# Generation of high energy, 30 fs pulses at 527 nm by hollow-fiber compression technique

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**Abstract:** The compression of 300-fs-long, chirp-free laser pulses at 527 nm down to 30 fs is reported. The laser pulses, originated from a frequency-doubled, mode-locked Nd:glass laser, were compressed by a 0.7-m-long, 150-μm-bore-diameter, argon-filled hollow fiber, and a pair of SF10 prisms with a final energy of 160 μJ. These are the shortest, high energy pulses ever produced by direct pulse compression at the central wavelength of 527 nm. The spectral broadening of the pulses propagating inside the hollow fiber was experimentally examined for various filling-gas pressures and input pulse energies. The spectral width of the pulses was broadened up to 25 nm, and 27 nm for argon- and krypton-filled hollow fiber, respectively, at a gas pressure lower than 2 bar. The physical limitations of the hollow-fiber pulse compression technique applied in the visible range are also studied.



**OCIS codes:** (320.5520) Pulse compression, (320.7110) Ultrafast nonlinear optics

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## 1 . Introduction

The hollow fiber pulse compression technique was used to generate high energy femtosecond pulses [1-4]. In this technique, the optical pulse spectrum is broadened inside a gas-filled hollow fiber by self-phase modulation (SPM) or by filamentation. Filamentation is a highly non-linear process which involves several effects such as group velocity dispersion, self-phase modulation, and self-steeping [5]. Typically, a filament is formed in a gas medium for laser pulse energy in the millijoule range, and a pulse duration in the order of several tens of fs [6], so that the peak power of the laser pulses in the gas medium exceeds a critical value for onset of both self-focusing and multi-photon ionization.

Optical pulse compression requires two essential steps: nonlinear spectral broadening, and dispersion compensation. Dispersion compensation can be achieved in many ways, among which mostly utilized are prism pairs or chirped mirrors. A prism pair is used to compensate the second-order dispersion, but it needs to be combined with grating-pairs, or spatial light modulators, if compensation of third or higher order dispersions is required [7]. Chirped mirrors with low loss are flexible for dispersion compensation. They have been used to generate high energy pulses as short as 5 fs or less [8] in the near-infrared region, at the 780 nm wavelength of the Ti:sapphire laser source. Nevertheless, while chirped mirrors are expensive to be designed and manufactured at shorter wavelengths, such as 527 nm, many research activities in physics, chemistry, bioengineering, and medicine need ultrashort laser pulses in the visible (see for instance [9]), or UV spectral region (see, for instance, [10]). To produce such intense fs pulses in the visible or UV domain , Ti: sapphire chirped-pulse amplification systems , which are equipped with frequency doubling or tripling stages, are widely used, although they are very expensive and complicated. Another quite recent ultrashort-pulsed laser system is constituted of a mode-locked Nd:glass laser source with chirped-pulse amplification, which is subsequently frequency-doubled and compressed [11, 12], representing a new high energy ultrashort pulse source.

In a recent paper [13] some of the authors reported the first application of the hollow fiber compression technique to laser pulses in the visible region, and pulses 40 fs long with energy of approximately 150 μJ, were achieved. In this paper, we report further improvements of the technique leading to even shorter, compressed, laser pulses. We also discuss the physical and practical limitations of the hollow fiber pulse compression technique, which can be of general interest when developing and using such relatively simple, compact, and versatile source of ultrashort laser pulses. It is worth stressing that the final peak power of the compressed pulses is as high as several gigawatts, which allows the achievement of peak intensities higher than $10^{14}$ W cm$^{-2}$, under typical focusing conditions.

The experimental setup description is followed by the analysis of the spectral broadening of the pulses propagating through an argon-filled hollow fiber. The spectral width of the pulses versus initial pulse energy and filling-gas pressure is then analyzed, and the optimal conditions for pulse compression are presented. Finally, we extensively discuss the physical and practical limitations of this technique in terms of the theory of light propagation in hollow fibers.

## 2. Experimental results and discussion

The scheme of our experimental setup is depicted in Fig. 1. Optical pulses emitted from a mode-locked Nd:glass laser ($\lambda = 1055$ nm), with chirped pulse amplification, are frequency-

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doubled (λ =527 nm), and temporally compressed down to ≈ 300 fs through a type-II phase-matching KDP crystal [12]. The frequency-doubled and compressed pulses are linearly polarized, and characterized by a well-behaved, Gaussian spatial fundamental mode ($M^2 \leq 1.3$), with pulse energy up to 1.1 mJ, and a repetition rate of 33 Hz.

A 30-cm-focal-length lens is used to focus the above pulses into a fused-silica, hollow fiber which is placed in a gas-filled chamber with quartz windows. The fiber is 0.7 m long and its bore diameter is 150 μm. Another lens having a 1-m focal length follows the fiber and collimates the output pulses. The pulses are then compressed with a pair of Brewster-angled SF10 prisms. The beam traveling in the backward direction through the prism pair slightly deflects vertically from the original direction, and it is reflected by a mirror to propagate into a grating spectrometer for spectrum analysis, or into a second-order, single-shot autocorrelator for pulse duration measurements.

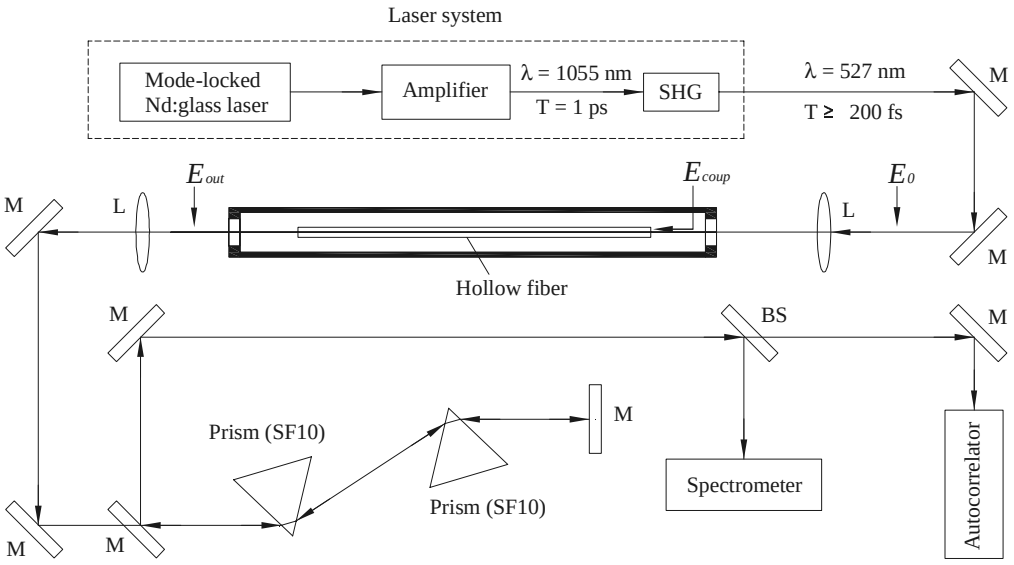


Fig. 1. Scheme of the experimental setup. BS: beam splitter; L: lens; M: mirror.

With reference to Fig.1, $E_0$ is the laser energy entering the gas cell (typically, ranging between 0.5-1 mJ), $E_{coup}$ represents the fraction of the input energy actually coupled to the hollow fiber fundamental mode, and $E_{out}$ is the energy at the output of the hollow fiber. $E_{coup}$ can be estimated by measuring $E_{out}$ , and using the relation $E_{out} = E_{coup} \exp(-\alpha L)$, where $\alpha$ and $L$ are the attenuation constant of the fiber, and the fiber length, respectively. The attenuation constant, $\alpha$, is evaluated as [1]

$$\alpha = \left(\frac{2.405}{2\pi}\right)^2 \frac{\lambda^2}{a^3} \frac{\nu^2 + 1}{\sqrt{\nu^2 - 1}}, \tag{1}$$

where $a$ is the fiber bore radius, $\lambda$ the pulse wavelength, and ν is the ratio between the refractive indices of the external fused silica and the internal gas medium. Thus, $E_{coup}$ reprents the fraction of the initial laser energy actually producing self-phase modulation in the gas medium, and is utilized as input parameter in the numerical model developed to simulate the experimental results, and described in the following.

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As a first step we verified that our initial, 300-fs-long laser pulse was actually chirp-free. This check was performed by directly sending the unperturbed pulse to the compressor, and verifying, by detuning the prism distance, that the ≈300-fs pulse duration actually corresponded to the minimum achievable duration. This implies a vanishingly small GVD of the initial pulse, which is the chirp dominant contribution.

We examine, first of all, the spectral broadening of the original pulse. In our experimental setup the laser intensity inside the hollow fiber is lower than the critical value for multi-photon ionization, which, for argon, is typically of the order of $10^{13} – 10^{14}$ W/cm$^2$ [5, 14]. So, the spectral broadening of laser pulses propagating inside the hollow fiber only depends on self-phase modulation occurring in the filling gas. Fig. 2(a) shows the experimental and theoretically simulated power spectrum of the pulse at the fiber output, in the optimal case of 2 bar of Ar, and of a coupled pulse energy of 0.38 mJ. The solid line is the experimental result, while the dashed line represents the simulated curve. The simulated spectrum, and its phase have been obtained through numerical integration of the Nonlinear Schrödinger Equation, by means of the Split-Step Fourier method [15]. The spectral broadening is accompanied by an oscillatory structure with five peaks. This behavior is typical of strong self-phase modulation in gases, and is due to the fact that the same instantaneous frequency is achieved at two different instants of time during the pulse [15]. From Fig. 2(a) one can see that the measured and simulated spectra are in good agreement, indicating that SPM is the dominant nonlinearity of the investigated process inside the gas in our experimental conditions.

Figure 2(b) shows the measured and simulated spectral widths of the fiber output pulses as a function of the gas pressure inside the hollow fiber for both argon and krypton. Experimental conditions, such as $E_{coup} \approx 0.35$ mJ and initial pulse duration **(**300 fs**),** were the same for the two noble gases. The figure indicates that the optical spectrum is broader when using krypton gas as compared to the argon case. As an example, when using argon, the spectral width of the output pulses was 17 nm at the pressure of 1.9 bar, while it amounted to 27 nm with krypton at the same pressure. The difference in pulse spectra for argon and krypton is caused by the fact that the nonlinear refractive coefficient of krypton is about twice as large as the nonlinear refractive coefficient of argon. In Fig. 2(b) the dashed lines represent the expected spectral widths vs the gas pressure, based on our numerical simulations. The theoretical behavior, based on the assumption of perfect coupling with only the fundamental mode of the fiber, is a linear increase of the spectral width with the gas pressure. Clearly, the

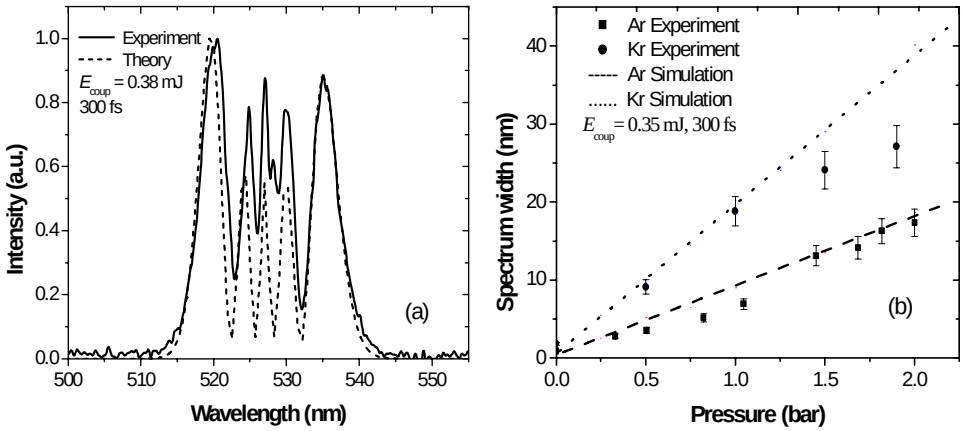


Fig. 2. (a) Measured (solid) and corresponding simulated (dashed) pulse optical spectrum in the best experimental conditions. (b) Spectral width.

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agreement with the measured values is very good for argon, whereas for krypton the experimental points show a saturation at high pressures, which is not reproduced by simulations. This discrepancy is due to strong coupling to higher-order modes of the fiber arising at high pressure (see next section), which implies a higher energy loss through the fiber, and less efficient overall self-phase modulation and spectral broadening. This effect is more severe for krypton than for argon due to its higher nonlinear refractive index, and starts at lower filling pressures for krypton than for argon. This also implies that for practical purposes argon must be preferred to krypton, despite its lower nonlinearity.

Let us now examine the dependence of the optical spectrum features of pulses at the fiber output on the initial pulse energy, when using 2 bar of argon as the nonlinear medium inside the fiber. Pulse spectra at the fiber output are similar to the one reported in Fig. 2(a), and are not shown here. Rather, in Fig. 3 we show the spectral width of pulses at the fiber output as a function of $E_{coup}$. In the figure we also report the result of our corresponding simulation. The

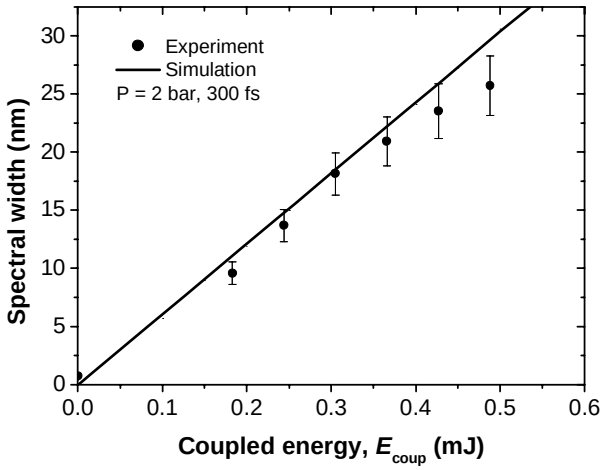


Fig. 3. Spectral width of the fiber output pulses as a function of the coupled energy.

simulated spectral width increases linearly with increasing initial pulse energy. The agreement between theory and measurement is very good up to coupled laser energies in the 0.4-0.5 mJ range, where the experimental points saturate. Once again, we attribute this discrepancy to our assumption throughout the simulation that laser pulses propagate only in the fundamental fiber mode. As we shall discuss in the following, at high input pulse energies, $E_0$, beam coupling to higher-order modes of the fiber becomes non-negligible. Therefore, pulse energy losses increase, and the final spectral width will not broaden any further, even by increasing $E_0$.

Figure 4 displays the compressed pulse duration versus $E_{coup}$. Squares represent the measured values, while full dots represent the simulated results in the case of ideal compression corresponding to the given initial pulse parameters. Ideal compression means that the phase response of the pulse compressor matches exactly the spectral phase of the pulses coming from the fiber, and that the dispersion of the pulse compressor can exactly compensate for all higher-order dispersion terms which are induced by propagation through fiber, lenses, and other relevant components of the optical system. As in the case of figure 3, the initial pulse duration is about 300 fs, and the argon pressure inside the fiber 2 bar. It can be

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seen that the compressed pulse duration monotonously decreases with the coupled pulse energy down to the shortest measured pulse duration of 30 fs at 527 nm, corresponding to a coupled pulse energy of 380 μJ. In this case, the pulse energy at the fiber exit was measured to be 310 μJ, giving rise to a final, measured energy of the compressed pulse of 160 μJ. This result is shown in Fig. 5 and represents our best achievement.

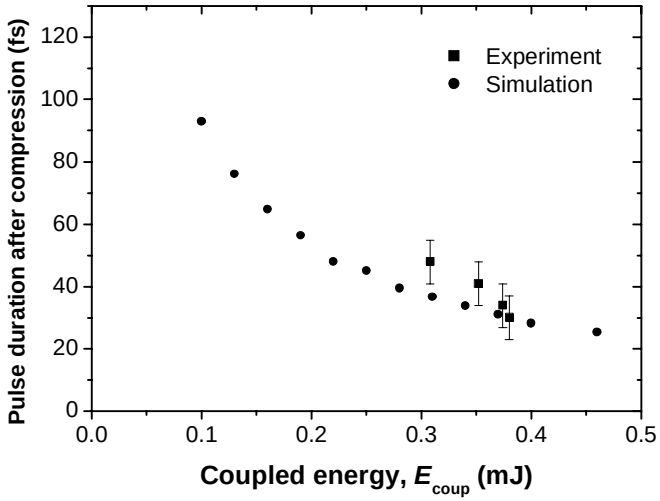


Fig. 4. Pulse duration versus the pulse energy coupled into the fiber.

Figure 5(a) shows the 2D autocorrelation pattern of the compressed pulse measured by the single-shot, second-order autocorrelator which makes use of a 1-mm-thick KDP crystal. On the *y*-axis the autocorrelation time-delay is reported, whereas the *x*-axis represents a

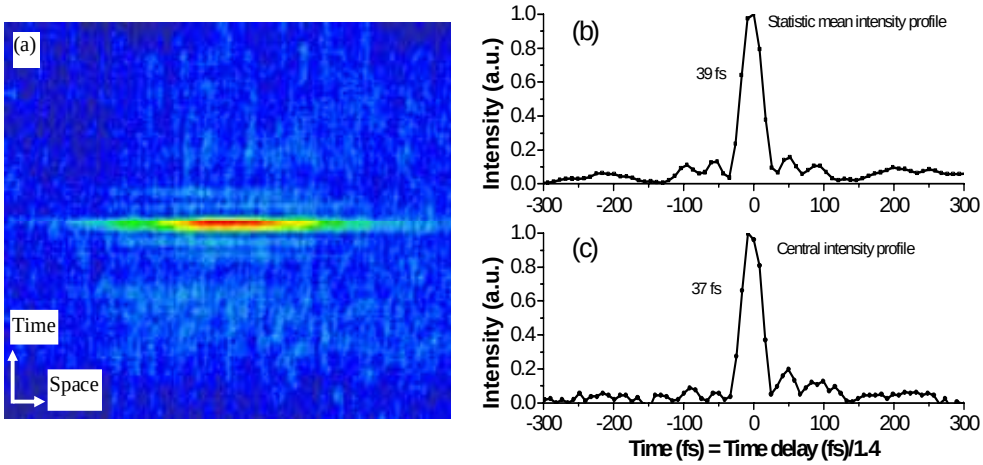


Fig. 5. (a) 2D autocorrelation pattern of the compressed pulses. (b) Spatially integrated autocorrelation traces of the compressed pulse. (c) Autocorrelation trace of the compressed pulse as read at the spatial centre of the wavefront.

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transverse spatial coordinate of the wave-front. Figures 5 (b) and (c) show the autocorrelation traces of the compressed pulses corresponding to the image shown in Fig. 5(a) when integrating over the whole spatial window (b), or corresponding to a specific section of the trace at the wave-front center where the pulse reaches its highest intensity (c). The temporal axis of the Figures 5(b) and (c), corresponding to the autocorrelation time delay, is directly reported in pulse intensity time units, obtained by dividing the time-delay by $\sqrt{2}$ . This corresponds to assuming a Gaussian profile for the pulse intensity temporal behavior, as it is typically done. The traces imply high quality pulses with a central lobe lasting 37 fs [FWHM, (c)], 39 fs [FWHM, (b)], respectively, and carrying about 75% of the total pulse energy. By subtracting the systematic pulse lengthening of ≈8.4 fs, induced by propagation through a 1-mm-thick KDP crystal in the single-shot second-order autocorrelator, we end up with a duration of the compressed pulse of about 30 fs.

Finally, Figure 6 shows the retrieved pulse temporal profile corresponding to the measured spectrum in Fig. 2(a). The corresponding time phase of the pulse is also reported. It is worth stressing that the temporal phase is flat in correspondence of the main peak of the compressed pulse, thus showing the absence of any residual temporal chirp. The retrieved result is in very good agreement with the estimated duration of the intensity profile obtained through the autocorrelation traces of Fig. 5(b) and (c). Since we do not directly measure the pulse spectral phase at the fiber output, we base our retrieval procedure on the following four steps. First the structure of the broadened measured spectra is carefully reproduced by means of our numerical simulation. Secondly, we infer the spectral phase of the spectrally broadened pulse by assuming that it is well reproduced by our calculated phase. The third step consists in accounting for prisms compression, by subtracting to the spectral phase the negative GVD contribution of the compressor. Fourth, an inverse Fourier transform gives the pulse intensity and phase in the time domain.

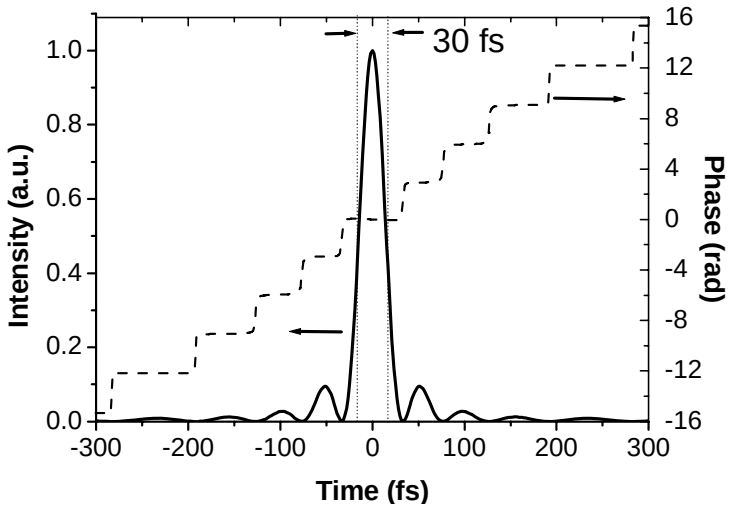

Fig. 6. Retrieved temporal shape of the pulse intensity profile (solid) and phase (dashed).

## 3. Physical limitations

In principle, we could achieve broader spectra of the optical pulses, shorter pulse durations after compression, and higher final pulse energies, by increasing the gas pressure inside the hollow fiber, or by improving optical pulse coupling into the hollow fiber, which would increase the actual optical intensity used for self-phase modulation inside the fiber filled with gas. However, spectral broadening of the pulses is substantially limited by the fact that a

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higher gas pressure, and/or higher initial pulse energy, leads to pulse propagation through the fiber in higher-order modes. In turn, this results in a much higher energy loss through the fiber with a consequent, limited pulse temporal compression and final energy.

We have found that we could obtain the broadest pulse spectra only when achieving the best mode-matching between the fiber propagation mode and the input beam spatial mode. In this case, pulses propagate in the fiber fundamental mode inside the argon-filled hollow fiber, and the fiber has a very small attenuation coefficient. Experimental results confirmed that the hollow fiber produced lower energy pulses, with narrower spectral widths, when the laser pulses propagated in a high order mode inside the hollow fiber.

As can be appreciated from Fig. 7, $E_{out}$ rolls over at input pulse energies higher than about 1 mJ. In fact, Figure 7 shows both the measured pulse energy at the fiber output (circles and solid line), and the measured efficiency of optical propagation through the fiber (squares and solid line), as a function of the input pulse energy $E_0$. The optical propagation efficiency of the fiber is defined as the measured value of $E_{out}/E_0$. It can be seen that the optical propagation efficiency decreases with increasing input pulse energy. The output pulse energy increases until it reaches a maximum when the input pulse energy is ≈ 1 mJ. We attribute the subsequent decrease of the output energy to the propagation of energy in higher-order modes inside the gas-filled hollow fiber. In principle, propagation in the sole fundamental fiber mode would lead to a linearly increasing function of the input energy (see the dashed line in Fig. 7).

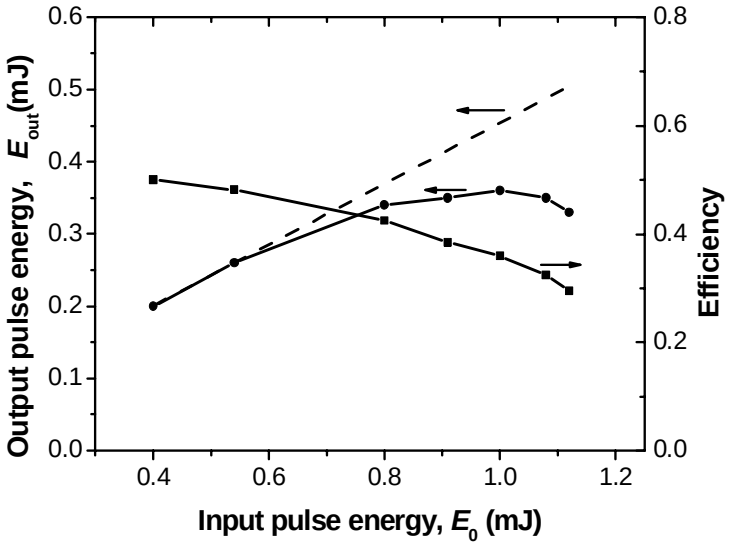


Fig. 7. Measured pulse energy at the fiber output and measured efficiency of optical propagation through the hollow fiber as a function of the input pulse energy. Solid circles and line: pulse energy at the fiber output versus the input pulse energy. Squares and solid line: optical propagation efficiency versus the input pulse energy. Dashed line: theoretical pulse energy at the fiber output in the case of propagation of the pure fundamental fiber mode.

Finally, Figure 8(a) shows the fiber *V*-parameter, which accounts for coupling to fiber higher-order modes, versus filling gas pressure at various initial pulse energies. The *V*-parameter is calculated as [15, 16]

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$$V = \frac{2\pi}{\lambda} a \sqrt{n_{core}^2 - n_{cladding}^2} \approx \frac{2\pi}{\lambda} a \sqrt{2 n_2 I_0}\,, \tag{2}$$

$n_{core}$ and $n_{cladding}$ being the maximum and minimum values of the refractive index inside the fiber, $n_2$ the nonlinear index coefficient, and $I_0$ the pulse peak intensity. For argon we have $n_2/p \approx 9.67 \times 10^{-24}$ $m^2/W\ bar$ [1], $p$ being the gas pressure. As shown in the literature [15, 16], if $V < 2.405$, the optical pulses will propagate in the fundamental fiber mode inside the fiber, whereas if $V > 2.405$, the optical pulse propagation will also take place in higher-order fiber modes. As shown in Fig. 2, the spectrum of the pulse at the fiber output is broadened when the gas pressure is increased inside the hollow fiber. Yet, too high pressures do not lead to bigger spectral broadening because the refractive index of the gas medium increases with gas pressure, thus increasing $V$ in Eq. (2). This can be seen in Fig. 8 (a), obtained by using Eq. (2), where $V$ increases monotonously with the filling-gas pressure at fixed initial pulse energy. As an example, $V$ increases from 1.02 at 1 bar to 2.51 at 3 bar, when the initial pulse energy is 0.6 mJ. The greater the $V$ parameter, the larger the number of modes the fiber will support and, as a consequence, the higher the propagation loss in the fiber. As it can be seen in Fig. 8 (a), $V$ also increases by increasing the pulse energy at constant gas pressure. For example, at the gas pressure of 2 bar, $V>2.405$ when the initial pulse energy is larger than 0.8 mJ. This means that a relevant part of high energy pulses propagates in higher-order modes inside the hollow fiber, leading to higher energy losses, with a consequent reduction of the field intensity inside the fiber. Thus, the spectral width of the pulse will not be efficiently broadened in this latter case. As a matter of fact, the effect of depleting the optical power stored in the fundamental fiber mode starts even before reaching the $V$ threshold value of 2.405, as it can be appreciated in Fig. 8 (b), where the optical power coupled to the fundamental fiber mode is plotted versus $V$ when the fiber input pulse energy is kept constant [16]. This is due to the fact that when approaching the threshold value $V = 2.405$ from below, the attenuation coefficient of the fundamental mode increases [16]. Thus, when $V \geq 1.8$, the output energy, stored in the

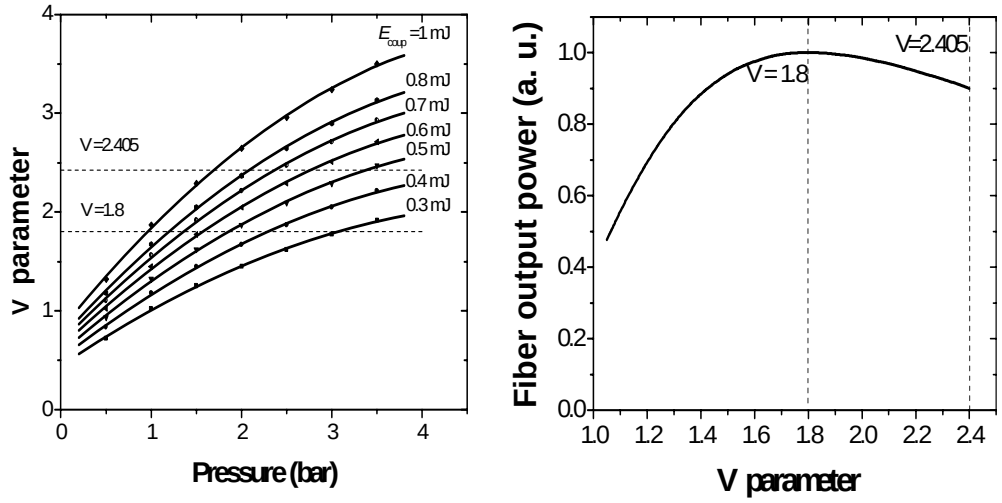


Fig. 8. (a) Normalized frequency versus filling-gas pressure at the differently initial pulse energy. (b) Fiber output power in the fundamental mode

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fundamental mode, appears to rollover as shown in Fig. 8(b). Interestingly, the experimental cases of our best pulse compression correspond to the *V*-parameter ranging within 1.6 and 2.2, thus matching very well the best possible region for propagation in the fundamental fiber mode only.

It is also worth stressing that the above physical limitation to the possibility of temporally compressing high-energy, ultrashort laser pulses by means of the hollow fiber technique is more severe in the visible wavelength range as compared to the infrared (e.g., Titanium-Sapphire laser source emitting at ≈ 800 nm) due to the presence of the laser pulse central wavelength in the denominator of Eq. (1).

## 4. Conclusion

In conclusion, we demonstrated that, by using an argon-filled hollow fiber, and a prism-pair compressor, 200 ~ 300 fs, 1-mJ, 527-nm laser pulses can be shorten down to 30 fs, with a final pulse energy of 160 μJ.

Initial pulse energy and duration, and gas pressure inside the argon-filled hollow fiber all play an important role in the spectral broadening of laser pulses propagating through the fiber. However, such spectral broadening is limited by the need of propagating pulses only in the fundamental mode of the fiber, in order to achieve the minimum final pulse duration coupled to high pulse energy, and good spatial quality of the output beam.

Both experimental measurements and theoretical simulations, based on the numerical integration of the Nonlinear Schrödinger Equation with the Split-Step Fourier Method, indicate that the spectral width of the laser pulses increases linearly with both gas pressure, and initial laser pulse energy actually coupled to the fiber, only when the energy is propagated through the hollow fiber in the fundamental fiber mode. In particular, argon is to be preferred as nonlinear gas medium to achieve the best compromise between short pulse duration, high final pulse energy and spatial quality of the final beam. We obtained the best temporal pulse compression, with a final pulse length of ≈ 30 fs, in correspondence of a broadened spectral width of ≈ 25 nm, at a gas pressure slightly lower than 2 bar.

Finally, it is worth stressing that in the best conditions, a peak power as high as ≈ 5.3 GW was reached by the compressed pulse. Since this high peak power is coupled to a remarkable spatial coherence of the final beam, peak intensities in excess of $10^{15}$ W/cm$^2$ can easily be obtained through proper focusing, thus also allowing high-energy physics applications of the compressed pulse.

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