$, $I_0$ is the peak intensity. The complex propagation constant $\beta(\omega)$ of the EH$_{1m}$ mode is given by (Shen et al., 1984)

$$\beta(\omega) = \frac{\omega \eta(\omega)}{c} \left[ 1 - \frac{1}{2} \left( \frac{v_m c}{\omega \eta(\omega) a} \right)^2 \right] + \frac{i}{a} \left( \frac{v_m c}{\omega \eta(\omega)} \right)^2 \frac{\omega^2(\omega)+1}{\sqrt{\omega^2(\omega)-1}}$$

where $\eta(\omega)$ is the refractive index of the gas, $\omega$ is the laser frequency, and $v(\omega)$ is the ratio between the refractive indexes of the external (fused silica) and internal (gas) media. The refractive index $\eta(\omega)$ can be calculated at standard conditions by tabulated dispersion relations then the actual refractive index can be easily determined in the operating conditions used for pulse compression (Lehmeier et al., 1996). Many modes can be excited when the laser beam is injected into the capillary. However single-mode operation is generally required for pulse compression and mode selection must be exists. This aim can be achieved by optimal coupling between the input laser beam and the fundamental fiber mode EH$_{11}$. By assuming a Gaussian linearly polarized input beam, it is possible to determine the equation for the coupling efficiency between the input beam and the capillary modes. Propagation along hollow fibers can be occurred at grazing incidence reflections of the dielectric inner surface. This grazing angle reduces the losses caused by these multiple reflections by suppresses the higher order modes. Thus, only the fundamental mode can propagate, in a sufficiently long fiber (Lehmeier et al., 1996). On the other hand, with the proper mode matching for an optimum value of $w_0/a \approx 0.65$, where $w_0$ is the spot size at the fiber entrance and $a$ is the capillary radius, the coupling efficiency of the EH$_{11}$ mode with the laser beam is $\sim 98\%$, while higher-order modes show a value lower than $0.5\%$. Thus, the incident intensity profile as a function of the radial coordinate $r$ is given by

$$I_0(r) = I_0 j_0^2 (2.405 \ r/a)$$

where $I_0$ is the peak intensity and $j_0$ is the zero-order Bessel function (Marcatili et al., 1984). It is worth pointing out that even if higher-order modes were excited, mode discrimination would be achieved anyway, owing to the higher loss rate of EH$_{1m}$ with respect to fundamental mode. Mode discrimination in the capillary allows one to perform a spatial filtering of the input beam. By applying the same mode on the complex propagation
constant $\beta(\omega)$, $\beta$ the real phase constant of Eq. (8), and imaginary, $\alpha/2$ (field attenuation constant), parts of the propagation constant are given by:

$$\beta = \frac{2\pi}{\lambda} \left[ 1 - \frac{1}{2} \left( \frac{2.405 \lambda}{2 \pi \alpha} \right)^2 \right]$$  \hspace{1cm} (9a)

$$\frac{\alpha}{2} = \left( \frac{2.405 \lambda}{2 \pi \alpha} \right)^2 \frac{\lambda^2}{2 \alpha} \frac{\omega^2 + 1}{\omega^2 - 1}$$  \hspace{1cm} (9b)

where $\lambda$ is the laser wavelength in the gas medium. Assuming Gaussian pulse profile and neglecting dispersion and self-focusing, the maximum broadening spectrum after propagating a length of $l$ can be written as

$$\delta \omega_{\text{max}} = 0.86 \int_0^l \gamma(z) P_0 \xi e^{-az} dz / T_0$$  \hspace{1cm} (10)

where $z$ is propagating distance, $\alpha$ is given by Eq. (9b), $P_0$ is the peak power; $T_0$ is the half-width (at the 1/e intensity point) of the pulse; $\gamma$ is the nonlinear coefficient and is given by $\gamma = n_2 p(z) \omega_0/c A_{\text{eff}}$ [$n_2$ is given by Eq. (2)], where $n_2$ is the nonlinear index coefficient, $\omega_0$ is the laser central frequency; $c$ is the light speed in vacuum, $\xi$ is the coupling efficiency, $A_{\text{eff}}$ is the effective mode area (Nisoli et al., 1996). In the statically gas-filled case, the pressure is constant along the fiber. While in the differentially pumped case, the pressure is chosen to be a minimum (0 bar) at the entrance and gradually increases along the fiber. This leads to the pressure distribution

$$p(z) = \left[ p_0^2 + \left( \frac{z}{\xi} \right)^2 (p_L^2 - p_0^2) \right]^{1/2}$$  \hspace{1cm} (11)

where $p_0$ and $p_L$ are the pressure at the entrance and the exit, respectively. Then the bandwidth broadened in both the cases can be expressed as

$$\Delta \omega_{SF} = 0.86 \omega_0 n_2 p_L P_0 \xi (1 - e^{-az}) / c T_0 A_{\text{eff}} ,$$  \hspace{1cm} (12)

$$\Delta \omega_{DP} = 0.86 \omega_0 n_2 p_L P_0 \int_0^z \sqrt{\frac{x}{\xi} e^{-ax}} A_{\text{eff}} dx .$$  \hspace{1cm} (13)

So that the bandwidth broadening depends on both the input laser characteristics and the gas used in the hollow fiber. The nonlinear coefficient $\gamma$ takes the values $7.4 \times 10^{-25}$ m$^2$/W bar, $9.8 \times 10^{-24}$ m$^2$/W bar, and $2.78 \times 10^{-23}$ m$^2$/W bar for neon, argon, and krypton respectively (Nisoli et al., 1997, Robinson et al., 2006). For short pulses (20-fs), it is found that a much larger spectral broadening is obtained than long pulses (140-fs). This is because at shorter pulse duration, gas dispersion, in addition to SPM, also plays an important role during pulse propagation while for longer pulses (140 fs) a purely SPM-broadened highly modulated spectrum is observed (Nisoli et al., 1996). The higher order nonlinear effect of Self-steepening has to be taken into account as well for the short input pulses. This higher order nonlinear effect is due to the intensity dependence of the group velocity and leads to an asymmetry in the SPM-broadened spectra with a larger broadening on the blue side (blue-shift) (Baltuska et al., 1997).

In the following, we reviewed recent results of some working groups in that field; In 2006 Joseph Robinson et al., group used 700-µJ, 30-fs laser pulses with a 50-nm bandwidth centered at 800 nm and 1-kHz repetition rate 15 mm diameter to generate SC for
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Few cycle generation (Robinson et al., 2006). The beam was focused into a 1-m-long hollow fused-silica fiber and reach intensity of about $2 \times 10^{14}$ Wcm$^{-2}$. The fiber is mounted inside a larger fused-silica support capillary, with an outer diameter of 6 mm and filled with neon pressures of 0–3 bar, in both the SF and DP cases. They found that the transmission remains constant with increasing pressure for the DP case, in contrast to the SF case, which exhibits a sharp drop in transmission with increasing pressure. They conclude that performance is due to defocusing at the fiber entrance, which causes an increase in the focal spot size and beam aberrations and consequently a reduced coupling efficiency. In these experiments, they observed a spectra broaden in both cases up to a pressure of 1.5 bar, with the DP bandwidth lagging that of the SF case, due to the reduced average gas density for the DP case. At higher pressures, while the DP spectra continue to broaden, the SF spectra begin to reduce in bandwidth due to the reduced energy throughput at these pressures. Finally, a Spectral broadening of $\Delta \omega = 7 \times 10^{14}$ rad s$^{-1}$ with an energy throughput of 40% and corresponding output pulse energies of 290 pJ and subsequent compression to 6.5 fs has been achieved in the DP fiber filled with neon at 3 bar.

In 2008 Katsumi Midorikawa et al., group used 800 nm, 5 mJ, 25-fs laser pulses 1-kHz repetition rate to generate SC for few cycle generation. The beam was focused into a 220 cm-long hollow fused-silica fiber and reach intensity of about $2.4 \times 10^{14}$ Wcm$^{-2}$. The fiber is mounted inside a larger fused-silica support capillary, and filled with neon in gradient pressures of 0 – 1.6 bar (Samuel et al., 1996). They observed the spectral broadening for various neon pressures on the exit side as shown in Fig. 1(a) (Fig. 1(a), (b) is reproduced with permission from Akira Suda (Samuel et al., 1996)). Then they used the same parameters as those used in the experiments to make simulations for these broadenings based on a method described elsewhere (Nurhuda et al., 2003) in order to understand the mechanism driving the spectral broadening as shown in fig.1 (b). A good agreement was found between the experimental and the theoretical simulations. By investigating the simulation data in details they recognized that the spectral broadening is predominantly driven by the self-phase modulation. Moreover, they concluded that the pressure gradient method prevents the beam from collapsing due to self-focusing as well as from plasma defocusing, while allowing the pulse to undergo self-phase modulation during its passage through the hollow fiber. However, the spectral broadening of more than 300 nm was obtained at a pressure of 1.6 atm. Under these conditions, they could obtain 5 fs laser pulse after compression.

In 2009 Chang H. Nam et al., group used a Ti:sapphire, 5 mJ, 29-fs laser pulses 1-kHz repetition rate to generate transform-limited 3.5 fs pulse (Park et al., 2009). The beam was focused into a 100 cm-long hollow fused-silica fiber. The fiber is mounted inside a larger fused-silica support capillary, and filled with neon in gradient pressures of 0 – 2.0 bar. They observed the spectral broadening for various neon pressures on the exit side as shown in Fig. 2 (Fig. 2, (3) are reproduced with permission from Chang H. Nam (Park et al., 2009)). The output beam from the pulse compressor was recollimated by a concave silver mirror, and two sets of chirped mirrors. The first pair of the chirped mirrors has GDD of - 80 fs$^2$ and TOD of +150 fs$^3$ (510 nm to 920 nm), and the second pair has -90 fs$^2$ and +1270 fs$^3$, respectively, (600 nm to 1000 nm). They used SHG FROG (second-harmonic generation frequency-resolved optical-gating) for temporal characterization of the compressed output pulses. They found that the shortest pulse measured was 4.1 fs at neon pressure of 1.6 bar and also at 1.8 bar. For pressures in excess of 1.8 bar, the pulse duration increased due to the high-order chirp induced by self-focusing and ionization. The FROG measurement showed...
that the GDD increase with neon pressure was much larger than that theoretically estimated for the increase of neon pressure. In these studies they also studied the effect of laser chirp on spectral broadening. That was done by changing the grating distance in the pulse compressor of the kHz CPA Ti: Sapphire laser as shown in Fig. 3 where the output spectra changed very delicately to laser chirp. Their obtained results showed that the spectral broadening was sensitive to the direction of the laser chirp. With negatively chirped pulses,
spectral broadening was reduced, while the broadening was effective with positively chirped pulses. Furthermore, they observed 3.4-fs pulses in the transform-limited case using positively chirped 31-fs pulses, while the negatively chirped 30-fs pulses resulted in a longer transform-limited duration of 4.4 fs. They found that while the compressor cell was evacuated, the energy transmission was 62% (3.1 mJ), but it decreased to 28% (1.4 mJ) with 1.6-bar neon.

In 2010, Wei Zhi-Yi group compared the spectral broadening from statically neon-filled and differentially pumped hollow fibers (Wei Zhang et al., 2010). They used 800 nm Ti: sapphire 25fs 800 μl laser which is focused into a fused silica 1 m hollow fiber filled with neon. The spectral broadening was obtained at different pressures, as shown by the spectra in Fig. 4 (Fig. (4),(5) are reproduced with permission from Wei Zhi-Yi (Wei Zhang et al., 2010)). They found that for the statically filled case, the spectrum broadens obviously as the gas pressure increases, even at very low pressure. For the optimum pressure of 1.5 bars, the broadening bandwidth covers the range from 460nm to 930nm with transmission efficiency of 50%. However, at pressure 2.0 bars they observed a narrow bandwidth with lower transmission and significant spot splitting. They concluded that this was due the energy loss due to ionization and defocusing at the fiber entrance at very high pressure. For the differentially pumped case, it was noticed that the bandwidth is almost unchanged at the pressure lower than 1 bar. However, the spectrum continuously broadens with nearly constant transmission efficiency with the increase of the pressure. The pressure was limited by the windows pressure maximum capacity at 2.5 bars. At the later pressure, the broadest spectrum was obtained in region from 420 nm to 960 nm with transmission efficiency of 68.8% for 0.55 mJ pulse energy. In this case, both of the bandwidth and the transmission efficiency are much higher than the statically filled case. These results show the obvious advantages of the differentially pumped hollow fiber, which supports 3.3 fs transform limited pulses with higher transmission efficiency. Based on a set of negative dispersion chirped mirrors and _ne adjustment of the small angle wedges, laser pulses as short as 4.4 fs are obtained, as shown in Fig. 3. It corresponds to only 1.6 cycles at the central wavelength. The compressed pulse duration obtained is 1.3 times the Fourier transform limitation; the reason is that the higher order dispersions cannot be compensated perfectly.

In general, the same equations used to describe pulse propagation in optical fibers can be used for gas-filled hollow fibers. Both of the dispersion and the weights relative of SPM can be evaluated using characteristic parameters such as the dispersion length $L_D$, and the nonlinear length $L_{NL}$ where

$$L_{NL} = 1/(\gamma P_0)$$

$$L_D = T_0^2 / |\beta_2|$$

where $P_0$ is the peak power of the pulse and $T_0$ is the half-width (at the 1/e-intensity point) of the pulse and $\beta_2 = \frac{d^2n}{d\omega^2}$ is the GVD (Group-velocity dispersion) of the fiber filled with gas.

The fiber length L is a very important factor to affecting the nonlinear processes that if L exceeds the dispersion length $L_D$ and the nonlinear length $L_{NL}$, both of SPM and dispersion will play an important role in pulse propagation through the fiber. The optimum fiber length $L_{opt}$ for best pulse compression, i.e. for interplay between GVD and SPM for the generation of linearly chirped pulses, is approximated by $L_{opt} = (6 L_{NL} L_D)^2$ (Tomlinson et al., 1984). For the case of higher pulse energies there are two considerations set the limit
Fig. 3 (a) Measured (black solid) and retrieved (blue dotted) spectra and spectral phase of the laser output obtained with 1.6-bar neon and positively chirped 33-fs laser pulse and (b) temporal profile of the laser pulse measured using the SHG FROG method. The inset shows the FROG trace.

Fig. 4. Spectral broadening in the hollow fiber for the statically gas-filled (a)–(d) and differentially pumped (e)–(h) cases as a function of gas pressure. The dashed lines show the spectrum of the input pulse.
when scaling this approach to supercontinuum generation. First, the input laser peak power must be smaller than the $P_c$ critical power for self-focusing $P_c = \lambda^2/2\pi n_2$ (for a Gaussian beam) which constrains the type of noble gas used and its pressure. Second, to avoid ionization, the input laser peak intensity should be smaller than the multiphoton ionization threshold which constrains the hollow fiber diameter and the type of gas used. The second constraint can be reduced by using shorter pulses, since the threshold for multiphoton ionization increases with decreasing pulse duration.

In the following Sections, we will demonstrate our experimental findings for the generation of few-cycle pulses using supercontinuum generation in hollow fiber filled with neon gas.

Fig. 5. Calculated and measured bandwidths broadening at the full-width-half-maximum (FWHM) for both the differentially pumped (upper panel) and statically gas filled (lower panel) cases.
5. Experimental setup

Figure 6 the experimental setup used for SC generation and pulse compression. A typical chirped-pulse amplifier (CPA) based on a commercial multipass Ti:sapphire system (Femtopower Pro, Femtolasers GmbH) delivers pulses 900-μJ, 28-fs laser pulses with a 55-nm bandwidth centered at 800 nm and 3-kHz repetition rate (Rolland et al., 1988, Nisoli et al. 1997, Cerullo et al. 2000, Marcatili et al. 1964). A Ti:sapphire oscillator (Rainbow; Femtolasers GmbH) produces sub-10 fs seed pulses with 2.4 nJ energy at 76 MHz. The oscillator was pumped by a continuous wave cw diode-pumped frequency doubled Nd:YVO4 laser (Coherent Verdi) at 532 nm and 3.10 watt. The oscillator output pulses are then stretched by 30 mm of SF57-glass before coupling them into the amplifier.

A nine-pass amplifier with three-mirror configuration was used to boost the pulse energy to the millijoule level. This amplifier consisted of a pair of dielectric-coated spherical mirrors (R/C=800, 500 cm) and a flat silver mirror overcoated with MgF2. A 5-mm-long Brewster-cut 0.25% titanium-doped-sapphire crystal system was placed at the confocal position of the two spherical mirrors and was pumped by 527 nm, 25.6 watt Nd:YLF laser (Quantronix 527DQ). About 90% of the focused pump total energy was absorbed by the Ti:sapphire crystal. To reduce thermal lensing effects, the crystal was cooled to -100 °C by using immersion cooling. After the fourth pass through the crystal the beam is coupled out by mirror and fed into the single pulse selecting system. This system consists of a Pockels cell between two crossed polarizers, a Berek compensator and an acousto-optic programmable dispersive filter (DAZZLER) (FASTLITE), which improves the pre pulse...
suppression ratio of the single pulse selection, provides fine dispersion control, and preventing gain narrowing before coupling back to the amplifier. After amplified in another four passes, the pulse passes through the 8th-pass periscope followed by a telescope which adapts the beam for efficient gain saturation across the entire pumped volume for the last two passes through the crystal. The beam is finally picked out of the multipass system by the 9th-pass PICK-OFF prism. The output of the amplifier was more than 1.1 mJ with hundreds of picosecond duration and 3 kHz repetition rate. After amplification, the beam sent into a grating compressor. The compressor consists of two high-efficiency transmission gratings and a vertical retro-reflector to get 23 fs with about 80% throughput efficiency of 0.9 mJ.

The beam from this laser system was focused into a fused silica hollow fiber with a length of 1 m (250 μm inner diameter, and 750 μm outer diameter) by a lens with 1.5 m focal length. The focusing was chosen to be 65% with respect to the hollow fiber diameter to satisfy the maximum coupling criterion, resulting in an intensity of 2×10¹⁴ Wcm⁻². The fiber was kept on a V-groove and then placed inside a tube chamber made of stainless steel with Brewster-cut input and output windows (0.5-mm-thick fused silica), making small enclosures at both ends of the hollow fiber. Stainless steel tubes are attached to both ends of the support capillary using o-ring seals. A CCD camera was used to image the entrance of the fiber, enabling qualitative measurement of the focal spot size. Pulse energy and spectral broadening were measured at the output of the fiber for neon pressures of 0–3 bar, using static pressure. The vacuum chamber was filled with neon with pressure of about 2 bars. Initially, the chamber is vacuumed to 2 × 10⁻² torr using a mechanical vacuum pump. Furthermore, the beam from the hollow fiber was collimated using a silver concave mirror with curvature radius of 4 m. The dispersion induced by gas in the hollow fiber, air and optical components such as lens, two windows, was compensated for by a set of ten broadband chirped mirrors and a pair of fused silica wedges. Each pair of chirped mirrors had group delay dispersion (GDD) of $-40 \text{ fs}^2$ for the wavelength range from 510 nm to 920 nm. The compressed pulse duration is measured with an autocorrelator.

6. Results and discussions

The result of supercontinuum relies on nonlinear phase shift due to self-phase modulation and self-steepening, it is crucial to optimize the dispersion of the input laser pulse for obtaining broadband supercontinuum so that the laser pulse maintains high peak power and steep temporal shape. Dispersion controlled supercontinuum spectra are presented at Fig. 7(a) and their 2D spectrogram at Fig. 7(b).

To control the dispersion of the input pulse, group delay dispersion (GDD) was adjusted by the DAZZLER for the range from -200 to +160 fs². It’s clearly shown that the dependency of supercontinuum bandwidth on the dispersion of the laser pulse exists. The optimal GDD is about -50 fs², which indicates this initial negative chirp compensates the positive dispersion from both the focusing lens and hollow fiber. The maximum bandwidth covers single octave from 450 nm to 950 nm wavelength. The transform limited pulse duration of 2.9 fs FWHM can be achieved using this spectrum as shown in Fig. 7(c). Output pulse energy is about 0.5 mJ and no significant dependence on the dispersion is observed.

Not only the dispersion of the pulse affects the spectral broadening but also the pressure of noble gas plays an important role on supercontinuum generation. In general, by increasing the pressure of Ne gas in hollow fiber, more spectral broadening occurs due to the
Fig. 7. (a) super-continuum spectra vs group delay dispersion (GDD) (b) super-continuum spectrogram; red:intense, blue:no-light.
increasing nonlinear susceptibility $\chi$. Fig. 8 shows the measured spectra of supercontinuum for various Ne pressures from 0.0 bar to 2.6 bar. Below the pressure of 2 bar, the spectral bandwidth increases with the increasing pressure of Ne gas, while the intensity of supercontinuum decreases over the pressure of 2 bar due to the absorption in the gas medium. One of the bandwidth limitations of supercontinuum from hollow fiber is energy lose due to leaky mode from mode matching condition for marginal wavelength. For fixed core diameter, only single wavelength satisfies the EH$_{11}$ mode matching condition which allows the best coupling efficiency while other wavelength components suffer energy loss during propagation. So as shown in Fig. 8, the spectral bandwidth does not really cover shorter than 400 nm and longer than 1000 nm wavelength. To overcome this limitation one should consider about multiple stages of supercontinuum generation in parallel or in serial with different parameters such as length and core diameter of hollow fiber, type of gas and gas pressure. Indeed, the serial supercontinuum generation has been successfully demonstrated to enhance the supercontinuum bandwidth (Schenkel et al., 2003), and parallel supercontinuum generation also can be implemented for multi-channel waveform synthesis (Goulielmakis et al., 2007).

The supercontinuum output is sent to compressor which consists of 5 pair of double-angle chirped mirrors for residual dispersion compensation (Pervak et al., 2007). The chirped mirror is originally designed for the wavelength range from 450 to 900 nm and reflectivity drops below 400 nm while reflectivity for other wavelength is above 98%. So after compressor the pulse energy drops to 0.4 mJ depending on how much spectrum is lost below 450 nm.

![Image](https://www.intechopen.com)
Compressed pulse duration of 3.8 fs is measured by using second harmonic autocorrelator and the autocorrelation trace is shown in Fig. 9 (a). Precise chirp compensation can be made by either adjusting wedge thickness or slightly changing the pressure of Ne gas which introduces positive material dispersion. The pulse duration for various gas pressures is shown in Figure 9 (b). The shortest pulse duration is achieved as 3.7 fs at 2.4 bar pressure.

Fig. 8. Supercontinuum spectra vs Ne pressure (0 bar ~ 2.6 bar).

Fig. 9. (a) the measured pulse duration of 3.8 fs.
7. Conclusion

Compression of high-energy pulses down to the few cycles regime is a well-established technology nowadays, essentially based on the hollow fiber technique. Using this tool, significant successes have been performed in ultrafast physics and nonlinear optics, in particular in the field of attosecond pulses. Nonlinear-optical interactions of ultrashort laser pulses can efficiently generate an artificial white light with a controlled and short pulse duration, unique spectral properties, and a high spectral brightness. We have reviewed the basic theory of supercontinuum generation especially in nonlinear hollow fiber optics. In that review, we demonstrated a comparison between the spectral broadening in both SF and DF using hollow fiber filled with gas (usually neon). Also, we have showed our experimental results for the generation of few cycle pulses using different static gas pressures. We successfully observed 3.8 fs, 0.5 mJ laser pulses at 3 kHz with high through output.

In future prospective, ultrashort pulses are expected to lead to breakthroughs in communication and optical computing. Optical computing based on ultrafast logic units has the potential for revolutionizing the field of computers. On the other hand, using SPM more spectrally wider supercontinuum pulses could be achieved and hence compression down to 1 fs and possibly even into attosecond regions may be possible over the next decade using uv pulses.
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