Production of single-cycle laser pulses through nonlinear pulse compression

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Today most ultrashort lasers are capable of producing pulses in the order of dozens of femtoseconds, using modelocked Ti:Sapphire oscillators. These ultrashort pulses are the tool of choice for exploring electron dynamics inside atoms, molecules, solids, and nanostructures. As time resolution is limited by pulse duration, achieving shorter pulses allows us to look at the previously hidden processes. In particular, few-cycle pulses enable the generation of secondary sources of isolated attosecond pulses (IAPs) produced through few-cycle laser-driven high-harmonic generation (HHG), unlocking diagnostic tools like attosecond x-ray diffraction, and spectroscopy with tabletop sources.

Here we present a post-compression installation at the VOXEL laboratory (GoLP/IPFN at Instituto Superior Técnico). The setup consists of a Coherent Astrella laser (40 fs, 800 nm, 3 mJ, 1 kHz) whose output is focused onto a 250 μm inner diameter differentially pumped hollow core fibre, pressurized with Argon gas. The spectrally broadened output is compressed and measured in time by a d-scan system from Sphere Ultrafast Photonics. We have achieved sub-4 fs with minimal group delay dispersion (GDD) and transmission of 33%. The setup represents a powerful tool for table-top single-cycle laser pulses in laboratories and we are now ready to produce attosecond pulses in the XUV region through high-harmonic generation.

We also studied post-compression techniques in the *Laboratoire d'Optique Appliquée* (LOA) and present the results here.

1. INTRODUCTION

In Instituto Superior Técnico, pulses at the femtoseconds regime have been delivered since 2017 at the VOXEL laboratory. Being one of few facilities in Portugal with such capabilities, the VOXEL group takes advantage of a process called High Harmonic Generation (HHG) to create some of the shortest man-made pulses. These pulses need to be in higher frequencies since for each wavelength there's a limit to the pulse duration. In the NIR region, like our femtosecond pulses, the limit is set at around 2.5 fs, for soft X-Ray pulses the limit is much lower, in the attosecond range.

HHG is achieved by using femtosecond lasers and a gas cell to produce pulses in the extreme ultraviolet region with hundreds of attoseconds. Having achieved a reliable coherent XUV source [1] a few years ago, the group is now looking to progress towards Isolated Attosecond Pulses (IAP), i.e. pulses in the attosecond timescale. These even shorter durations promise new insight into physical, chemical, and biological processes, thanks to methods of attosecond x-ray diffraction and spectroscopy. Imaging techniques with IAP require hundreds of μJ of energy on target [2] with pulse durations close to the single-cycle regime, i.e. less than 4 fs in 800 nm.

Pulses of such capabilities are on the front edge of laser technology and can only be achieved through a new technique called post-compression. This is the case since the limited gain bandwidth of the amplifying medium of commercial lasers limits the possible pulse duration of its beam.

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1.1. Theory of Post-Compression

To achieve the desired single-cycle pulses, we need to start by looking at the fundamental behaviour of electromagnetic wave packets: a pulse can be fully described either in the time or in the frequency domain which are connected by the Fourier transform. This is, of course, analogous to particles in quantum mechanics where the domains in question are momentum and space. And so, we have the bandwidth theorem where the product of the representation of the two domains can not be arbitrarily small but must respect a minimum value. In quantum mechanics this is known as Heisenberg's uncertainty relation, in ultrafast optics, it reads as

$$\Delta\omega \cdot \Delta t \geq \text{constant}, \tag{1}$$

where $\Delta\omega$ is the spectral bandwidth and Δt is the pulse duration. When the product is at its minimum value we can say that it is at the Fourier Transform Limit (FTL).

We have then two goals: (i) we need to achieve a broad enough bandwidth to allow a shorter FTL pulse and (ii), all the components of the spectral phase have to be constant over the entire spectrum (to achieve FTL). However, most of the frequently used lasers have too narrow a gain bandwidth due to the finite widths of the energy levels of the amplifying medium. Therefore the spectrum of their pulses cannot be broadened within the laser itself. Instead, the beam needs to be broadened outside the laser cavity before it can be compressed to a single-cycle duration [3]. This technique is called **post-compression** (compression after broadening) and is the core of this work.

There are several ways to achieve a broadened spectrum. In an approach of free propagation, the techniques mostly used are (i) free propagation in bulk,

(ii) multi-plate continuum generation and (iii) multi-pass cells (MPC). To perform spectral broadening in a more controlled way, it is more common to resort to gas-filled Hollow Core-Fiber (HCF). At the VOXEL laboratory, we chose to take advantage of the latter with resource to Self-Phase Modulation

Self Phase Modulation is the nonlinear phase modulation of a beam, caused by its own intensity via the Kerr effect, a third-order nonlinear optical effect. It occurs when the intensity of a light pulse propagating in a medium is high enough to distort the medium's atomic potential. This forces the valence electrons to oscillate an-harmonically and changes the macro properties of the medium. In particular, the refractive index becomes intensity-dependent.

$$n = n_0 + n_2 I(t) \tag{2}$$

Since the instantaneous frequency is proportional to the refractive index,

$$\omega_{inst}(t, z) = \frac{\partial \phi}{\partial t} = \omega_0 - \frac{\omega_0}{c}\frac{\partial n(t, z)}{\partial t} z, \tag{3}$$

an intensity-dependent refractive index will create new frequencies and our spectrum's bandwidth will increase, we call this spectral broadening.

This description of self-phase modulation with a time-dependent refractive index is somewhat simplified and accurate only for pulses longer than 10 fs (in the NIR region). In the VOXEL laboratory, we need to extend the concept since we are creating single-cycle pulses. There, we need to take into account the effects of self-focusing and self-steepening, as well as other nonlinear effects.

The spectral broadening of the pulse can only occur if there are changes to the spectral phase. These changes result in pulses on the order of hundreds of femtoseconds. To correct this, we need to temporally compress the pulse. For that we need chirp management, a process that brings all spectral components in phase and makes the spectral phase constant. Most compression setups include a series of chirped mirrors and additional wedges of fused silica that apply continuous tuning of the chirp, such is the case of our project [4]. Once 1.5 or fewer optical cycles are achieved, then there's a need to incorporate additional chirp compensation such as third-order dispersion (TOD) compensation using a combination of bulk materials with different ratios of the group-delay dispersion (GDD) to the TOD [5].

2. POST COMPRESSION AT THE VOXEL LAB

To install the post-compression technique at the VOXEL laboratory, we first had to analyse the existing pulse that exits the Astrella laser system. For this, we set up a Second-Harmonic Frequency Resolved Optical Gating [6] tool to measure the original pulse duration. We also used a CinCam CMOS-1201 beam profiler to study how much the pulse drifts. The horizontal and vertical deviations of the laser beam are plotted in figure 1. The spectral broadening will occur in a fibre with an inner core of 250 μm, in the same order of magnitude as the drift, it is then clear that there's a need to control the vertical drift. For this, we have commissioned an Active Beam Stabilisation system by TEM Messtechnik GmbH.

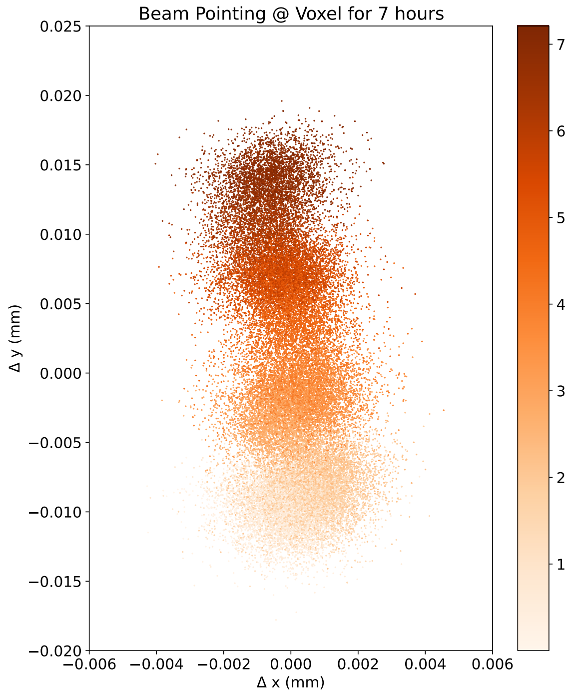

FIG. 1: Horizontal and vertical deviation of the beam centre measured with the beam profiler for over 7 hours. Note the axis scale.

With proper knowledge of the limitations of our laser we finally started the implementation of the Hollow-Core Fibre (HCF). We moved both the Astrella and the Hydra lasers to create space and implemented a new beamline in between the lasers to be directed into the fibre. We aligned the on-axis line of the fibre with both the NIR and HeNe lasers. After, we placed an achromatic lens and found the focus plane where we could achieve optimal coupling into the fibre. We also placed a beam profiler to monitor any changes in beam pointing and coupling into the fibre. We could then place the rail where fibre would lay as well as the gas and vacuum systems that were connected to a gas station. We aligned this setup to the previously fixed beam line. Finally, we placed the 1-meter-long Hollow-Core Fibre provided by John Tisch from Imperial College London Consultants.

The fibre supports up to 2 mJ of energy due to differential pumping (the input of the fibre remains in vacuum while the output is pumped with a noble gas). This approach is used since it allows for more energy intake by reducing plasma at the entrance and increasing the limit for ionisation. With this, we achieved a transmission of 33% but by controlling the beam pointing it can go up to around 50% [7, 8].

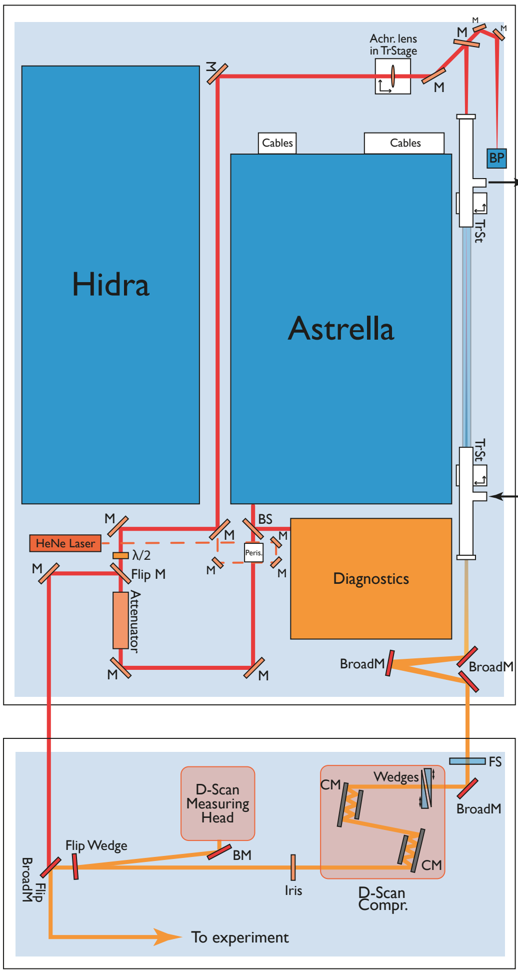

FIG. 2: Scheme of the setup of the HCF in the VOXEL laboratory. In the first optical table, we find the spectral broadening system composed of an attenuator, a focusing lens and the HCF (which includes also the gas and vacuum structure). In the second table, we see the D-Scan system made of the compressor and the measuring head. It's also here that the beamline to HHG is placed.

The reason why the transmission is subpar derives from two main flaws in our system (described in figure 2): the lack of stability of the beam pointing and the lack of polarisation control. The vertical drift of the beam affects the transmission of the fibre by not allowing a proper and stable coupling of the beam into the fibre. The polarisation effect is more complex: when the beam moves into the second table, there is a difference in height that doesn't maintain the parallel polarisation set by the Brewster windows in the spectral broadening system. This needs to be precompensated which requires realignment of the fibre and a more deficient control of the polarisation.

Having achieved a broadband pulse, we started by studying the spectrum at the output of the fibre. The amount of pressure defines how much broadening happens since $n_2 = p \cdot k_2$ in gases and so a study was done in order to analyse the amount of broadening for each pressure of Argon ($n_2 = 10.8 \times 10^{-20}\text{cm}^2/\text{W}$ at atmospheric pressure [9]).

The broadening started to occur with a pressure of 0.6bar and increased until 1.8bar where the system set a hard limit of input pressure. The evolution of the spectrum is represented in figure 3.

There was also an attempt to do this study with Helium ($n_2 = 0.3 \times 10^{-20}\text{cm}^2/\text{W}$ at atmospheric pressure) and Neon ($n_2 = 1 \times 10^{-20}\text{cm}^2/\text{W}$ at atmospheric pressure) but the first needed metal tubes to prevent leaks due to the small size of the atom and the second is out of reach due to the current worldwide shortage.

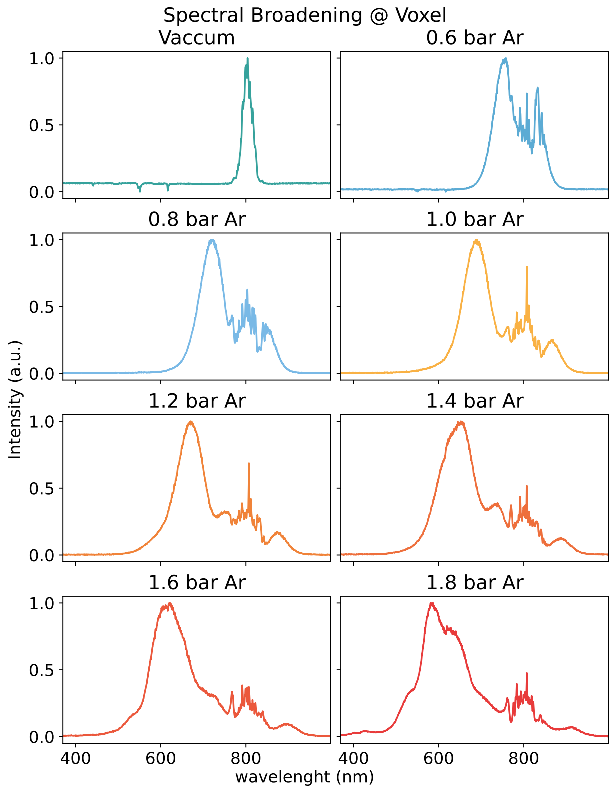

FIG. 3: Evolution of the normalised spectral shapes with increasing gas pressure in the HCF of the VOXEL lab.

With a broadband pulse we could finally start to install the temporal compression system. For that, we use a D-Scan system from Sphere Ultrafast Photonics that simultaneously measures and compresses the pulse. This is done by inserting a variable dispersion on the pulse, through the movement of two fused silica wedges. This movement is encoded into the measuring head that resolves the spectra. After, using MIIPS technology[10], we are provided with information on the temporal profile of the pulse.

At 1200 mbar we achieved the maximum pulse compression so far: a pulse duration of FWHM= 3.81 fs representing only 1.5 optical cycles. This is represented in figures 4, 5 and 6, where the D-Scan trace is presented along with the retrieved temporal and spatial profiles.

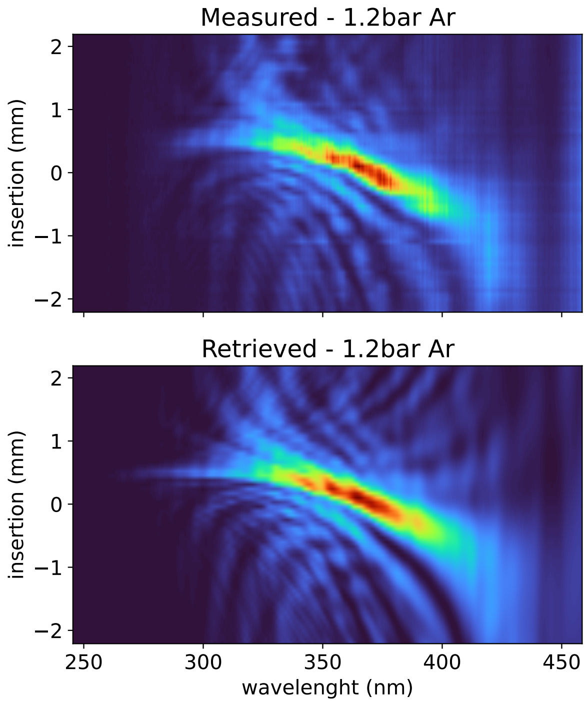

FIG. 4: Measured (top) and Retrieved (bottom) D-Scan traces of the optimal HCF broadening in the VOXEL lab with 1.2 bar of Argon, $E_{in} = 797\mu$J and transmission of 27%.

A quick look into the D-Scan traces reveals a thin but obviously tilted D-Scan trace, this corresponds then to a low Group Delay Dispersion (GDD) but an intense Third-Order Dispersion (TOD). This unfortunately cannot be solved at VOXEL, only material with proper GDD/TOD ratio or with only TOD compensation can correct this. Only with this correction will we be able to achieve FTL pulses.

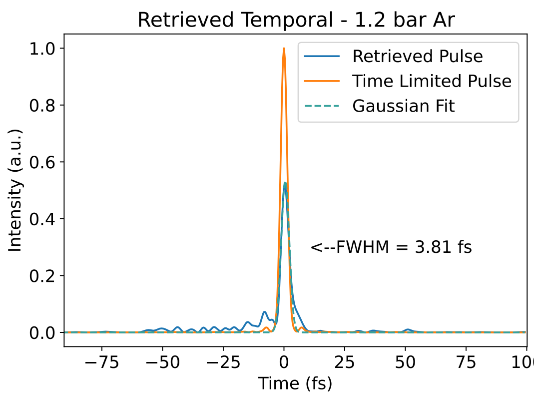

FIG. 5: Temporal retrieval of the optimal HCF broadening in the VOXEL lab.

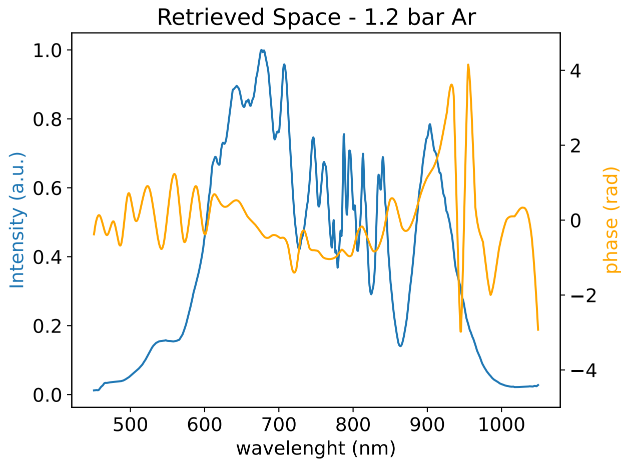

FIG. 6: Spatial retrieval of the optimal HCF broadening in the VOXEL lab.

3. POST COMPRESSION AT LOA

To understand the HCF method of post-compression, we had the privilege of collaborating with colleagues from the *Laboratoire d'Optique Appliquée* (LOA) a part of *Institut Polytechnique de Paris*. More specifically, we collaborated with the Optical-Cycle Physics (PCO) group, created in 2005 and led by Dr Rodrigo Lopez-Martens. The group studies the interaction of high-intensity lasers with matter to follow and control the collective movement of relativistic electrons.

We studied post-compression in both the Hollow-Core Fibre setup and the Multipass Cell. At LOA, 8 mJ are successfully compressed by an HCF with 50% efficiency while the MPC compresses 1.2 mJ with 67% transmission. These results will be used in the design of the VOXEL laser extension where up to 5 mJ will be used for compression.

3.1. Hollow Core Fibre

At the Salle Noire of LOA we found a hollow core fibre with 2.5m and an inner core of 536μm. Just as before, the fibre is differentially pumped with high-purity Helium gas, creating a stable pressure gradient across the waveguide from pressures from $<$ 0.1mbar in the entrance to 2bar at the output. In this setup, the light is, however, circularly polarised to mitigate defects from multi-photon ionisation and self-focusing.

We had the opportunity to analyse how the spectral broadening changed with an increase of pressure inside the fibre. This evolution is plotted in figure 7.

At Lisbon, we have the optimum pressure around 1200 mbar with a resource to Argon. At *Salle Noire*, we find a similar spectral width when employing 1000 mbar of Helium on the output of the HCF. In the Helium-filled fibre, it is clear that a lot less self-focusing and ionisation occurs, as shown by the lack of a "bump" in the 500-600 nm range.

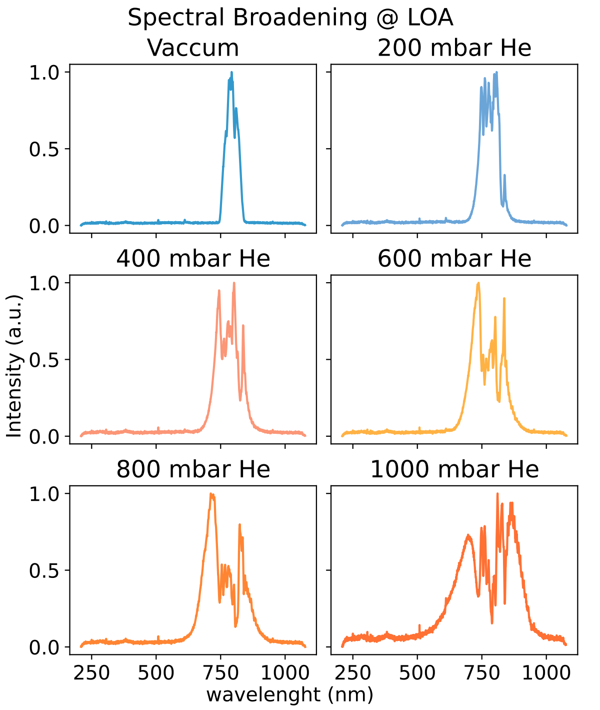

FIG. 7: Evolution of the normalised spectral shapes with increasing gas pressure in the HCF of Salle Noire of LOA.

At a pressure of 1 bar of Helium we achieved the maximum compression point before fatal distortions appeared. The D-Scan traces, along with the temporal and spatial retrieved profiles are shown in figures 8, 9 and 10.

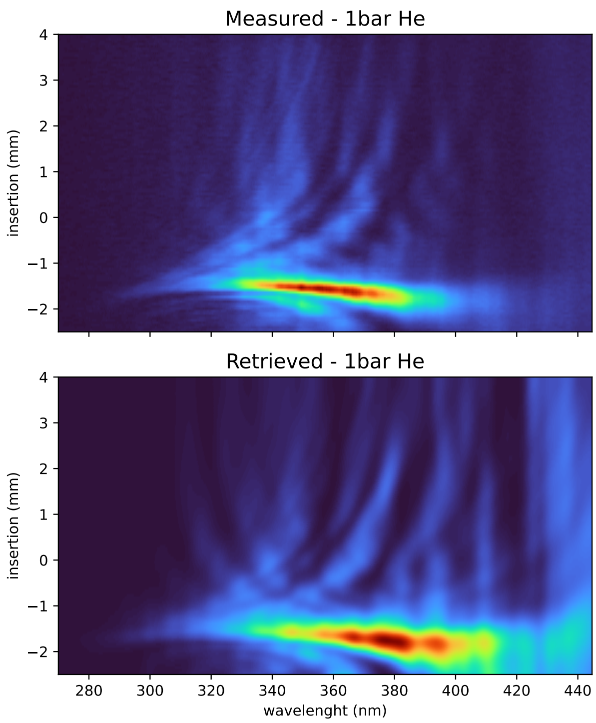

FIG. 8: D-Scan trace of the optimal HCF broadening in Salle Noire 2.0.

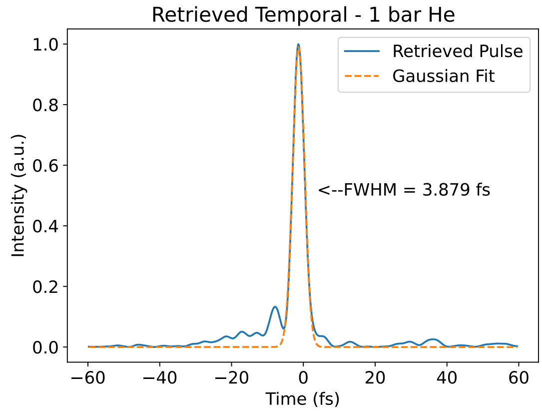

FIG. 9: Temporal retrieval of the optimal HCF broadening in Salle Noire 2.0.

In the case of the setup at LOA, the TOD is already corrected without the need for passive dispersion optics with specific GDD/TOD. This happens since the whole setup is mostly in vacuum, only having a noble gas in the spectral broadening area. Having the beam interact only with fused silica adds a lot of GDD but almost no third-order dispersion, which, as we know is much harder to compress.

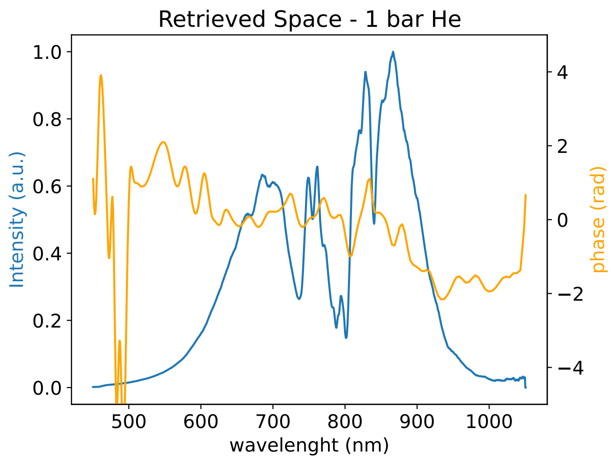

FIG. 10: Spectral retrieval of the optimal HCF broadening in the Salle Noire 2.0.

3.2. Multipass Cell

An MPC consist of a cavity where one can find either a nonlinear crystal or a noble gas. Through multiple passes, allowed by the Herriott-cell structure of an MPC, the spectral broadening occurs in a less intense way. A scheme of an MPC is presented in figure 11.

At LOA the existing MPC uses gas, in particular, Argon as the nonlinear medium. For this reason, the

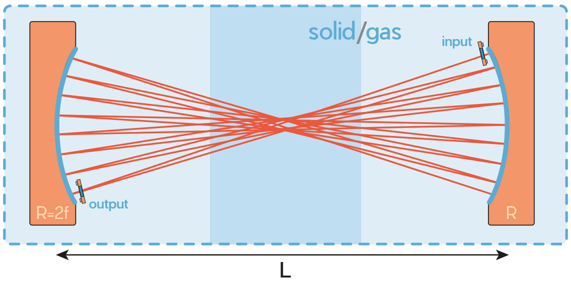

FIG. 11: Schematic layout of a nonlinear MPC. The nonlinear medium is either a bulk slab inserted inside the cell (dark blue) or a gas surrounding the cell (light blue). Adapted from [11].

output spectrum was studied through several gas pressures having always 18 passes in the MPC. This is plotted in figure 12. One can immediately see that the spectrum is much more symmetrical throughout the whole spectral broadening range.

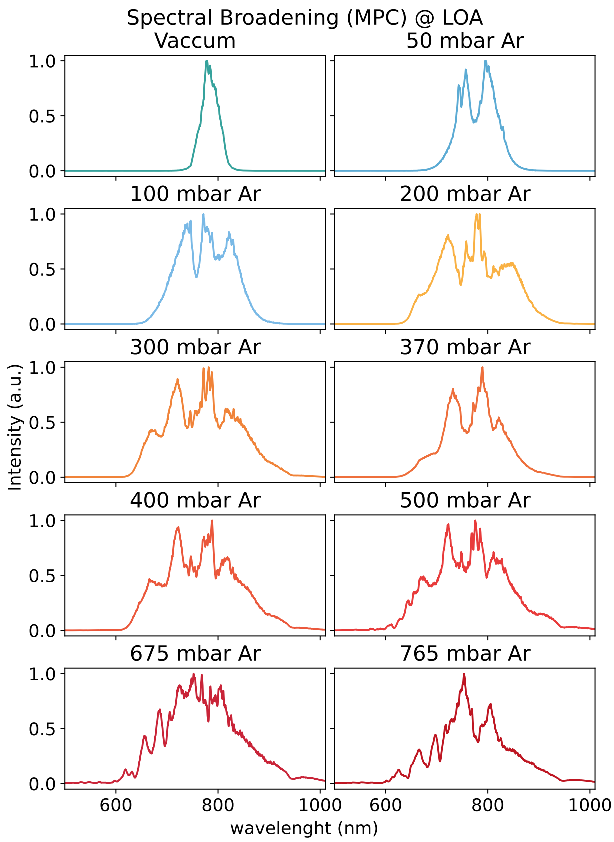

FIG. 12: Evolution of the normalised spectral shapes with increasing gas pressure in the MPC of Salle Noire of LOA.

The MPC is limited by a saturation point after 400 mbar and so we found our optimal pressure (before self-steepening started occurring) at 370 mbar. The experimental and retrieved FROG traces of the post-compressed pulse at the 370 mbar are represented in figures 13 and 14. The retrieved temporal and spatial profiles are plotted in figures 15 and figures16.

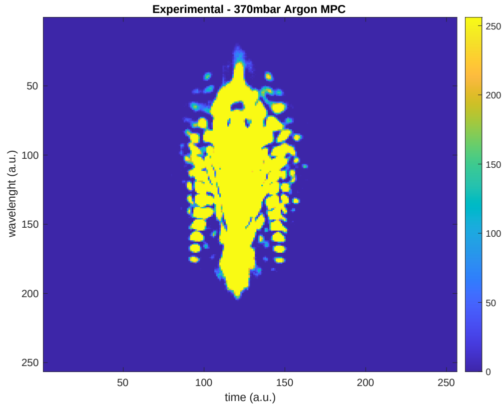

FIG. 13: Experimental FROG trace of the optimal MPC broadening in the Salle Noire.

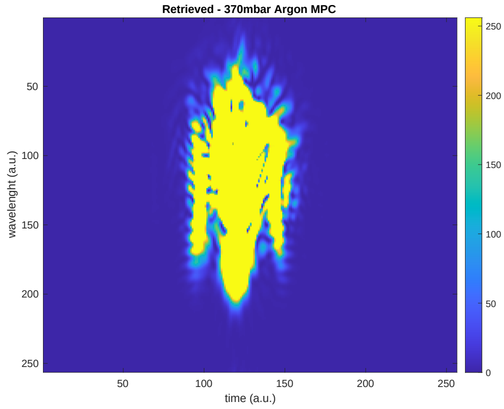

FIG. 14: Retrieved FROG trace of the optimal MPC broadening in the Salle Noire.

The output energy after the MPC is usually around 79% due to the losses that arise from the inherently flawed mirrors, which have a reflectivity of 98.5% for each pass over the entire bandwidth. After the 9 pairs of chirped mirrors that introduce a group-delay dispersion of $378 fs^2$ and the two wedges that also lose energy, the whole post-compression efficiency was measured to be 67%.

3.2.1. Cross-polarised wave generation in MPC

During the visit to Paris, there was also the opportunity to take part in the new experiment being developed by the PCO group [12].

The group developed the first implementation of cross-polarised wave generation (XPW) in an MPC. XPW generation is a well-established technique that is known to improve temporal contrast by three orders of magnitude with up to 33% internal conversion efficiency [13], given that the input beam is spatially filtered and has a super-Gaussian profile.

It was proven then that pulse shortening and temporal contrast enhancement of mJ-pulses can be si-

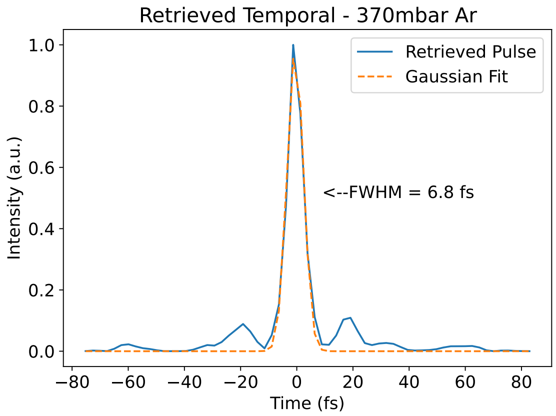

FIG. 15: Temporal retrieval of the beam at optimal MPC broadening in the Salle Noire.

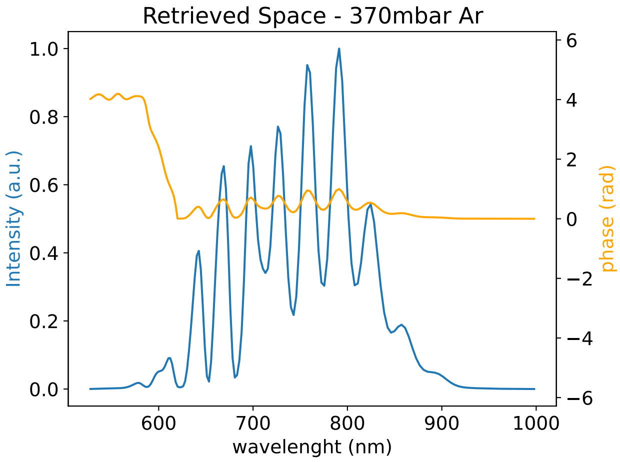

FIG. 16: Spatial retrieval of the beam at optimal MPC broadening in the Salle Noire.

multaneously achieved. In this case, the experiment has 20% global efficiency giving 18.2 fs pulses with an enhancement of measured temporal contrast of at least 3 orders of magnitude.

4. CONCLUSIONS

We successfully achieved almost octave-spanning spectral width. This resulted in the achievement of sub-4fs pulses with minimal group delay dispersion and transmission of 33%. These results represent a powerful tool for table-top single-cycle laser pulses in the VOXEL laboratory. We also installed a new single-cycle beam line to create broadband harmonics.

In LOA we studied what constitutes the state of the art of post-compression into the single-cycle regime, both in hollow-core fibres and multipass setups. We also collaborated in the creation of a novel setup concerning cross-polarised wave generation in multipass cells, achieving simultaneous pulse shortening and temporal contrast enhancement.

Future Work

The installation of the beam stabilisation system already commissioned is crucial for the continuous operation of the post-compression system. This needs to be installed and will require a new, hopefully, less arduous, optimisation of the spectral broadening. An autocorrelator should arrive any day now and will permit a more systematic analysis of the dispersion of the pulse.

After these corrections on the setup, not achievable before due to time constraints and lack of the resources just mentioned, the setup is prepared to be employed in the creation of the isolated attosecond pulses that gave the motivation for this thesis. A novel attosecond holography setup [14] designed with the capability of retrieving sub-fs, 100 nm-scale information has never been demonstrated with true attosecond pulses. This experiment has been planned and should be implemented in the next months.

Finally, work performed at LOA allowed us to study the compression of much higher energy pulses. This will allow the design of a new post-compression station at the expanded VOXEL laboratory, currently under design.

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