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Spectral broadening and compression of picosecond pulses in air using a multi-pass cell

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Abstract: We report the spectral broadening and post-compression of picosecond pulses using a multi-pass cell (MPC) operating in air. A 2 m-long bi-concave cavity with 41 reflections and 96.4% transmission was designed and implemented, optimized for the compression of 5.5 ps, 4.5 mJ pulses from a Yb:YAG laser operating at 1030 nm. A 19.3-fold spectral broadening was obtained, increasing the bandwidth from 0.3 nm to 5.8 nm, which enabled pulse compression down to 547 fs with a symmetric temporal profile close to the transform limit. These results demonstrate the feasibility of using open-air MPCs for high-energy picosecond pulse post-compression, without the need for sealed chambers, while preserving high mode quality and transmission. Scaling to pulse higher energy is discussed.

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1. Introduction

Nonlinear spectral broadening (NSB) is a widely used technique as a preliminary step to post-compression of short or ultrashort pulses that have already reached their transform limit. This process has numerous applications that include strong-field physics, attosecond science, and industrial material processing [1–3].

Ytterbium-based laser systems have the advantage of being pumped directly with commercially available diodes. Although they achieve high pulse energy, high average power, and high efficiency [4], they naturally deliver pulses limited to the picosecond or several hundreds of femtoseconds range due to their relatively narrow bandwidth. Therefore, NSB has become a key solution to generate femtosecond pulses [5–7].

Multi-pass cells (MPCs) [1,8] are currently one of the most versatile and efficient methods for NSB. They consist of a two-mirror cavity formed by a pair of facing curved mirrors, typically concave, in which the beam undergoes many successive reflections. This allows for the accumulation of a large total nonlinear phase as a result of multiple weak nonlinear interactions at each pass, while maintaining high transmission and good spatial homogeneity. Their configuration also minimizes spatial and temporal distortions and allows control of the NSB by adjusting the number of passes, mode size, or nonlinear medium [1]. NSB results have been reported both in MPCs fully filled with gas [9–11] and in setups where certain sections are occupied by bulk materials [12–14].

Although NSB effects in bulk materials can be up to three orders of magnitude higher than in gases, they present greater complications at high intensity due to self-focusing. Therefore, typical compressed energies range from μJ to a few mJ [12–14]. Thus, the possibility of allowing higher pulse energies in the absence of self-focusing-induced damage makes spectral broadening in gases particularly attractive.

Noble gases such as argon, helium, neon, or xenon are commonly used, enabling pulse compression down to the tens or hundreds of femtoseconds regime. For example, in the case of argon-filled MPCs, Lavenu et al. [10] demonstrated the compression of 160 μJ , 275 fs pulses down to 33 fs at 7 bar; Russbuehl et al. [11] compressed 1.1 mJ, 590 fs pulses to 26.5 fs at 3.5 bar; Kaumanns et al. [9] achieved compression of 18 mJ, 1.3 ps pulses to 41 fs at 0.6 bar and, in a subsequent work, Kaumanns et al. [15] used a donut-like mode to compress 112 mJ pulses from 1.3 ps to 37 fs. In helium, Pfaff et al. [16] recently demonstrated the compression of 200 mJ pulses from 0.5 ps to 50 fs in a 10 m long cell. In neon, Ueffing et al. [17] reported spectral broadening and compression of 2 mJ, 210 fs pulses down to 37 fs in a gas-filled multipass cell. In the case of xenon, its large nonlinear index enables strong spectral broadening at relatively low pressure; for example, Guichard et al. [18] demonstrated efficient nonlinear compression of sub-picosecond pulses in xenon-filled multipass configurations at the microjoule level.

It is worth noting that the use of these gases requires the construction of a chamber to isolate the MPC from the environment and contain the chosen gas. This becomes complicated when the mirror spacing becomes large (several meters), which is necessary as the laser energy increases, in order to keep the beam size on the MPC mirrors below the damage threshold. It is well known that there is a trade-off between how small the beam waist can be at the center of the MPC to avoid gas ionization, and how low the beam divergence can be without causing mirror coating damage [1]. In this context, air represents a particularly interesting nonlinear medium since it does not require a sealed cell. Recently, Omar et al. [19] demonstrated the spectral broadening of 2.1 mJ, 670 fs pulses, later compressed to 134 fs, in a compact convex–concave MPC filled with air and operated under ambient conditions, showing that air-filled MPCs can induce substantial broadening without coating damage or plasma formation. Later, Feng et al. [20] demonstrated nonlinear post-compression of 145 μJ , 185 fs pulses to sub-20 fs using a MPC filled with air. Similarly, Gao et al. [21] reported multi-pass compression in air of 400 μJ , 206.6 fs pulses down to 28.2 fs. In the UV regime, Hariton et al. reported spectral broadening of 220 fs, 12 μJ pulses in an air-filled MPC, supporting compression to 27 fs [22].

Alternatively, Schönberg et al. demonstrated spectral broadening of 8 mJ, 1 ps pulses in a compact folded MPC operated in air, achieving compression down to 51 fs [23]. It employs a non-conventional compact MPC that virtually increases the distance between two concave mirrors (ROC = 25 m) through multiple reflections between flat mirrors. This configuration requires the use of a large beam waist of 3.5 mm ($1/e^2$) to prevent mirror damage as the fluence increases during successive focusing reflections. In a different approach, Takil et al. [3] demonstrated the feasibility of using atmospheric air as a nonlinear medium for 65 mJ, 5.5 ps pulses in a configuration different from a standard MPC. In that setup, a collimated beam with a 5 mm ($1/e^2$) diameter propagated freely over a distance exceeding 60 m after undergoing multiple reflections between flat mirrors. The pulses were temporally split into two subpulses and later recombined, achieving compression to 1.1 ps. While this method is effective for pulses of increased energy, it lacks the advantage of the frequency mixing characteristic of a conventional MPC. Moreover, in methods involving long-distance propagation of large-diameter beams, as in the two approaches described above, the high sensitivity to wavefront inhomogeneities can lead to filamentation. Consequently, to the best of our knowledge, post-compression of picosecond mJ pulses into the femtosecond regime has not yet been demonstrated in conventional open-air MPC cavities while preserving the advantages of high spectral homogeneity and excellent beam quality.

2. Air MPC design

For the design of the MPC, the following factors were considered. First, the total length (including optomechanics) had to fit within the available 2 m space in the laboratory. Second, the intensity at the center of the MPC should not exceed the Raman effect threshold intensity in air, 2.15 TW/cm² [24]. This threshold is defined here as the intensity above which the asymmetry in the broadened spectrum makes the pulse uncompressible (as shown in Section 4). Third, the mirror diameter had to allow for the inclusion of a one-inch in/out-coupling mirror, avoiding the need to drill holes in the mirrors. Fourth, the number of reflections was set so that the beam enters and exits the MPC through the in/out-coupling mirror. Fifth, the fluence on the mirrors must remain below the coating damage threshold.

A bi-concave configuration was chosen, consisting of 8-inch (203.2 mm) circular mirrors with a radius of curvature $R_C = 1$ m, made of UVFS substrates with a high-reflectivity (HR) HfO₂/SiO₂ stack at 1030 nm (reflectivity = 99.99%) at normal incidence, deposited in-house using ion beam sputtering. The measured damage threshold for the mirror coatings is 3.6 J/cm². A 1000-on-1 LIDT measurement was conducted with 5.5 ps pulses with a 1.16 mm 1/e² diameter and 19 mJ energy. However, the damage threshold of the in/out-coupling mirror HR at 1030 nm at 45° is lower, 0.7 J/cm² (measured for single 5.5 ps pulses with a 1.16 mm 1/e² diameter and 3.7 mJ energy). Therefore, the maximum working fluence we set for operation is 0.4 J/cm², corresponding to a safety factor of 1.75, which, based on our experience, provides an adequate margin for safe and stable operation. Note that the experiment could be conducted with a smaller mirror diameter (e.g., 7 inches); however, we chose 8-inch mirrors for increased flexibility in conducting the experiment.

For a stable cavity, the mirror separation must satisfy $L < 2R_C = 2$ m [8]. Using ray matrices [25], the position (x_n, y_n) and propagation angle ($\theta_{x,n}, \theta_{y,n}$) of each reflection on the mirrors are determined according to the equation:

$$\begin{pmatrix} q_n \\ \theta_{q,n} \end{pmatrix} = \begin{pmatrix} 1 & 0 \\ -\frac{2}{R_C} & 1 \end{pmatrix} \begin{pmatrix} 1 & L \\ 0 & 1 \end{pmatrix} \begin{pmatrix} q_{n-1} \\ \theta_{q,n-1} \end{pmatrix} = \begin{pmatrix} q_{n-1} + L\theta_{q,n-1} \\ -\frac{2}{R_C}q_{n-1} + \left(1 - \frac{2L}{R_C}\right)\theta_{q,n-1} \end{pmatrix} \quad (1)$$

where q generally denotes the spatial coordinate x or y , and the free-space propagation matrix over a distance L and the reflection matrixes for a curved mirror were used. From this equation, the pattern shown in Fig. 1 was constructed. For a length of $L = 1988$ mm, a total of 41 reflections are sufficient for the first (1-input) and the last pass (41-output) beams to strike a one-inch (25.4 mm) mirror without clipping from neighboring reflections. Allowing the reflections on the in/out-coupling mirror to not overlap exactly at the same spot enables a simple separation of the output beam.

For this configuration, mode matching for a perfectly Gaussian beam with a wavelength of 1030 nm is achieved with a waist at the center of the MPC of $w_0 = 159.1$ μm (see inset of Fig. 1) and a beam radius of $w = 2.05$ mm (1/e²) on the mirrors. With these parameters, the maximum pulse energy that does not exceed the previously defined fluence limit of 0.4 J/cm² (due to the limitations of the in/out coupling mirror) is 26 mJ. However, to remain at least 10% below the Raman threshold in air, the pulse energy should not exceed 4.5 mJ, which corresponds to a maximum intensity at the center of the MPC of 1.93 TW/cm² for 5.5 ps pulses.

On the other hand, from the B-integral for this configuration [1]:

$$B = N \frac{16\pi n_2 P_0}{\lambda^2} \tan^{-1} \left(\frac{L}{2z_R} \right) \simeq 13\pi \quad (2)$$

where N is the number of roundtrips, n_2 is the nonlinear refractive index at 1030 nm (3.8×10^{-23} m²/W) [3], P_0 is the peak power, λ is the wavelength, and z_R is the Rayleigh length of the Gaussian beam.

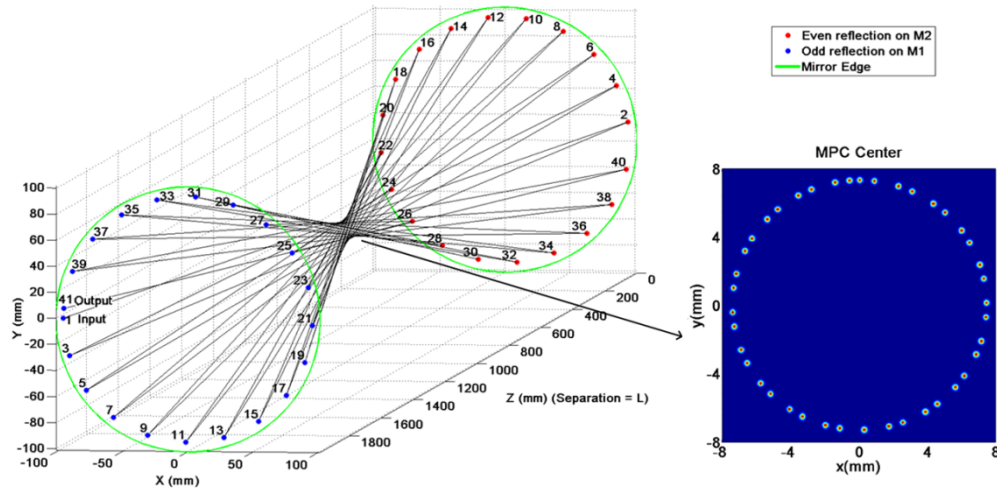


Fig. 1. Simulated reflection pattern inside the bi-concave MPC cavity showing 41 reflections that fit within the mirror aperture while allowing for the separation of the input and output beams on the in/out coupling mirror. The inset shows the Gaussian profiles of each reflection at the center of the MPC. The simulation was performed using the following parameters: mirror radius of curvature $R_C = 1000$ mm, mirror separation $L = 1988$ mm, initial horizontal position $X_0 = 0.0$ mm, initial horizontal angle $\theta_{x,0} = 0.4^\circ$, initial vertical position $y_0 = -96.0$ mm, initial vertical angle $\theta_{y,0} = 5.5^\circ$, and mirror diameter = 203.2 mm.

With this B integral it can be expected, under ideal conditions, that the $\tau_{in} = 5.5$ ps pulse can be compressed to a transform limited pulse of:

$$\tau_{out} = \frac{\pi}{B} \tau_{in} \simeq 420 \text{ fs} \quad (3)$$

3. Experimental setup

Figure 2 shows a schematic of the experimental setup. A diode-pumped thick-disk Yb:YAG chirped-pulse amplification (CPA) laser, similar to that described in [26,27] and operating at a repetition rate of 10 Hz, is used. The beam exits collimated, and to achieve mode matching with the MPC, a tunable three-lens zoom system is used, consisting of two positive lenses (L_{Z1} and L_{Z2}) and one negative lens (L_{Zn}). By changing the position of lenses L_{Zn} and L_{Z1} , the beam radius w can be continuously varied within the range $[w/2, 2w]$, providing a wide flexibility. The beam is then directed, using two 45° HR dielectric mirrors (M_5 and M_6), toward a focusing lens (L_f), which can be translated to align the beam waist with the center of the MPC after the beam passes through a periscope (mirrors M_3 and M_4) and the in/out-coupling mirror (M_0).

After multiple reflections in the MPC, the resulting beam is directed to a gold grating compressor, which is followed by an autocorrelator for pulse duration measurements. A small portion of the beam energy is directed into a grating spectrometer (SpectraPro 2750 with 0.1 nm spectral resolution) to measure the spectral broadening.

Figure 3 shows the beam profiles at the MPC input ($M^2 = 1.08 \times 1.16$) and output ($M^2 = 1.14 \times 1.13$), after achieving mode matching, along with the M^2 measurements corresponding to the beam exiting the MPC (focused with a lens of $f = 1500$ mm). A good preservation of the mode is observed, with $w(1/e^2) = 4.7$ mm, measured in the far field at the position of mirror M_5 (see Fig. 2). The MPC throughput is 96.4%.

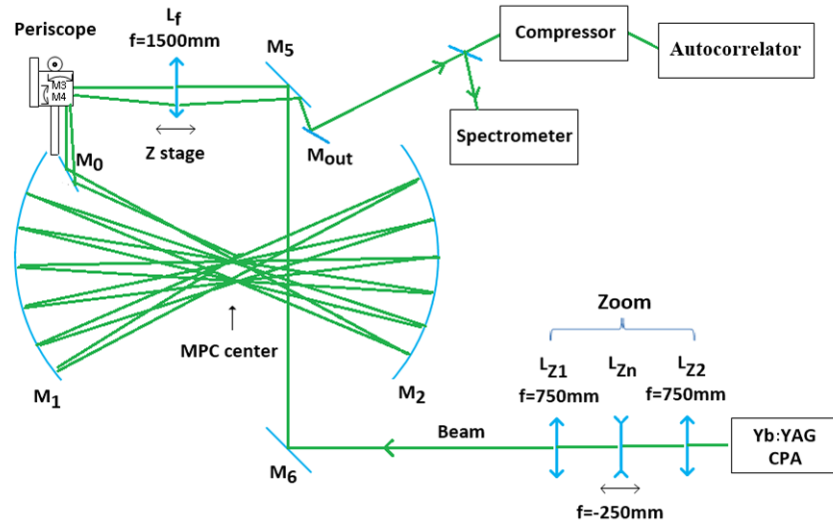


Fig. 2. Schematic of the experimental setup showing the Yb:YAG CPA laser system, the MPC, and the tunable three-lens zoom system.

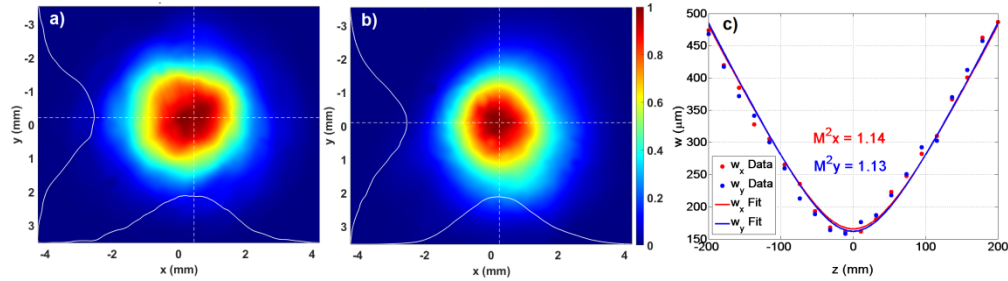


Fig. 3. Beam profiles at the MPC (a) input and (b) output after mode matching (c) M^2 measurements corresponding the beam exiting the MPC.

4. Results

Figure 4(a) shows a comparison between the average spectrum of 180 pulses at 4.5 mJ measured at the MPC output and the input pulse spectrum. A spectral broadening by a factor of 19.3 is observed, increasing the original bandwidth from 0.3 nm to 5.8 nm. The averaging is performed to improve the signal-to-noise ratio; however, the measured spectrum is very stable, as shown in Fig. 4(b), where each of the 180 consecutive single shots is plotted. These data were obtained without an enclosure which could be added to further improve stability. These spectra were recorded over a period of one hour, with a time interval of 20 seconds between measurements. The result corresponds to the spatial integration of the output pulse.

When the energy is increased to 5 mJ, the intensity at the center of the MPC reaches the threshold of 2.15 TW/cm^2 , and the onset of the Raman effect is observed (Fig. 5), which creates an asymmetric spectrum with incompressible components.

Output pulses with 4.5 mJ energy were compressed to 547 fs using a gold grating compressor (throughput $\sim 65\%$) assembled with 1200 l/mm gratings available in our lab. Figure 6 shows the comparison between this pulse and the transform-limited pulse of 446 fs, calculated from the spectrum in Fig. 4(a), a value that is close to that predicted by Eq. (3). The spectral

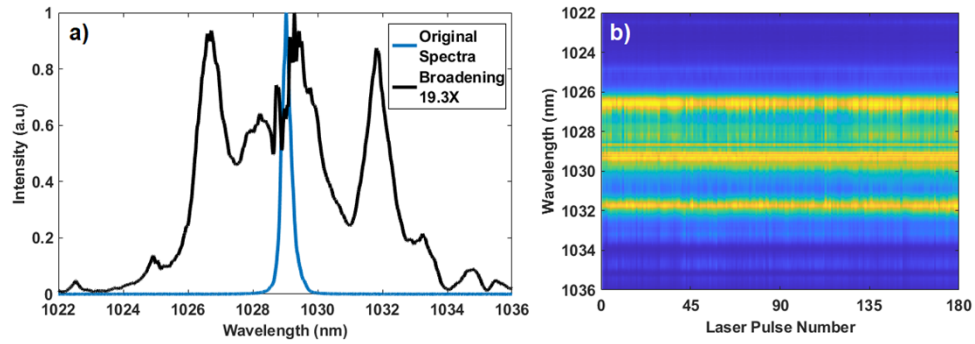


Fig. 4. (a) Comparison between the input spectrum and the averaged output spectrum at 4.5 mJ. (b) Output spectral stability over one hour.

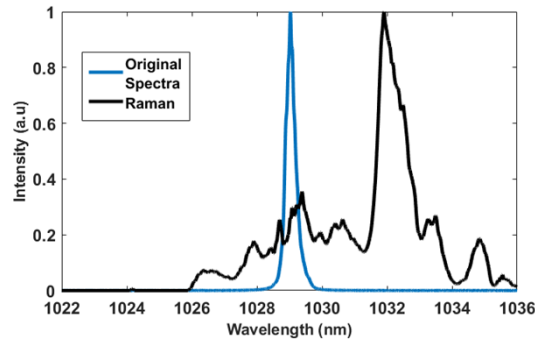


Fig. 5. Comparison of the input spectrum and the output spectrum at 5 mJ showing Raman-induced shift.

broadening in the MPC introduces predominantly second-order dispersion, whereas the grating compressor is expected to provide both second- and third-order dispersion. As a result, while the second-order dispersion can be largely compensated by adjusting the compressor, some residual third-order dispersion may remain, potentially leading to a slight degradation of the compression, resulting in this case in a pulse duration that differs by 18% from the transform limit. The temporal measurements were performed using a home-built single-shot second-harmonic autocorrelator based on a 20 mm diameter, 1 mm thick KDP crystal and a CCD camera with an 8.45 mm $\times$ 7.07 mm sensor.

It is worth noting that, in our system, the primary limitation to increasing the pulse energy is the Raman threshold intensity in air. Since the maximum intensity occurs at the center of the MPC, modifying the MPC dimensions so that mode matching occurs with a larger beam radius at the center allows the same intensity to be maintained at higher energy.

By performing a simulation equivalent to that shown in Fig. 1, it can be shown that reducing the MPC length by 345 mm changes the beam size in that region after mode matching from $w_0 = 159 \mu\text{m}$ to $w_0 = 354 \mu\text{m}$. This results in an increase of the central beam area by a factor of approximately 5, which would allow the energy to be increased from 4.5 mJ to 22 mJ while maintaining the same intensity of 1.93 TW/cm² used in this work, for which no Raman was observed.

This modification of the cell and the corresponding increase in energy would also lead to an increase in the fluence on the mirrors to 2 J/cm². Although this value still leaves a safety factor of 1.8 for the 8-inch mirrors, it would require replacing the in/out coupling mirror for one with

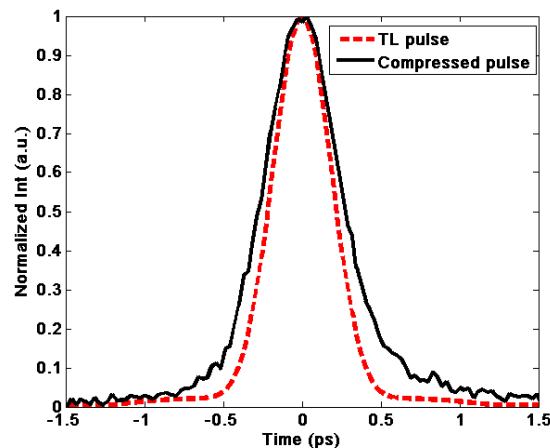


Fig. 6. Comparison between the compressed pulse (547 fs) and the transform-limited pulse (446 fs) (FWHM). The signals correspond to the autocorrelation peaks, one measured with a single-shot autocorrelator and the other obtained from the numerical autocorrelation of the TL pulse, calculated from the spectrum in Fig. 4(a). The temporal scale has been adjusted by a factor $\sqrt{2}$ so that the full width at half maximum matches the width of the deconvolved peak, as expected for Gaussian pulses.

higher damage threshold, similar to that of the MPC mirrors. Under these conditions, Eq. (2) can predict that it is possible to achieve a B-integral of 32.4π for 37 reflections, which, for the new mirror separation, would allow the last reflection to be extracted using the same in/out coupling mirror. In this way, from Eq. (3) we can predict that for 22 mJ pulses in air, pulse compression with values approaching the transform-limited 170 fs could be achieved.

5. Conclusion

The experiment presented in this work demonstrates that efficient spectral broadening of multi-mJ picosecond pulses can be achieved in an ambient air multi-pass cell without the need for a sealed gas chamber. The conventional air-filled MPC enabled a 19.3-fold spectral broadening of 5.5 ps, 4.5 mJ pulses, followed by compression to 547 fs. The output beam exhibited excellent spatial quality and temporal stability, indicating that nonlinear propagation in air can produce uniform broadening without detrimental effects such as self-focusing or plasma formation when operating below the Raman threshold. A reconfiguration of the MPC can be expected to allow for the post compression of higher energy pulses.

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Data availability. Data underlying the results presented in this paper are not publicly available at this time but may be obtained from the authors upon reasonable request.

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