



Development of a dual comb spectroscopy experiment in the ultraviolet range

Abel Feuvrier

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Development of a dual comb spectroscopy
experiment in the UV range

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Abstract

In the context of climate change, air quality studies and atmospheric physics and chemistry, the development of sensitive and precise trace gas detection techniques is needed. DCS spectroscopy has demonstrated its applicability to the remote detection of atmospheric trace gases. Indeed, the short acquisition feature of this spectroscopy technique can outrun atmospheric turbulences fluctuations and its high resolution allows the simultaneous detection of atmospheric trace gases. In particular, its use in the UV range could allow to detect radical species that are critical to understand the physico-chemistry of the atmosphere. This work aims at developing a UV-DCS experiment, in the scope of atmospheric trace gas detection.

Dual Comb spectroscopy uses the beating of two mutually coherent optical combs with slightly different repetition rates. This beating maps the optical spectra at lower frequencies (radiofrequency (rf) domain) that fall within the bandwidth of standard photodetectors. Hence, the optical absorption lines are imprinted on the rf spectrum: optical spectroscopic information can be retrieved from rf temporal measurements. DCS allows to retrieve high resolution spectra (up to 100 MHz) with high sensitivity (up to ppb) in short acquisition times (up to ms). the combination of these features makes DCS particularly relevant for broadband spectroscopy with applications from molecular metrology to atmospheric remote sensing.

The mutual coherence between the two combs of our experiment relies on the use of a bidirectional laser cavity. This cavity emits two optical combs, with the same pump laser, gain medium and cavity. Thus their mutual coherence is intrinsically high, and they can be used for DCS without any active stabilisation. A solid-state titanium-sapphire (Ti:Sa) laser was used to ensure high output power and low intensity noise. Moreover, the Ti:Sa emission range allows to reach the near UV by a single harmonic generation stage, limiting the phase coherence loss. The Kerr-lens mode-locked laser cavity was homebuilt prior to this work. In this work, the two generated optical combs were further characterised, in terms of beam size, pulse length and temporal shape, spectral width, and repetition rate difference between the two combs (Δf_{rep}). The link between Δf_{rep} and experimental parameters of the cavity, and with its other output quantities, was studied. We showed quantitatively that the microscopic origin of Δf_{rep} can be explained by the self-steepening of the pulses in the Ti:Sa crystal.

Using this cavity, IR DCS measurements were performed. The signal/noise ratio (SNR) of the previous proof-of-concept spectroscopic measurement on the A band of O_2 around 760 nm was improved by a factor two. An upper bound of 1 GHz on the resolution of the spectrometer was retrieved. This SNR improvement allowed to bring out asymmetric, Fano-type lineshapes on the O_2 lines. To better understand the origin of these lineshapes, a DCS simulation was coded. It showed that the Fano-type lineshapes can be caused by Δf_{rep} fluctuations.

A proof-of concept UV-DCS measurement was performed on atomic Cs lines, with a resolution of the order of 50 GHz. To test if this DCS experiment is relevant to detect atmospheric trace gases, we performed NO_2 spectroscopy using a in-lab cell. We retrieved NO_2 spectroscopic signature over a spectral range of 3 nm ($2\text{e}4\text{ cm}^{-1}$, 6 THz) centered at 389 nm ($2.57\text{e}6\text{ cm}^{-1}$, 771 THz) in 4 ms. This experiment is among the first demonstrations of UV-DCS with resolved absorption lines. The particularity of this work lies on the use of Ti:Sa cavity, that allows high versatility in terms of spectral range and position with high output power.

The perspectives of this work include the development of post-correction algorithms and adaptive clock strategies to further increase the signal/noise ratio of the spectroscopic results.

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Chapter 1

Introduction

Introduction

In the context of climate change and growing atmospheric pollution, atmospheric trace gas concentration ranging methods are needed. Dual comb spectroscopy (DCS) is a method fit for trace gas concentration retrieval, because of its combination of several features: broadband spectral measurements and high spectral resolution to measure several gas simultaneously and identify their individual spectral lines, and fast acquisition times to be immune to turbulence-related biases in the atmosphere. However, DCS is not much implemented yet in the UV range, where many pollutant trace gas (NO_x , BrO, HONO, etc.) have their highest absorption cross section.

This work aims to develop a DCS experiment in the UV range, using a homemade free-running bidirectional ring titanium-sapphire (Ti:Sa) cavity, in the scope of an application for atmospheric trace gas detection. A prior study evaluated the feasibility of this approach.

To achieve this objective, an experimental characterisation of the bidirectional cavity was done and a second harmonic generation (SHG) setup was used to reach the UV range from the cavity near-IR emission. To better understand experimental spectroscopic results, an analytical modelisation of the DCS method was developed and used to implement a numerical DCS simulation.

This chapter introduces the context and motivation behind this experiment. The first section recalls the importance of trace gas detection in the context of climate change and air quality and gives a quick review of the existing detection techniques. The second section focuses on optical frequency comb spectroscopy techniques. The third presents DCS and provides an overview of its application in the UV range. Finally, the fourth section highlights the use of a freerunning bidirectional ring Ti:Sa cavity for this work.

1.1 Trace gas detection in the context of climate science and air quality

1.1.1 Importance of the detection of trace gas

Climate change and air quality are major challenges. The 2023 report of the intergovernmental panel on climate change (IPCC) [1] gives a survey of the enormous stakes related to this issue, environmental [2], social [3], and political [4]. In this context, atmospheric science is needed. In particular, the study of trace gas is relevant to climate and air quality issues. Pollutant trace gas are emitted by human activity. Their concentration are especially high in urban areas, where the population keeps increasing as the air quality keeps decreasing due to pollution [5]. Trace gas concentration is linked with acid rains, public health and mortality [6]. In particular, radicals such as NO_2 were found to have harmful effects on public health [7–10].

Moreover, trace gas, especially the radical HO, participate to regulate the physico-chemistry of the atmosphere, through redox reaction that regulate the concentration of molecules such as CH₄, CO or VOC (volatile organic compounds) [11,12].

To better monitor the concentration of trace gas and better understand their dynamics, reliable sensing methods are needed [13,14].

Many relevant trace gas feature absorption lines in the near UV frequency range, as illustrated on figure 1.1.

Thus, the remote sensing of these trace gas calls for a sensing technique that is:

- usable in the near UV range
- able to detect and discriminate several species over this spectrum
- fast, to be free of the measurement bias related to atmospheric fluctuations.

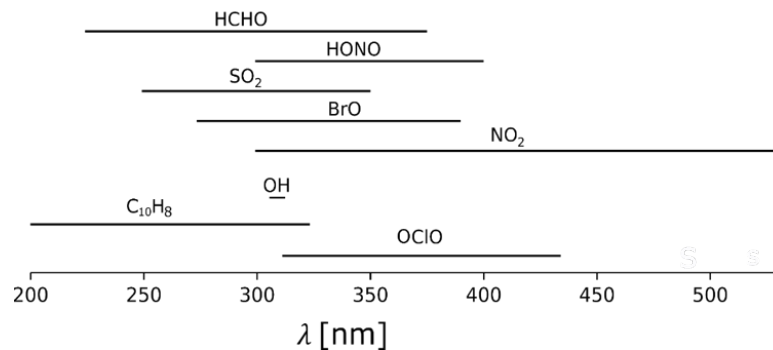


Figure 1.1: Near UV frequency range with absorbtion bands range of atmospheric radicals (figure taken from [15])

1.1.2 Trace gas detection techniques in the UV range

Several complementary methods of trace gas detection in the UV are available, as surveyed in [16–18].

Non-optical techniques

Non-optical methods are usually less costly than optical ones, while still featuring sensitivity and fast acquisition times.

Laboratory analysis methods such as chromatography [19] use the affinity of gases with custom solid or liquid phases to detect them in an extracted gas sample. To extract the sample, chemical-based [20] or photo-ionisation devices [21] can be used. These methods are sensitive and have high molecule selectivity. However, they are difficult to integrate.

Other methods are integrated on portable devices, enabling portability and real-time measurement. The gas concentration can be retrieved from the variation of a physical quantity in presence of the gas: electric resistivity [22], acoustic waves propagation parameters [23], or chemically-triggered quantities such as colour, fluorescence or electric current [20].

Optical techniques

Optical methods are complementary to these methods. Though they are more complex, they are easier to implement, allow in-situ analysis and faster acquisition time [18], which is necessary for our application. They are surveyed in [16,18,24–28].

Non-absorption-based techniques can detect very low levels of trace gas concentration. Detectors based on chemiluminescence are especially used to detect NO_x [29], while UV-fluorescence is used for sulphur-related compounds [30]. In both cases, the molecules absorb UV radiation and emit at lower frequencies. This latter emission is detected by photodetectors. The gas concentration can then be retrieved through calibration of the instruments.

These techniques are very sensitive to dedicated molecules and can suffer for bias from other compounds contained in the probed air, while our goal is to ultimately be able to simultaneously detect different gaseous compounds, as depicted in figure 1.1.

Absorption-based methods are more agile in terms of molecules selectivity. Cavity ring-down spectroscopy (CRDS), allow very high sensitivity trace gas measurement using compact setups [31, 32]. This technique relies on high-finesse optical cavities, in which laser sources are injected. The concentration is retrieved from the measurement of the optical decay of the output of the cavity after the source is turned off. However, this technique cannot be used on long atmospheric open-path, as it needs an extracted sample of the characterised medium. This limit prevents its use in the open path atmosphere.

On the other hands, remote sensing techniques allow free space measurements. Differential absorption light detection and ranging (DIAL) allows space resolved, free space concentration retrieval [33–37]. This technique relies on the time-resolved detection of the retrodiffused light on an open path to map the spatial gas concentration. Differential optical absorption spectroscopy (DOAS) does not allow spatially-resolved measurement like DIAL, but reaches higher sensitivity [38–40]. This technique relies on spectral measurements. The spectrum of a beam that travelled through an optical path can be decomposed into a baseline and absorption features, as given by Beer-Lambert’s law:

$$I(\lambda) = \underbrace{I_0(\lambda)}_{\text{baseline}} \underbrace{e^{-\int_{\text{opt. path}} \sum_i \sigma_i(\lambda) C_i(x) dx}}_{\text{absorption feature}} \quad (1.1)$$

with $I_0(\lambda)$ the intensity taking into account every phenomenon but absorption (mainly diffusion), σ_i the absorption cross-section and C_i the concentration of the species contained in the medium indexed over i . Hence, the concentration can be retrieved from the light intensity decrease.

This latter technique is widely used for trace gas detection [41,42], in the UV range [43–46] and meets our requirements of ability to detect and discriminate several gas and of fast acquisition, thus it was used in this work.

1.2 Optical frequency comb spectroscopy

A frequency comb is a field whose spectrum is composed of evenly-spaced, phase-coherent discretised frequencies. The corresponding temporal field is a train of pulses.

A comb is characterised by several quantities, some of them presented in figure 1.2. In particular, the frequencies of the comb teeth are determined by the repetition rate f_{rep} and the offset frequency f_0 .

1.2.1 Optical frequency combs development

Using the above definition, any mode-locked laser produces an optical frequency comb (OFC). Hence, the first OFC can be traced back to the first mode-locked lasers, from active [47] to Q-switch [48] and passive continuous-wave (cw) [49] emissions. The pulse durations were then shortened (thus the

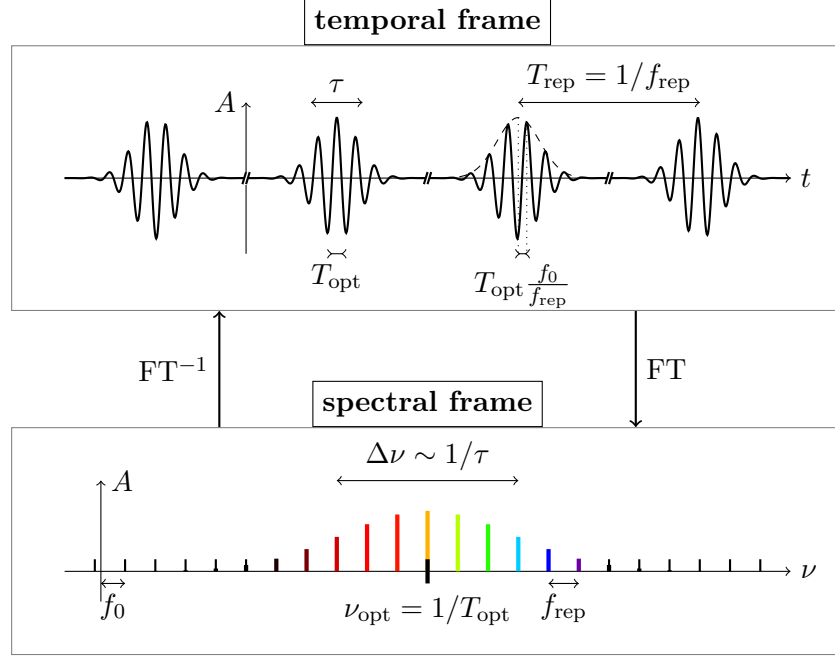


Figure 1.2: Temporal and spectral representations of a frequency comb with characteristic quantities: pulse duration τ , optical frequency T_{opt} and repetition period T_{rep} in the temporal frame, linked respectively with spectral width $\Delta\nu$, central frequency ν_{opt} and repetition rate f_{rep} in the spectral frame. Other relevant parameters, such as phase-carrier offset and linewidth of the comb teeth, are not represented here for the sake of simplicity but will be discussed later.

spectral width widened), from picosecond using dye lasers [50] to femtosecond, using Kerr-lens mode-locked solid-state lasers such as titanium-sapphire (Ti:Sa) cavities [51, 52]. Since then, other types of solid-state lasers have been used to produce frequency combs [53–56] and solid-state lasers remain relevant tools for OFC production, due to their high power and broad emission spectra [57–60].

In 2000, nonlinear optical fiber technology [61–63] enabled to widen the comb spectrum from a Ti:Sa cavity over one octave. This spanning over one octave allowed to stabilise the offset frequency f_0 thanks to the $f-2f$ stabilisation technique, introducing self-referenced mode-locked lasers [64–66]. This breakthrough, combined with microwave-optical frequency conversion technology, unlocked the spectral coverage of virtually all of the spectrum [67] from mid-IR [68, 69] to XUV [70–72]. Fiber-based optical combs proceeded to become the most widespread source of frequency combs [73–77]. During this fast and fruitful expansion of the frequency comb field, the 2005 physics Nobel prize was awarded to Hänsch and Hall for their contribution [78, 79].

The difference frequency generation (DFG) and optical parametric oscillators (OPO) technologies then enabled sources spanning over several octaves [80–82] in the mid-IR, extending the available spectral coverage up to 27 μm wavelength [83], while high harmonic generation techniques in gas extended it in the XUV down to sub-20 nm wavelength [84, 85].

Finally, electro-optic modulators provided sources with fast tunable repetition rates, high sidebands power, and reconfigurability of the comb spectrum [86, 87].

During the past 20 years, tremendous progress has also been achieved on OFC with lower size, weight and power (SWAP) [88–91]. New laser technologies enabled OFC to become photonics-integrated and chip-based [92], using quantum cascade laser (QCL) [93, 94], mode-locked integrated external cavity surface-emitting lasers (MIXSELs) [95, 96] or microresonators [97, 98].

1.2.2 OFC applications

Applications of OFC range from very fundamental research to applied technology.

In fundamental research, stabilised OFC have revolutionised metrology [99], becoming the most accurate available time and frequency measuring tool by reaching relative precision up to 10^{-18} [100–102], allowing to test general relativity through geodesy [103], synchronising clocks with a relative precision of 10^{-15} at the scale of a continent [104] and even pushing for a new definition of the second [67].

OFC were applied, among others surveyed in [105], to calibration of astronomical spectrographs [106, 107], quantum computing [108], optical communication [109, 110], light detection and ranging (LIDAR) [111, 112], development of attosecond science [113], trace gas remote sensing [114], and, of course spectroscopy, for which several OFC-based techniques were developed.

1.2.3 Single frequency comb spectroscopy techniques

Optical combs feature large bandwidth and high spectral resolution, which makes them a powerful spectroscopic tool. Several techniques were developed to harness their potential, surveyed in [115, 116], some of them schematised in figure 1.3.

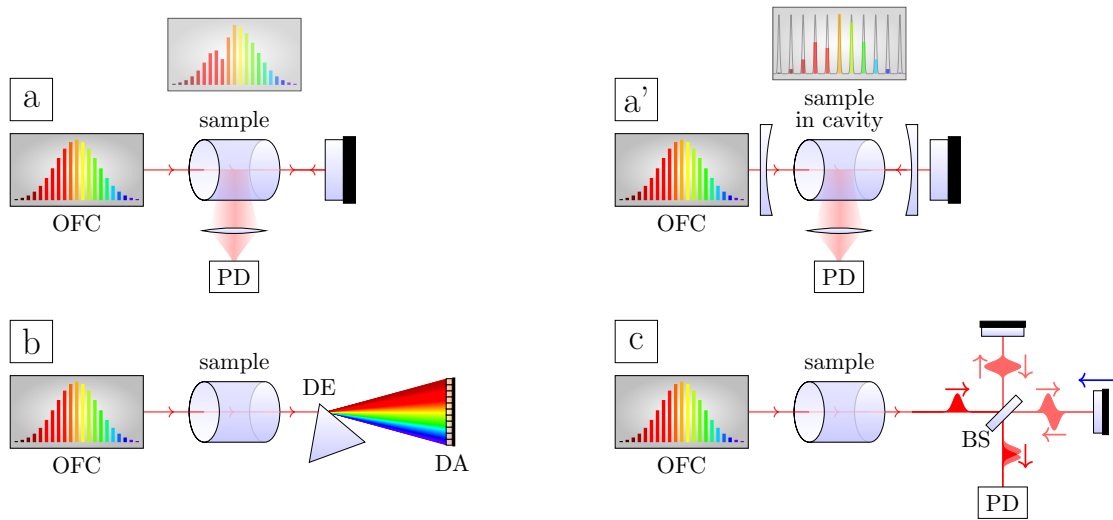


Figure 1.3: Schemas of some spectroscopy techniques using optical frequency combs (OFC). **(a)** Direct fluorescence/Ramsey spectroscopy. The OFC is sent through the absorbing sample and the resulting fluorescence of the sample is detected by a photodetector (PD). The OFC at the output of the cavity is represented over the cavity, with an absorption line. In the case of Ramsey spectroscopy, only some pulse pairs are selected and amplified. **(a')** The coupling of the comb with a cavity enables to filter the comb teeth not matching of the cavity modes and enhance the sensibility of the spectrometer. The comb and the cavity can be tuned or detuned (Vernier). **(b)** Direct OFC spectroscopy using a dispersive element (DE) that separates spatially the different comb frequencies. The resulting dispersed beam is detected by a detector array (DA), each detector corresponding to a frequency range. **(c)** Fourier-transform (FTIR) spectroscopy. The absorbed OFC is sent through an interferometer with one moving arm. The output intensity of the interferometer is recorded as the length of one arm varies. The recording gives interferograms. The Fourier transform of these interferograms give the spectrum of the absorbed OFC.

Direct OFC spectroscopy is the simplest approach to OFC spectroscopy [117]. Two-photon excitations techniques allow to perform spectroscopy without a spectrometer, but are limited to spectra with few transitions [50, 118]. Using dispersion-based spectrometers, more complex spectra can be

retrieved, with resolutions down to 100 MHz thanks to the virtually imaged phase array (VIPA) technique [119–121]. Fourier-transform spectroscopy with Michelson-based interferometers can achieve even better resolution, but at the expense of the acquisition time [122,123]. Complementary to these techniques, dual comb spectroscopy combines fast acquisition time and high resolution, as will be seen on next section (section 1.3).

The coupling of OFC spectroscopy with optical cavities allows to greatly enhance its sensitivity and precision, as surveyed in [124]. Direct OFC spectroscopy [125], FTIR [126] and DCS [127] have been successfully coupled with cavities, and performances of techniques such as cavity ring-down spectroscopy (CRDS) [128,129] or Vernier [130–132] keep improving.

1.2.4 OFC spectroscopy in the UV range

UV optical frequency combs (UVOFC) was used in Ramsey spectroscopy to perform precision metrology on noble gas transitions [133,134]. Fluorescence spectroscopy was also performed using UVOFC [135] for atomic spectroscopy. Direct spectroscopy with UVOFC allowed to perform electronic transitions spectroscopy and quantum electrodynamics tests [84,136]. Recently, UVOFC were applied to astrocombs [137], and UVOFC were accurately characterised using spectroscopy [138].

More and more broadband UVOFC sources are being developed and used for spectroscopy [139]. Some recently developed sources span over several octaves, which is promising for integration and spectral expansion [140,141].

The development of XUV OFC spectroscopy in the future holds great potential for precision metrology and attosecond science [142,143], as well as for accurate measurement of physics fundamental quantities [144].

1.3 Dual comb spectroscopy

1.3.1 Principle of DCS

Dual Comb Spectroscopy (DCS) is a spectroscopy technique that relies on the interference of two frequency combs with a slightly different repetition rate. This repetition rate difference is noted Δf_{rep} . Due to this Δf_{rep} , the beating between the two optical combs produces a third comb at much lower frequencies (radiofrequencies rf). These frequencies fall within the bandwidth of standard photodetectors. Hence, the measurement of the temporal trace of the beating (interferograms) allows to retrieve the rf spectrum by Fourier transform, and the rf spectrum allows to retrieve the optical spectrum. This principle is illustrated in figure 1.4. A more developed formalism, based on temporal description, will be developed in chapter 2.

The main advantage of DCS is that it combines the high resolution and broadband spectral coverage of the frequency combs with a high acquisition speed [116,145]. Whereas the resolution of grating-based spectrometers is limited by the dispersive power of the dispersive element, the resolution of DCS spectrometers is only limited by the repetition rate (and can even go beyond using spectral interleaving [146,147]). As for the acquisition speed, whereas the acquisition speed of Michelson-based spectrometers is limited by the frequency of the moving part (acoustic frequencies), the acquisition speed of DCS is only limited by the Δf_{rep} parameter (radiofrequencies) (and even this limit can be unlocked [148]).

This combination makes it especially fit for atmospheric trace gas detection, as discussed previously in section 1.1.

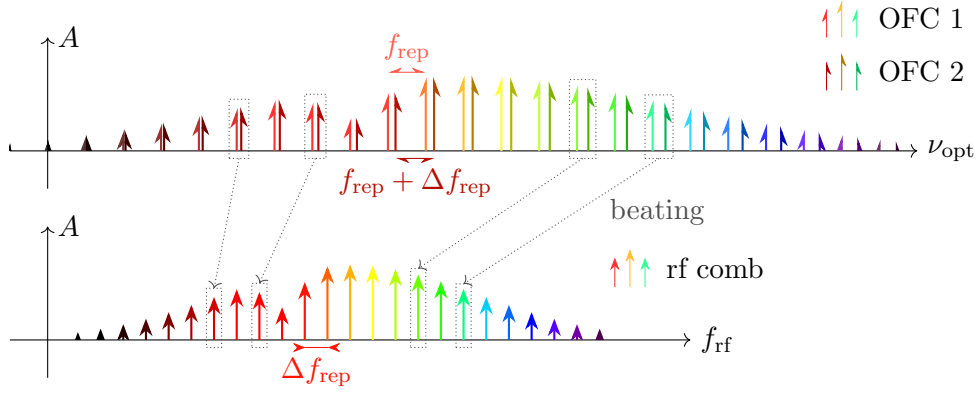


Figure 1.4: Schema of the principle of DCS. Two optical frequency combs (OFC) with slightly different repetition rates beat. Their beating produces a third, radiofrequency (rf) comb, that falls within the bandwidth of standard photodetectors. The repetition rate of this rf comb is the difference between the repetition rates of the two optical combs. Any absorption feature appearing on the OFC is imprinted on the rf comb: for example here, an absorption line in the red frequencies.

1.3.2 Brief overview of DCS

History of DCS

DCS was first proposed in 2002 [149]. The first results were published in 2005 in the mid-IR range [150]. The spectral coverage of the technique was then extended to the whole visible to mid-IR range with increasingly broadband sources [122, 151–154] following the progress of OFC technology [145]. The resolution THz was also covered but falls out of the scope of optical combs, and UV will be treated in a next subsection (subsection 1.3.3).

The coupling of DCS with cavities was achieved in 2010 [155] and proceeded to enhance sensitivity, which is especially useful in remote sensing [156–159].

The field keeps maturing, with recent breakthroughs such as free-form DCS that allows to reduce the acquisition time while conserving the signal/noise ratio (SNR) [148], or use of quantum technology to reduce shot noise [160, 161].

Spectral resolution

The ultimate resolution of DCS spectra depends on the repetition rate of the combs, as can be seen on figure 1.4. This quantity depends on the OFC that is used and can range down to kHz [162, 163]. However, not all dual-comb sources allow to retrieve comb-resolved spectra, depending on the mutual coherence of the two combs that are used for DCS, as will be discussed in next subsection (subsection 1.4.1).

Applications of DCS

DCS has been applied to various fields, from molecular metrology to enhance molecular spectra database [165, 166] to microscopy [167], chemical kinetics study [168], plasma characterisation [169], solid characterisation [170] and medical imagery [171].

In particular, DCS is used for remote sensing of atmospheric trace gas. DOAS methods are used with high sensitivity and resolution up to 100 MHz [172–175], which allows to discriminate different gas [176, 177], over long paths up to 100 km long [178]. DIAL methods were recently implemented as well with GHz spectral resolution [179].

The feasibility of the DOAS method over km-long path with our UV-DCS setup was studied quantitatively in [180].

1.3.3 State of the art of UV-DCS

The use of DCS in the UV range (UV-DCS) is particularly interesting for remote sensing of atmospheric trace gas, as explained in section 1.1. Roadmaps to achieve UV-DCS were proposed from 2020 [180,181]. The first spectroscopic results were published in 2024 on formaldehyde [182], Fe I and Fe II [183], cesium and rubidium [184], while UV dual-comb spectrometers were published with characterisation and prospects of application to plasma spectroscopy [185], remarkable spectral coverage [186] and XUV [187]. Some of the main features of these reported experiences are summarised in table 1.1.

Publication (1 st author)	Laser source	Average power	Central wavelength	Spectral coverage	Spectral resolution	Acquisition time
Fürst [182]	Yb fiber + SHG twice	-	344 nm	14 nm	134 GHz	100 μ s
McCauley [183]	Yb fiber + SHG twice	-	265 nm	0.2 nm (sgl) 3 nm (tot)	1.2 GHz (avg)	200 μ s (sgl) 1.1 s (avg)
Xu [184]	AOM + SHG twice	1 nW	388 nm	0.03 nm (sgl) 1.76 nm (tot)	500 MHz	255 s (tot)
Chang [185]	-	-	386 nm	-	100 MHz	-
Muraviev [186]	Cr:ZnS + HHG	1 mW 0.1 mW	391 nm 333 nm	38 nm 17 nm	80 MHz	1 s (tot)
Li [187]	Yb:YAG thin-disk	300 mW	265 nm	4 nm	71 MHz	-

Table 1.1: Features of UV-DCS publications. When publications feature averaged spectra, (sgl) stands for single acquisition and (avg) for averaged. When publications feature tunable laser source, the spectral coverage is noted (sgl) for a single acquisition and (tot) for the total coverage of the source. In, case of information we do not have access to, cells are left empty.

1.4 Bidirectional Ti:Sa ring cavity

1.4.1 Mutual coherence between two combs

Dual Comb Spectroscopy relies on the detection of interference between two frequency combs. This interference is dependent on the mutual coherence level between the two combs. Thus, the two frequency combs used for DCS must have a high degree of mutual coherence. To ensure this coherence, several tools exist, schematised in figure 1.5 and briefly reviewed hereafter.

Most DCS setups use stabilised lasers to ensure mutual coherence between the two combs. Using feedback loops that correct in real-time the emission of each laser with respect with one single reference, one can achieve very high spectral resolution (MHz or less) and mutual coherence. The mutual reference can be an optical continuous-wave laser, a lower frequency light source, an atomic clock, etc. [153,154,164,186,188,189].

These feedback devices allow high performances but are cumbersome to set up.

In the case where the emission is not stabilised enough to achieve a sufficient degree of mutual coherence, one can also use adaptive detection. Its principle is presented hereafter and a modelisation is given in section B.

Since DCS relies on the beating between the two combs, only relative phase noise has an effect on the temporal trace and has to be corrected (see appendix B). Thus, to tackle phase noise, one must sample relative phase noise. To do so, one can make one tooth of each direction beat with the

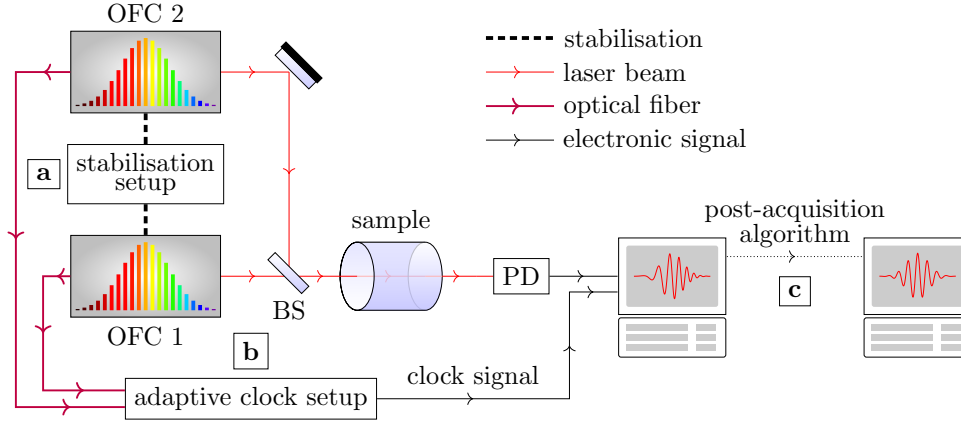


Figure 1.5: Schema of a DCS experiment with coherence-enhancing tools. Two optical frequency combs (OFC 1 and 2) are superimposed on a beamsplitter (BS), go through an absorbing sample and are focused on a photodetector (PD). The output signal of the photodetector is then sampled on a computer. To enhance the mutual coherence of the two optical combs, they can be stabilised using a common reference (a), the acquisition rate can be adapted according to the temporal fluctuations of the combs using an adaptive clock setup (b), and the acquired signal can be treated with a post-acquisition algorithm (c).

same stable, spectrally narrow laser. This samples the absolute phase noise of each direction. Then, by making these two beating signals beat, one obtains the relative phase noise of the two combs. By referencing the acquisition clock with this signal, the relative phase noise vanishes.

(In the analogy where DCS is compared to a pump-probe experiment, adaptive correction can be seen as a jitter correction, namely a correction for the shift of the "clock" ("pump") signal from rigorous periodicity.)

Adaptive detection can improve the Signal-to-Noise Ratio (SNR) of DCS measurements by several orders of magnitude, leading to better resolution and sensitivity [151].

The limits of these devices is that the beating signal must have a signal/noise ratio (SNR) high enough to be manipulated [190]. The threshold depends on the processing of the beating signal, which can include filtering, amplification and more beating.

The aforementioned tools for mutual coherence enhancement are to be put up before or during the acquisition. After the acquisition, post-correction algorithms can be used.

The principle of most post-correction algorithm is to sample phase noise on each interferogram of the temporal trace and to remove it numerically, to be able to average the interferograms coherently. Such methods have been used on Fourier-transform spectroscopy interferograms long before the emergence of frequency combs spectroscopy, with algorithms that can correct the fluctuations of one spectral tooth (namely a continuous-wave laser) [191]. In the case of DCS, one must take into account the fluctuations of the two degrees of freedom of the comb (Δf_0 and Δf_{rep}). To do so, one can use ambiguity maps, which allows to sample both Δf_0 and Δf_{rep} noise [192, 193].

The limits of these algorithms is that they cannot retrieve phase noise in cases where the spectral fluctuations exceed half of the repetition rate of the comb.

Finally, the mutual coherence between the two optical combs can be made intrinsically high without any stabilisation, adaptation or correction device, using a bidirectional cavity as the source of both combs.

In a bidirectional cavity, the two emitted beams share the same cavity, pump laser and gain medium, thus they have an intrinsically high degree of mutual coherence. The dual emission can be achieved through polarisation multiplexing [194–196], spatial multiplexing [154, 197–199], wavelength multiplexing in the gain medium [200, 201] or use of a ring laser cavity [15, 202–204], as surveyed in [205].

The mutual coherence between the two beams emitted by a bidirectional cavity is limited by the relative noise between the two beams. This relative noise can be corrected using the aforementioned coherence enhancement processes.

In our team, a freerunning bidirectional cavity was built, characterised and used for DCS in the IR range [15]. The choice to use a bidirectional cavity was motivated by the absence of cumbersome stabilisation device.

In this work, this cavity was further characterised (see chapter 3) and used for DCS in the IR and the UV ranges. Moreover, correction algorithms were used and an adaptive clock setup was started (see chapter 4).

1.4.2 Ti:Sa for power, low noise and spectral coverage

Titanium sapphire (Ti:Sa) laser cavities were involved in many breakthroughs in the development of laser science and spectroscopy, as surveyed in [206]. Their main advantages are high output power, low noise and broadband spectral coverage, which are important features for atmospheric trace gas remote sensing, as discussed in section 1.1. In particular, the use of a bidirectional Ti:Sa cavity for broadband DCS had already been reported by Ideguchi et. al. in 2016 [204].

Moreover, in our particular case, the targetted spectral range can be reached through second harmonic generation (SHG) and third harmonic generation (THG) from the gain spectral range of a Ti:Sa crystal, as illustrated in figure 1.6. This single-step UV generation process from our cavity is interesting for our measurements because coherence decreases with the order of the harmonic generation [207].

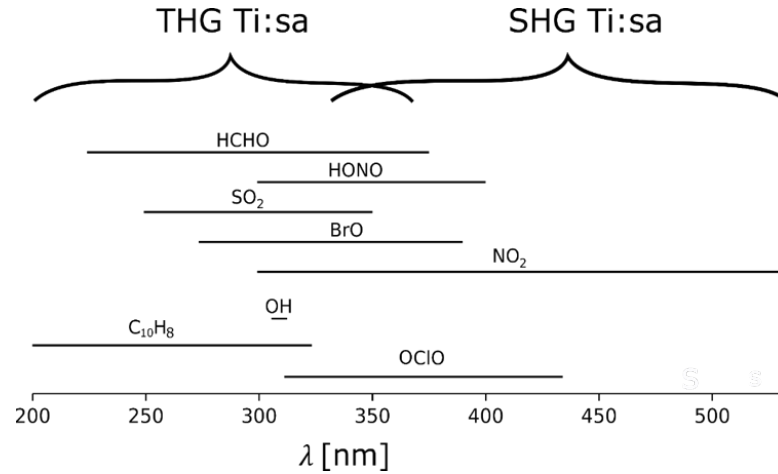


Figure 1.6: Near UV frequency range with absorption bands range of atmospheric radicals and ranges reachable by second harmonic generation (SHG) and third harmonic generation (THG) of a Ti:Sa laser cavity (figure taken from [15])

Conclusion

Trace gas remote sensing is important for climate science and air quality studies. This remote sensing can be done through spectroscopy. The absorption spectrum of many trace gases feature absorption lines in the near UV range, which calls for a fast, broadband, high-resolution spectroscopy technique in this range. UV-DCS meets these requirements.

To use DCS, two mutually coherent frequency combs are needed. To ensure this mutual coherence, a bidirectional cavity can be used. In particular, Ti:Sa cavities ensure broadband spectral coverage, high output power and low noise.

A more accurate modelisation of DCS will be developed in chapter 2. The characterisation of the bidirectional Ti:Sa will be developed in chapter 3. Finally, the DCS results will be presented in chapter 4.

Chapter 2

UV-DCS methodology

Introduction

This chapter aims to describe quantitatively the UV-DCS measurement methodology through the modelisation of our experimental data.

The main output of our experimental detection setup is the temporal trace of the filtered and amplified output signal of a photodiode, on which two frequency combs are focused. A typical output is presented on figure 2.1.

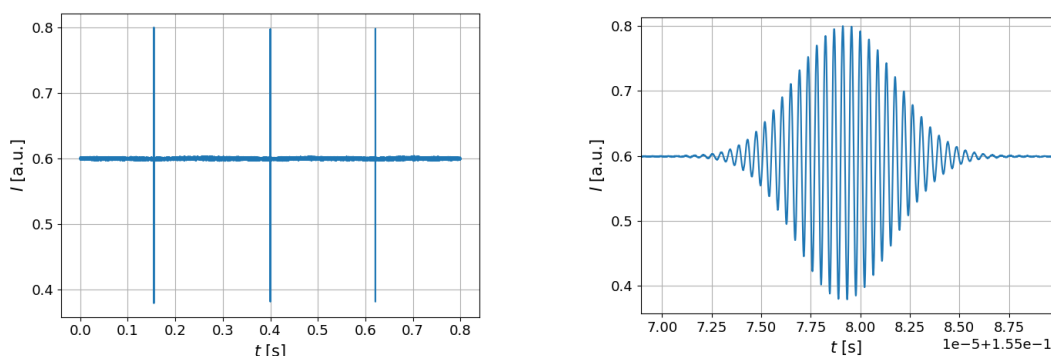


Figure 2.1: Typical output of our detection setup: several consecutive interferograms (left) and zoom on one interferogram (right)

This output presents a baseline, a succession of interferograms, and a free-induction decay term, that is not visible on the graph because of its relatively small amplitude compared with the center-burst, but that can be brought out by spectral analysis. The modelisation of all of these features is developed in this chapter.

The first section aims to develop a model for the experimental time trace and the corresponding spectrum.

First, the detection of a pair of pulses after their propagation through an absorbing medium is modelised at the femtosecond time scale. On a broader time scale (one interferogram, ~ 10 μ s), the detection of a train of pulses and the corresponding spectrum are then modelised as well. Finally, the total interferometric signal is modelised (~ 1 s scale). The hypotheses on which this first modelisation is built upon are detailed along the development.

The second section focuses on the retrieval of optical spectroscopic data from the analysis of these experimental temporal traces.

A mathematical formulation of the problem is proposed, then the different cases that can occur depending on the pulses parameters are treated. A general formula is finally given.

The analytical model that is developed accounts for some deviations from the ideal case: carrier-envelope phase, chirped pulses. However, some other deviations from the ideal case are not treated analytically in this work, especially Δf_0 and Δf_{rep} temporal fluctuations. To modelise them anyway, a simulation was developed, based on the analytical model. Using it, the fluctuations of Δf_{rep} and its consequences on the spectra at the scale of one interferogram are simulated. The qualitative results that are obtained were used in the interpretation of experimental spectra in chapter 4.

Finally, a short description of the Second Harmonic Generation (SHG) process, that enables to reach the near UV range from our Ti:Sa emission range, is given, insisting on the experimental consequences in our case (rather than a complete fundamental description of the process).

2.1 Experimental temporal trace and spectrum

The objective of this section is to modelise our experimental data (temporal trace and corresponding spectrum) analytically. To do so, a modelisation of the combs, the absorption process they go through during their propagation, their detection, and the detection of their beating, is developed.

2.1.1 Propagation of the field and thin medium approximation

This subsection aims to fix the notations that will be used afterwards concerning the field and the diluted medium. Complementary notations and strict definitions can be found in appendix A.

The medium's response to a propagating electric field is characterised by its susceptibility $\hat{\chi} = \sqrt{1+n}$ with n the optical index. The electric field in the absorbing medium obeys the wave equation:

$$\Delta \mathbf{E} - \frac{n^2}{c^2} \partial_t^2 \mathbf{E} = 0 \quad (2.1)$$

which gives by Fourier transform the dispersion relation:

$$k^2 = \frac{\omega^2}{c^2} (1 + \hat{\chi}). \quad (2.2)$$

$\hat{\chi}$ can be split into its real and imaginary part:

$$\hat{\chi}(\omega) = \hat{\chi}'(\omega) + i\hat{\chi}''(\omega). \quad (2.3)$$

The real part $\hat{\chi}'$ is linked with the dispersion due to the medium while the imaginary part $\hat{\chi}''$ is linked with the absorption by the medium. This will appear more clearly further on.

This decomposition allows to formulate a weak absorption hypothesis:

$$\begin{cases} \hat{\chi}'(\omega) \ll 1 \\ \hat{\chi}''(\omega) \ll 1 \end{cases} \quad (2.4)$$

This hypothesis is justified numerically in the atmosphere in the UV/visible wavelengths: $|\hat{\chi}| = |n^2 - 1| \sim 10^{-3}$ [208].

This hypothesis allows to simplify the dispersion relation at order 1 in $\hat{\chi}$:

$$k(\omega) = \pm \frac{\omega}{c} \left(1 + \frac{1}{2} \hat{\chi} \right). \quad (2.5)$$

The positive (resp. negative) solutions correspond to waves propagating towards the $+z$ (resp. $-z$) axis. From now, only the ones propagating towards $+z$ will be considered, without loss of generality, because the detected waves are the ones going towards the detector.

A temporal Fourier transform on the propagation equation 2.1 turns it into a second order, constant coefficient differential equation on the spectrum of the field $\hat{E}(\omega, z)$, z being the propagation direction:

$$\partial_z^2 \hat{E}(\omega, z) + \frac{\omega^2 n^2}{c^2} \hat{E}(\omega, z) = 0 \quad (2.6)$$

Here, the electrical field is assumed to be a plane wave. While the real field is not a plane wave, it can be decomposed into a sum of plane waves, hence there is little loss of generality in this assumption. The wavefront mangament of the real beams is dealt with experimentally, as described in section 4.1.1. The spectrum of the field after a propagation of length L in the medium reads [209]:

$$\hat{E}(\omega, L) = \hat{E}(\omega, 0) e^{ik(\omega)L} = \hat{E}(\omega, 0) e^{i\frac{\omega}{c}(1+\hat{\chi}'(\omega))L} e^{-\frac{\omega}{c}\hat{\chi}''(\omega)L}. \quad (2.7)$$

On this expression, it appears clearly that $\hat{\chi}'$ is linked with dispersion, giving the spectral phase of each spectral component, while $\hat{\chi}''$ is linked with absorption, giving the decrease of the field.

To linearise the latter equation, an optically thin sample hypothesis ($\chi k L \ll 1$) is used. This hypothesis is justified a posteriori as long as the intensity decrease due to absorption by the medium is small compared with the absolute intensity. In our case, this hypothesis does not hold very good experimentally. However, it will be kept for the following, because it greatly simplifies the following derivations and allows relevant, qualitative description of the experiment.

The linearised equation reads:

$$\hat{E}(\omega, L) = \hat{E}(\omega, 0) e^{i\frac{\omega}{c}L} \left(1 + i\frac{\omega L}{2c} \hat{\chi}(\omega) \right). \quad (2.8)$$

The absorption coefficient $\hat{\alpha}(\omega) = SN\hat{g}(\omega)$ and the $\hat{\chi}_p(\omega)$ are introduced as follows (to be consistent with the notations of the HITRAN database [210]), for one absorption line:

$$\frac{\omega}{c} \hat{\chi}(\omega) = \hat{\chi}_p(\omega) + iSN\hat{g}(\omega) \quad (2.9)$$

with S the strength of the absorption line (in Hz/mol·cm²), N the molecule density in the absorbing medium (in molecules/cm³) and $\hat{g}$ the normalised absorption line spectral density function (in Hz⁻¹).

Equation 2.8 can then be rewritten with these new notations:

$$\hat{E}(\omega, L) = \hat{E}(\omega, 0) e^{i\frac{\omega}{c}L} \left[1 + \frac{i}{2} L \hat{\chi}_p(\omega) - \frac{1}{2} SNL \hat{g}(\omega) \right]. \quad (2.10)$$

This equation links the spectrum of the incoming field with the spectrum of the outgoing field after the medium. To modelise the experimental temporal trace, the corresponding temporal field must be derived.

2.1.2 Temporal field

The temporal field of the outgoing field after the medium is given by the inverse Fourier transform of the previous equation (with $*$ denoting the convolution product):

$$E(t, L) = E\left(t - \frac{L}{c}, 0\right) + \underbrace{\frac{L}{2} \left[E\left(t - \frac{L}{c}, 0\right) * (i\chi_p(t) - SNg(t)) \right]}_{e(t)} \quad (2.11)$$

with the response of the medium, namely the free induction decay, contained in the second term $e(t)$. This latter term will be further simplified to improve the clarity of the model.

Medium response as a function of g

The term χ_p can be expressed as a function of the lineshape g to simplify the expression, thanks to causal properties of the response of the medium.

The function $\frac{\omega}{c}\hat{\chi}(\omega)$ is regular (analytic on the superior complex half-plan). Physically, it is so that the response of the material happens only after the excitation [209,211]. Formally, it obeys the Kramers-Kronig equation, which gives here [212]:

$$\hat{\chi}_p(\omega) = \frac{1}{\pi} \text{p.v.} \int_{-\infty}^{+\infty} \frac{SN\hat{g}(\omega')}{\omega' - \omega} d\omega'. \quad (2.12)$$

with p.v. short for principal value.

Thus, $\chi(t)$ can be rewritten using only the absorption lineshape function g :

$$i\chi(t) = i\chi_p(t) - SNg(t) = \frac{SN}{2\pi} \left[i \frac{\text{p.v.}}{\pi} \int_{-\infty}^{+\infty} d\omega \int_{-\infty}^{+\infty} d\omega' \frac{\hat{g}(\omega')}{\omega' - \omega} e^{-i\omega t} - \int_{-\infty}^{+\infty} \hat{g}(\omega) e^{-i\omega t} \right] \quad (2.13)$$

The previous equation is injected into the temporal expression of the field (equation 2.11):

$$e(t) = \frac{SN}{2\pi} \left[\overbrace{i \frac{\text{p.v.}}{\pi} \int_{-\infty}^{+\infty} dt' E\left(t' - \frac{L}{c}, 0\right) \int_{-\infty}^{+\infty} d\omega \int_{-\infty}^{+\infty} d\omega' \frac{\hat{g}(\omega')}{\omega' - \omega} e^{-i\omega(t-t')}}^{\text{dispersion}} - \underbrace{\int_{-\infty}^{+\infty} dt' E\left(t' - \frac{L}{c}, 0\right) \int_{-\infty}^{+\infty} d\omega \hat{g}(\omega) e^{-i\omega(t-t')}}_{\text{absorption}} \right]. \quad (2.14)$$

The dispersion term is then developed by reorganising the integral signs:

$$\begin{aligned} & \frac{\text{p.v.}}{\pi} \int_{-\infty}^{+\infty} dt' E\left(t' - \frac{L}{c}, 0\right) \int_{-\infty}^{+\infty} d\omega \int_{-\infty}^{+\infty} d\omega' \frac{\hat{g}(\omega')}{\omega' - \omega} e^{-i\omega(t-t')} \\ &= \frac{\text{p.v.}}{\pi} \int_{-\infty}^{+\infty} dt' E\left(t' - \frac{L}{c}, 0\right) \int_{-\infty}^{+\infty} d\omega' \hat{g}(\omega') e^{-i\omega'(t-t')} \int_{-\infty}^{+\infty} d\omega \frac{e^{i(\omega' - \omega)(t-t')}}{\omega' - \omega}. \end{aligned} \quad (2.15)$$

The following property is then used on the latter integral:

$$\text{p.v.} \int_{-\infty}^{+\infty} \frac{e^{i\omega(t-t')}}{\omega} d\omega = i\pi \text{sgn}(t - t') \quad (2.16)$$

with sgn short for sign, which gives, once reinjected into equation 2.14 and by making $g(t)$ appear:

$$e(t) = -SN \int_{-\infty}^{+\infty} dt' E\left(t' - \frac{L}{c}, 0\right) (1 + \text{sgn}(t - t')) g(t - t') \quad (2.17)$$

which finally gives:

$$e(t) = -2SN \int_{-\infty}^t dt' E\left(t' - \frac{L}{c}, 0\right) g(t - t') = -2SN \int_{-\infty}^{+\infty} dt' E\left(t' - \frac{L}{c}, 0\right) \theta(t - t') g(t - t') \quad (2.18)$$

with $\theta(t)$ the Heaviside step function.

This latter expression is a convolution product. It can be interpreted as a linear response from the medium, whose transfer function is θg . The presence of the Heaviside function shows explicitly that the response of the medium happens after the excitation.

Up to now, no hypothesis has been made on the incoming field. To further simplify the equations and improve the clarity of the model, the shortness of the incoming field (fs pulses) will now be taken into account.

Instantaneous excitation approximation

The next step is to take into account the fact that the pulses are much shorter than the decay time of the excited medium. Numerically, the pulses are approximately 100 fs long while the free induction decay of the studied molecules is approximately of the scale of the hundred of picoseconds [180].

Before using this property, a few more notations are introduced, using functions centered on 0. These will help to formulate the next hypothesis more clearly. The Fourier transform of the envelope of the incoming field and of the free induction decay are introduced:

$$\widehat{E}(\omega, 0) = \widehat{\mathcal{E}}(\omega - \omega_c) \quad (2.19)$$

and alike, for $\widehat{g}(\omega)$:

$$\widehat{g}(\omega) = \widehat{\mathcal{A}}(\omega - \omega_0). \quad (2.20)$$

Thus $E(t, 0) = \mathcal{E}(t)e^{-i\omega_c t}$ and $g(t) = \mathcal{A}(t)e^{-i\omega_0 t}$.

The response term $e(t)$ (équation 2.18) can be rewritten using these notations:

$$e(t) = -2SNe^{-i\omega_0 t}e^{i\omega_c \frac{L}{c}} \int_{-\infty}^{+\infty} dt' \mathcal{E}\left(t' - \frac{L}{c}\right) \theta(t - t') \mathcal{A}(t - t') e^{-i(\omega_c - \omega_0)t'}. \quad (2.21)$$

An instantaneous excitation hypothesis is done, by saying that $\theta(t)\mathcal{A}(t)$ varies much more slowly than $\mathcal{E}(t)e^{-i(\omega_c - \omega_0)t'}$. This hypothesis is justified physically by the fact that the considered pulses are approximately 100 femtoseconds long, while the observed free induction decay is typically on the order of nanoseconds. If this hypothesis is not true, the probing of the free-induction decay by the pulses is much less resolved in time. This allows to simplify the latter equation into:

$$e(t) = -2SNe^{-i\omega_0 t}e^{i\omega_c \frac{L}{c}} \theta\left(t - \frac{L}{c}\right) \mathcal{A}\left(t - \frac{L}{c}\right) \int_{-\infty}^{+\infty} dt' \mathcal{E}\left(t' - \frac{L}{c}\right) e^{-i(\omega_c - \omega_0)t'}. \quad (2.22)$$

and further (by reorganising the phase of the exponential):

$$e(t) = -2SNe^{-i\omega_0(t - \frac{L}{c})} \theta\left(t - \frac{L}{c}\right) \mathcal{A}\left(t - \frac{L}{c}\right) \underbrace{\int_{-\infty}^{+\infty} dt' \mathcal{E}(t') e^{-i(\omega_c - \omega_0)t'}}_{\widehat{E}_0(\omega_0)}. \quad (2.23)$$

Thus, by reinjecting the latter equation into the expression of the total field (equation 2.11), the expression of the temporal field is obtained:

$$E(t, L) = E\left(t - \frac{L}{c}, 0\right) - SNLe^{-i\omega_0(t - \frac{L}{c})} \theta\left(t - \frac{L}{c}\right) \mathcal{A}\left(t - \frac{L}{c}\right) \widehat{E}_0(\omega_0). \quad (2.24)$$

The first term corresponds to the transmitted exciting pulse, while the second term carries the medium's response. Both propagate at celerity c , because the only variable is $t - \frac{L}{c}$. Later on, only t will be written (which is equivalent to setting a new origin of time t , or looking at the field at one given place, for instance on the detector's surface).

2.1.3 Temporal intensity of a single train of pulse

Experimentally, what is measured is photodetectors' output current. This current is proportional not to the field E but to its intensity $I(t) = |E(t, L)|^2$.

Instantaneous intensity

To derive the expression of this intensity, the thin sample hypothesis is used again (the second term of equation 2.24 is much smaller than the first). The previous equation gives then:

$$I(t) = |E_0(t)|^2 + \left[-E_0^*(t) SNL e^{-i\omega_0 t} \theta(t) \mathcal{A}(t) \hat{E}_0(\omega_0) + \text{c.c.} \right] \quad (2.25)$$

which can be rewritten using the envelope functions:

$$I(t) = |\mathcal{E}(t)|^2 - SNL \left[\mathcal{E}^*(t) e^{-i(\omega_0 - \omega_c)t} \theta(t) \mathcal{A}(t) \int_{-\infty}^{+\infty} dt' \mathcal{E}(t') e^{-i(\omega_c - \omega_0)t'} + \text{c.c.} \right]. \quad (2.26)$$

This intensity is limited in time by $\mathcal{E}(t)$: this term does not give information on the free induction decay at time scales larger than the excitation pulse's duration.

Measured intensity

Experimentally, the output signal of the photodetector is amplified and filtered by a low-pass filter, whose cut-off frequency is approximately half the repetition rate of the pulse train (its precise value will be discussed later in section 2.2). This spectral filtering corresponds to a temporal averaging on a duration $T_d \simeq 1/f_{\text{rep}}$. The resulting filtered intensity is noted I_{exp} and can be defined by:

$$I_{\text{exp}}(t) = \langle I(t) \rangle_{T_d} = \frac{1}{T_d} \int_{t-T_d/2}^{t+T_d/2} dt' I(t'). \quad (2.27)$$

This latter function modelises the output signal of the photodiode, amplified and filtered. It is defined up to a constant, that depends on the photodiode, the amplification and the filter.

This definition allows to derive an expression for I_{exp} :

$$I_{\text{exp}}(t) = \int_{t-T_d/2}^{t+T_d/2} dt' |E_0(t')|^2 - SNL \left[\int_{t-T_d/2}^{t+T_d/2} dt' \mathcal{E}^*(t') e^{-i(\omega_0 - \omega_c)t'} \theta(t') \mathcal{A}(t') \hat{E}_0(\omega_0) + \text{c.c.} \right]. \quad (2.28)$$

In our case, the cut-off frequency of the low-pass filter is 50 MHz, slightly lower than half the repetition rate (59 MHz). Thus T_d is slightly higher than the duration between two pulses. Hence, every $[t - T_d/2; t + T_d/2]$ interval contains one pulse, and the measured signal is constant:

$$\int_{t-T_d/2}^{t+T_d/2} dt' |E_0(t')|^2 = \int_{-\infty}^{+\infty} dt' |E_0(t')|^2. \quad (2.29)$$

To be more rigorous, this reasoning would need a precise modelisation of the filter and the corresponding averaging in time. This more precise modelisation was not done during this work because experimentally, this signal was found to be constant, as expected, thus not calling for a finer modelisation.

This same remark also applies to the other term of equation 2.28:

$$\int_{t-T_d/2}^{t+T_d/2} dt' \mathcal{E}^*(t') e^{-i(\omega_0 - \omega_c)t'} \theta(t') \mathcal{A}(t') \hat{E}_0(\omega_0) = \int_{-\infty}^{+\infty} dt' \mathcal{E}^*(t') e^{-i(\omega_0 - \omega_c)t'} \theta(t') \mathcal{A}(t') \hat{E}_0(\omega_0). \quad (2.30)$$

Furthermore, this term can be simplified thanks to the instantaneous excitation hypothesis that was already done in subsection 2.1.3:

$$\int_{-\infty}^{+\infty} dt' \mathcal{E}^*(t') e^{-i(\omega_0 - \omega_c)t'} \theta(t') \mathcal{A}(t') \widehat{E}_0(\omega_0) = \theta(0) \mathcal{A}(0) \int_{-\infty}^{+\infty} dt' \mathcal{E}^*(t') e^{-i(\omega_0 - \omega_c)t'} \widehat{E}_0(\omega_0) \quad (2.31)$$

$$= \theta(0) \mathcal{A}(0) |\widehat{E}_0(\omega_0)|^2. \quad (2.32)$$

Finally, the complete expression of I_{exp} is independant of t :

$$I_{\text{exp}}(t) = \int_{-\infty}^{+\infty} dt' |E_0(t')|^2 - 2SNL \left| \widehat{E}_0(\omega_0) \right|^2 \theta(0) \Re(\mathcal{A}(0)) \quad (2.33)$$

with $\Re$ denoting the real part.

Up to now, it was shown that with our detection setup, the measurement of one train of pulses gives a constant intensity. This intensity is the baseline on which the interferences between the two trains will appear. It is the spectrum of these interferences that contains the spectroscopic informations of interest.

2.1.4 Interference between two pulses

The aim of this subsection is to modelise the interference between two trains of pulses. First one pair of pulse is considered, namely two different, time-shifted pulses, before considering a broader time scale.

Equation 2.24 allows to write:

$$\begin{cases} E_1(t) = \mathcal{E}_1(t) e^{-i\omega_{c,1}t} - SNL\theta(t)\mathcal{A}(t) e^{-i\omega_0 t} \widehat{E}_{0,1}(\omega_0) \\ E_2(t) = \mathcal{E}_2(t - \Delta t) e^{-i\omega_{c,2}(t - \Delta t)} - SNL\theta(t - \Delta t)\mathcal{A}(t - \Delta t) e^{-i\omega_0(t - \Delta t)} \widehat{E}_{0,2}(\omega_0) \end{cases} \quad (2.34)$$

The total field is given by the sum of both fields (according to the superposition principle), so the instantaneous intensity is given by:

$$I(t) = (E_1(t) + E_2(t))(E_1^*(t) + E_2^*(t)) = I_1(t) + I_2(t) + I_{\text{itf}}(t) \quad (2.35)$$

with $I_1(t)$ and $I_2(t)$ given by equation 2.26, and $I_{\text{itf}}(t)$ the interference term. The latter can be separated into the terms that do not depend on the response of the medium, namely the interferogram, given by the transmitted pulses, and the ones who do, namely the free induction decay:

$$I_{\text{itf}}(t) = I_{\text{IGM}}(t) + I_{\text{FID}}(t). \quad (2.36)$$

These two terms will be treated separately.

Centerburst terms

The centerburst terms are the terms of order 0 in the medium response $\mathcal{A}$, namely:

$$I_{\text{IGM}}(t) = \mathcal{E}_1(t) \mathcal{E}_2^*(t - \Delta t) e^{-i(\omega_{c,1} - \omega_{c,2})t} e^{-i\omega_{c,2}\Delta t} + \text{c.c.} \quad (2.37)$$

Just like above in subsection 2.1.3, what is measured is not the instaneous intensity but the averaged and filtered intensity $I_{\text{exp,IGM}}(t)$:

$$I_{\text{exp,IGM}}(t) = \langle I_{\text{IGM}} \rangle_{T_d}(t) = \frac{1}{T_d} \int_{t-T_d/2}^{t+T_d/2} I_{\text{IGM}}(t') dt'. \quad (2.38)$$

which gives:

$$I_{\text{exp,IGM}}(t) = e^{-i\omega_{c,2}\Delta t} \int_{-\infty}^{+\infty} \mathcal{E}_1(t') \mathcal{E}_2^*(t' - \Delta t) e^{-i(\omega_{c,1} - \omega_{c,2})t'} dt' + \text{c.c.} \quad (2.39)$$

Just like in section 2.1.3, this expression does not depend on t . That is to say, with our detection setup, for one given pair of pulses, the measured intensity is fixed, as long as the pair of the pulses is within the $[t - T_d/2 : t + T_d/2]$ interval (which is always the case). The experimental signal will vary, but on a broader temporal scale, considering more than one pair of pulses: indeed, from one pair of pulses to another, Δt will vary, and the latter expression depends on Δt . This will be treated on the next subsection (2.1.5).

Free induction decay (FID) term

The free induction decay term is given by:

$$I_{\text{FID}}(t) = -SNL \left(\mathcal{E}_1(t) e^{-i\omega_{c,1}t} \theta(t - \Delta t) \mathcal{A}^*(t - \Delta t) e^{i\omega_0(t - \Delta t)} \widehat{E}_{0,2}^*(\omega_0) \right. \\ \left. + \mathcal{E}_2^*(t - \Delta t) e^{i\omega_{c,2}(t - \Delta t)} \theta(t) \mathcal{A}(t) e^{-i\omega_0 t} \widehat{E}_{0,1}(\omega_0) + \text{c.c.} \right). \quad (2.40)$$

Just like above in subsection 2.1.3, what is measured is not the instantaneous intensity but the averaged and filtered intensity $I_{\text{exp,FID}}(t)$:

$$I_{\text{exp,FID}}(\Delta t) = \langle I_{\text{FID}} \rangle_{T_d}(t) = \frac{1}{T_d} \int_{t-T_d/2}^{t+T_d/2} I_{\text{FID}}(t') dt'. \quad (2.41)$$

To derive the next expression, the hypotheses that were done before are used once again (instantaneous excitation and averaging), leading to:

$$I_{\text{exp,FID}}(t) = -SNL \left(\theta(-\Delta t) \mathcal{A}^*(-\Delta t) e^{-i\omega_0 \Delta t} \widehat{E}_{0,1}(\omega_0) \widehat{E}_{0,2}^*(\omega_0) \right. \\ \left. + \theta(\Delta t) \mathcal{A}(\Delta t) e^{-i\omega_0 \Delta t} \widehat{E}_{0,2}^*(\omega_0) \widehat{E}_{0,1}(\omega_0) + \text{c.c.} \right). \quad (2.42)$$

The two terms of this latter equation are symmetrical. The first term corresponds to the probing of the FID of the second pulse by the first pulse: it can only happen if the first pulse happens after the second pulse, thus the $\theta(-\Delta t)$. The opposite can be said about the second term: it corresponds to the probing of the FID of the first pulse by the second pulse.

Experimentally, if only the first (resp. the second) pulse triggers a FID, namely if only the first (resp. the second) comb travels through the absorbing medium, the second (resp. first) of these two terms vanishes, and thus the corresponding signal will only appear before (resp. after) the centerburst of the interferograms. In the following, the case corresponding to our experimental setup will be considered, namely the one where both pulses trigger a FID.

To further simplify, the property $\mathcal{A}^*(-t) = \mathcal{A}(t)$ is injected. This property is due to the fact that $\widehat{\mathcal{A}}(\omega)$ is purely real by construction (see equations 2.9 and 2.20).

$$I_{\text{exp,FID}}(t) = -SNL \left(\mathcal{A}(\Delta t) e^{-i\omega_0 \Delta t} \widehat{E}_{0,1}(\omega_0) \widehat{E}_{0,2}^*(\omega_0) + \text{c.c.} \right). \quad (2.43)$$

Just like the centerburst term, this FID term does not depend on t but depends on Δt , which will be discussed in the next subsection (2.1.5).

Summary

Putting everything together, the expression of the measured intensity for one pair of pulses is obtained:

$$\begin{aligned}
I_{\text{exp}} = & \int_{-\infty}^{+\infty} (|\mathcal{E}_1(t')|^2 + |\mathcal{E}_2(t')|^2) dt' \\
& + \left(e^{-i\omega_{c,2}\Delta t} \int_{-\infty}^{+\infty} \mathcal{E}_1(t') \mathcal{E}_2^*(t' - \Delta t) e^{-i(\omega_{c,1} - \omega_{c,2})t'} dt' + \text{c.c.} \right) \\
& - 2SNL\theta(0)\Re(\mathcal{A}(0)) \left(\left| \hat{E}_{0,1}(\omega_0) \right|^2 + \left| \hat{E}_{0,2}(\omega_0) \right|^2 \right) \\
& - SNL \left(\mathcal{A}(\Delta t) e^{-i\omega_0\Delta t} \hat{E}_{0,1}(\omega_0) \hat{E}_{0,2}^*(\omega_0) + \text{c.c.} \right).
\end{aligned} \tag{2.44}$$

Each term of equation 2.44 corresponds to the detection of one term of the femtosecond-scale intensity, which is decomposed in figure 2.2. The detected terms can be interpreted:

- The first line corresponds to the intensity due to each pulse's "own" intensity, without interaction between the pulses. It is constant with respect with Δt , which means it will only contribute to set the level of the baseline in the interferograms' temporal trace.
- The second line corresponds to the centerburst. It can be interpreted as a cross-correlation function between the two transmitted pulses. Later on, the influence of other parameters (the relative phase, chirp, etc.) will be investigated. This term is at order 0 in the medium response.
- The third line corresponds to the probing of the medium by each pulse thanks to the excitation this same pulse caused. Just like the first term, it is constant with respect with Δt and will participate to set the level of the baseline. This term is at order 1 in the medium response.
- The fourth term corresponds to the probing of the free induction decay caused by one pulse thanks to the other pulse. As seen above, if only one pulse goes through the absorbing medium, this term will be multiplied by a factor $\pm\theta(\Delta)$, the sign being determined by which pulse is absorbed. This term is at order 1 in the medium response.

Up to now, the measurement of the interference of two pulses was modelised, which corresponds experimentally to one acquisition point. To modelise the full interferogram, a larger time scale must be considered.

2.1.5 At the time scale of the lab

At the scale of a single interferogram

The goal of this subsection is to explicit the link between the individual pulses time scale (fs scale) and the "macroscopic" time of our experimental acquisition at the scale of one interferogram, 1 to 100 μs .

The repetition rate difference between the two sources makes the time shift Δt between the two pulses vary. For a given Δf_{rep} , this variation $\delta\Delta t$ is given by:

$$\delta\Delta t = \frac{1}{f_{\text{rep}}} - \frac{1}{f_{\text{rep}} + \Delta f_{\text{rep}}} = \frac{\Delta f_{\text{rep}}}{f_{\text{rep}}(f_{\text{rep}} + \Delta f_{\text{rep}})} \simeq \frac{\Delta f_{\text{rep}}}{f_{\text{rep}}^2} \tag{2.45}$$

This latter simplification is justified by the fact that Δf_{rep} is much smaller than f_{rep} . Numerically, in our case, Δf_{rep} ranges from 1 to 400 Hz, while $f_{\text{rep}} \sim 118 \text{ MHz}$.

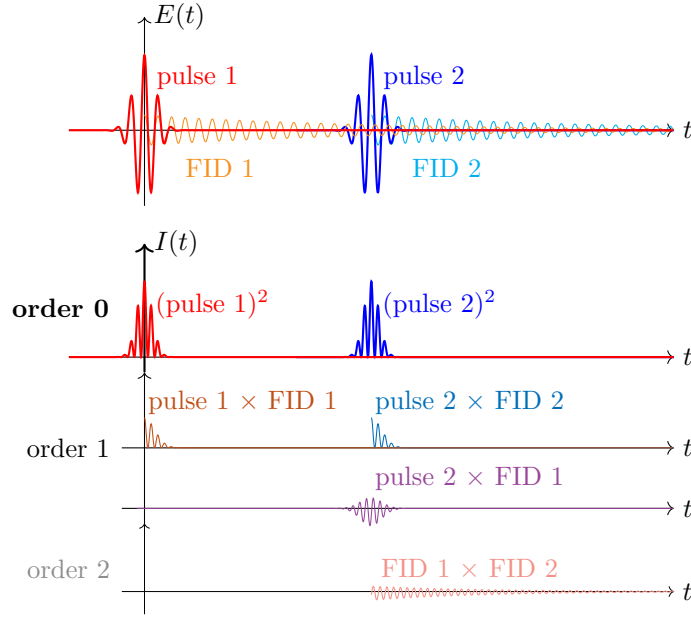


Figure 2.2: Femtosecond-scale field (a) and intensity (b) (decomposed in order 0, 1 and 2 in $\mathcal{A}$) in the case of two shifted pulses exciting a single transition of the medium. The four lines of the intensity, separated for the sake of clarity, correspond to (order 0, first line of equation 2.44) the intensity of the transmitted pulses, (order 1, third and fourth line of equation 2.44) the scanning of each pulses' own FID and scanning of one pulse's FID by the other pulse, (order 2) product of the FID of the two pulses. This latter term is of order 2 in $\mathcal{A}$ and thus neglected in the analytical modelisation, that relies on a development at order 1 (thin medium hypothesis from subsection 2.1.1). The scanning of one pulse by the other pulse, namely the centerburst term, second line of equation 2.44) is omitted in this case because it is null since the pulses are far apart in time. Experimentally, the pulses are ~ 100 fs long while the FID are ~ 1 ns long.

In this subsection, Δf_{rep} is considered as a constant. This approximation will be discussed experimentally in the next chapter (section 3.3) and numerically using a simulation later on section 2.3. Hence, Δt varies linearly in time, from one pair of pulses to another. If the following pairs of pulses are numbered by an index k , the corresponding Δt_k is given by:

$$\Delta t_k = \Delta t_0 + \frac{\Delta f_{\text{rep}}}{f_{\text{rep}}^2} k. \quad (2.46)$$

The instants t_k are then defined as such: the pulse from the first comb whose time shift with the closest pulse from the second comb is Δt_k reaches the photodetector at the instant t_k . (Hence, these instants are defined precisely up to the fs scale, which is much more accurate than any experimental detection device.) This definition is illustrated on figure 2.3.

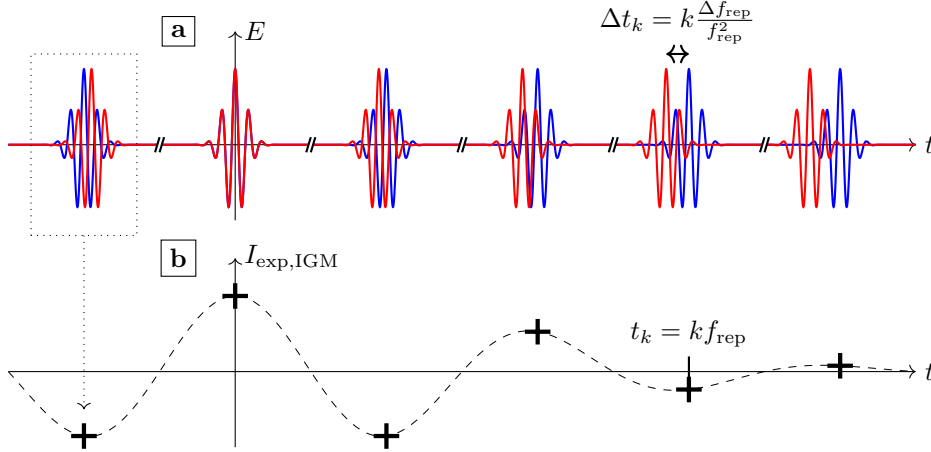


Figure 2.3: Illustration of the link between the femtosecond scale time and the IGM scale time. (a) Temporal field of the pulses of the two combs, one ~ 100 fs-long pulse once every ~ 10 ns. The interference between these two pulses give, at a much bigger time scale, a centerburst term (b). Each point of the interferogram corresponds to one single pair of pulses.

Formally, the link between $t_k = k/f_{\text{rep}}$ and Δt_k is:

$$\Delta t_k = k \frac{\Delta f_{\text{rep}}}{f_{\text{rep}}^2} = t_k f_{\text{rep}} \frac{\Delta f_{\text{rep}}}{f_{\text{rep}}^2} = t_k \frac{\Delta f_{\text{rep}}}{f_{\text{rep}}}. \quad (2.47)$$

This equation can be interpreted as the link between the "macroscopic" time (the scale of the laboratory) and the femtosecond scale. To each pulse corresponds an instant t_k , thus a Δt_k , thus an intensity $I_{\Delta t_k}$. The factor between the is noted $a = f_{\text{rep}}/\Delta f_{\text{rep}}$ and is on the order of 10^6 to 10^8 experimentally in our case.

In the case where the acquisition is synchronised with the pulses, each experimental point of the temporal trace corresponds to a pair of pulses. Hence, equation 2.47 gives an expression of Δt_k as a function of t_k , and the modelised experimental signal $S(t_k)$ can be written using equation 2.44:

$$S(t_k) = I_{\text{exp}, \Delta t_k = t_k/a} \text{ with } a = \frac{f_{\text{rep}}}{\Delta f_{\text{rep}}}. \quad (2.48)$$

However, in our case, the acquisition is not synchronised with the pulses. Nevertheless, experimentally, we observed that a variation of the acquisition rate (125 MHz, 100 MHz) does not affect the retrieved DCS spectrum, as it is filtered by a 50 MHz bandpass low-pass filter before acquisition. Thus, in the following of this modelisation, the synchronicity between the acquisition rate and the repetition rate will be assumed ($f_{\text{acq}} = f_{\text{rep}}$), allowing to write the modelised experimental signal $S(t)$ as:

$$S(t) = I_{\text{exp}, \Delta t=t/a} \text{ with } a = \frac{f_{\text{rep}}}{\Delta f_{\text{rep}}}. \quad (2.49)$$

Hence, the experimental signal is the femtosecond-scale intensity, stretched in time by a factor a . Numerically, in our case, this factor ranges from 10^6 to 10^8 as Δf_{rep} ranges from 1 to 100 Hz.

To further modelise the asynchronicity between the pulses and the acquisition, a precise modelisation of the detection setup (photodiode, amplification, filtering) would be necessary, which was not performed during this work. Besides, the effects of Δf_{rep} fluctuations will be studied in section 2.3.

To modelise our whole acquisitions, that contain several interferograms, an even longer time scale must be considered.

At the scale of several interferograms

The time shift between two consecutive interferograms is strongly linked with the repetition rate difference Δf_{rep} , as depicted in figure 2.4.

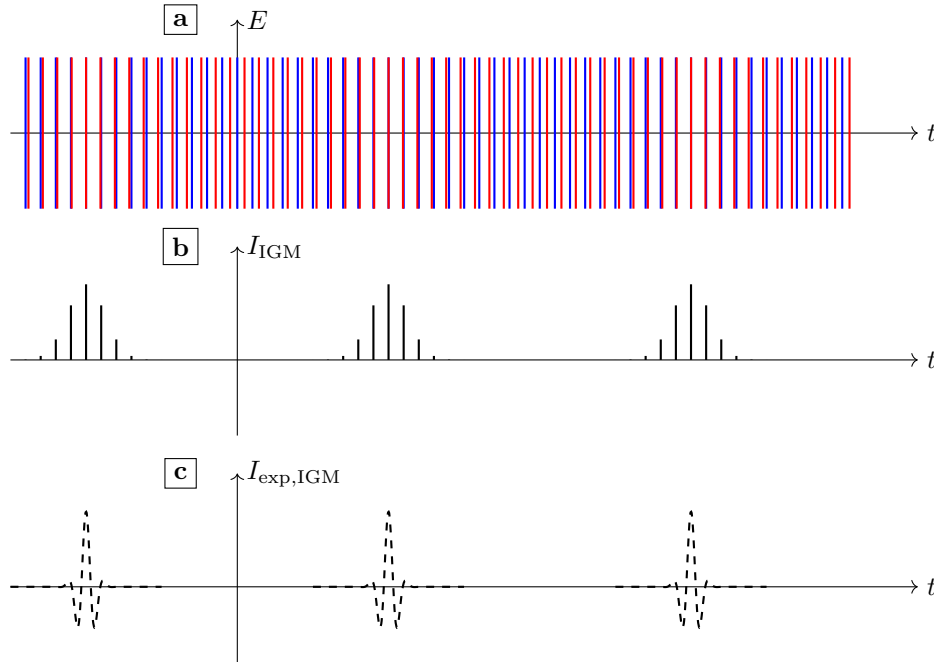


Figure 2.4: Depiction of the link between Δf_{rep} and the time shift between two consecutive interferograms. (a) The two trains of pulses (in blue and red) interfere only when two pulses are very close, giving an intensity (b) that is detected and averaged to give an interferogram (c). After one interferogram, the delay between the pulses from the two sources is almost zero and increases with t ; another interferogram appears when this delay reaches $1/f_{\text{rep}}$, namely after a $1/\Delta f_{\text{rep}}$ time according to equation 2.47. The graphs (a) and (c) correspond to a zoom out on the graphs from figure 2.3.

Formally, the delay between two consecutive interferograms is the time for Δt to reach $1/f_{\text{rep}}$, namely $1/\Delta f_{\text{rep}}$.

Hence, the complete experimental signal with every interferogram can be modelised by the repetition of the signal of one interferogram every $1/\Delta f_{\text{rep}}$. This modelisation implies that Δf_{rep} is a

constant and that all interferograms are exactly similar, which will be discussed in subsection 2.3 and 2.1.7 respectively.

$$S_{\text{tot}}(t) = \sum_{n=-\infty}^{+\infty} S(t - n/\Delta f_{\text{rep}}) \quad (2.50)$$

Having obtained a modelisation of the temporal trace, the modelisation of the corresponding experimental spectrum, which is obtained by Fourier transform of the temporal trace, can now be derived.

2.1.6 DCS spectrum

At the scale of one interferogram

Our experimental spectrum is obtained through FFT of the interferogram trace. Thus, to modelise this spectrum, the Fourier transform of our modelisation of the interferogram trace $S(t)$ is derived. Like in subsection 2.1.4, the centerburst term and the FID term will be treated separately.

The temporal interferogram term can be rewritten by making the Fourier transforms of both pulses appear:

$$\begin{aligned} S_{\text{IGM}}(t) &= I_{\text{exp}, \Delta t=t/a} = e^{-i\omega_{c,2}t/a} \int_{-\infty}^{+\infty} \mathcal{E}_{0,1}(t') \mathcal{E}_{0,2}^*(t' - t/a) e^{-i(\omega_{c,1}-\omega_{c,2})t'} dt' + \text{c.c.} \\ &= \frac{1}{(2\pi)^2} \int_{-\infty}^{+\infty} dt' \int_{-\infty}^{+\infty} d\omega \hat{E}_{0,1}(\omega) e^{+i\omega t'} \int_{-\infty}^{+\infty} d\omega' \hat{E}_{0,2}^*(\omega') e^{-i\omega'(t'+t/a)} + \text{c.c.} \\ &= \frac{1}{(2\pi)^2} \int_{-\infty}^{+\infty} d\omega \int_{-\infty}^{+\infty} d\omega' \hat{E}_{0,1}(\omega) \hat{E}_{0,2}^*(\omega') e^{-i\omega' t/a} \underbrace{\int_{-\infty}^{+\infty} dt' e^{-i(\omega-\omega')t'}}_{2\pi\delta(\omega-\omega')} \\ &= \frac{1}{2\pi} \int_{-\infty}^{+\infty} d\omega \hat{E}_{0,1}(\omega) \hat{E}_{0,2}^*(\omega) e^{-i\omega t/a} + \text{c.c.} \end{aligned}$$

Its Fourier transform can then be computed:

$$\begin{aligned} \hat{S}_{\text{IGM}}(\omega) &= \frac{1}{(2\pi)^2} \int_{-\infty}^{+\infty} dt \int_{-\infty}^{+\infty} d\omega' \hat{E}_{0,1}(\omega') \hat{E}_{0,2}^*(\omega') e^{-i\omega' t/a} e^{i\omega t} + \text{c.c.} \\ &= \frac{1}{(2\pi)^2} \int_{-\infty}^{+\infty} d\omega' \hat{E}_{0,1}(\omega') \hat{E}_{0,2}^*(\omega') \underbrace{\int_{-\infty}^{+\infty} dt e^{i(\omega-\omega'/a)t}}_{2\pi\delta(\omega-\omega'/a)} + \text{c.c.} \\ &= \frac{1}{2\pi} \hat{E}_{0,1}(\omega/a) \hat{E}_{0,2}^*(\omega/a) + \text{c.c.} \end{aligned} \quad (2.52)$$

This spectrum is the product of the spectra of the two pulses, stretched by a factor $1/a$. This stretching will be discussed further on this subsection.

The Fourier transform of the free induction decay term gives:

$$\hat{S}_{\text{FID}}(\omega) = -SNL\hat{A}((\omega - \omega_0)/a) \left(\hat{E}_{0,1}(\omega_0) \hat{E}_{0,2}^*(\omega_0) + \text{c.c.} \right). \quad (2.53)$$

Like the spectrum of the interferogram, the FID spectrum is stretched by a factor $1/a$ compared with the optical spectrum.

All of the terms are finally put together and the instantaneous excitation hypothesis is used (which is equivalent to say that the spectral width of $\hat{\mathcal{A}}$ is much smaller than the one of $\hat{E}_0$) once again:

$$\begin{aligned}\hat{S}(\omega) &= \frac{1}{2\pi} \left(\hat{E}_{0,1}(\omega/a) \hat{E}_{0,2}^*(\omega/a) - 2\pi SNL \hat{\mathcal{A}}((\omega - \omega_0)/a) \hat{E}_{0,1}(\omega_0) \hat{E}_{0,2}^*(\omega_0) + \text{c.c.} \right) \\ &= \frac{1}{2\pi} \underbrace{\left(1 - 2\pi SNL \hat{\mathcal{A}}((\omega - \omega_0)/a) \right)}_{\text{transmittance}} \underbrace{\left(\hat{E}_{0,1}(\omega/a) \hat{E}_{0,2}^*(\omega/a) + \text{c.c.} \right)}_{\text{envelope}}\end{aligned}\quad (2.54)$$

This expression can be interpreted as the product of an envelope term (the second term, corresponding to the transmitted pulses) and a transmittance function. It shows that in this model, the spectral phase of the envelope, contained in the envelope terms, does not affect the lineshape of the transmittance term. Namely, chirp or chirp difference between the two combs does not deform the absorption lines.

The stretch of a factor $1/a$ shifts the spectrum from optical frequencies ($\sim 10^2$ THz) to radiofrequencies ($\sim 10^1$ MHz). Thus these DCS spectra are often referred to as radiofrequency (rf) spectra. The mapping of optical frequencies to radiofrequencies will be considered in section 2.2.

At the scale of several interferograms

The Fourier transform of the total temporal trace S_{tot} (given by equation 2.50) gives, by property of the Fourier transform [213]:

$$\hat{S}_{\text{tot}}(\omega) = \sum_{n=-\infty}^{+\infty} \text{TF}(S(t - n/\Delta f_{\text{rep}}))(\omega) \quad (2.55)$$

$$= \frac{1}{2\pi\Delta f_{\text{rep}}} \hat{S}(\omega) \sum_{n=-\infty}^{+\infty} \delta(\omega - 2\pi n\Delta f_{\text{rep}}) \quad (2.56)$$

This spectrum is the spectrum of one single interferogram, discretised into a comb structure. The comb structure of this spectrum comes from the periodicity of the temporal signal.

Up to now, the hypothesis that all pulses from one direction are identical has been made. But experimentally, the parameters of the pulses are not controlled in our freerunning cavity. In particular, the presence of an envelope-carrier phase can be expected [145]. The objective of the next subsection is to modelise its effect.

2.1.7 Phase drift

The goal of this subsection is to modelise the effect of the carrier-envelope phase drift from one pulse to another, on both the temporal trace and the spectrum of the experimental signal.

Temporal trace

Formally, the envelope-carrier phase φ_o is modelised by an phase on the envelope term:

$$E(t) = \mathcal{E}(t)e^{-i\omega_c t} = |\mathcal{E}(t)|e^{-i\omega_c t}e^{-i\varphi_o}. \quad (2.57)$$

To modelise the variation of this phase from one pulse to another, the pulses are numbered with the index k , and a linear variation of φ_o is considered:

$$E_k(t) = \mathcal{E}(t)e^{-ik\varphi_o}e^{-i\omega_c t}. \quad (2.58)$$

This term is injected into the expression of the temporal field 2.8. The derivation of I_{exp} gives, with $\Delta\varphi_o = \varphi_{o,2} - \varphi_{o,1}$:

$$\begin{aligned} I_{\text{exp,IGM}}(t) &= \frac{e^{if_{\text{rep}}t\Delta\varphi_o}}{2\pi} \int_{-\infty}^{+\infty} d\omega \hat{E}_{0,1}(\omega) \hat{E}_{0,2}^*(\omega) e^{-i\omega t/a} + \text{c.c.} \\ &= \frac{e^{i\Delta\omega_0 t}}{2\pi} \int_{-\infty}^{+\infty} d\omega \hat{E}_{0,1}(\omega) \hat{E}_{0,2}^*(\omega) e^{-i\omega t/a} + \text{c.c.} \text{ with } \Delta\omega_0 = f_{\text{rep}}\Delta\varphi_o \end{aligned} \quad (2.59)$$

It appears that only the difference of envelope/carrier phase between the two combs $\Delta\varphi_o$ has an effect on the temporal trace of the interferogram (and not the absolute value of $\varphi_{c/e,1}$ or 2). This effect is an additionnal oscillation, at pulsation $\Delta\omega_0$.

The FID term undergoes the same change:

$$I_{\text{exp,FID}}(t) = -SNL \left(\mathcal{A}(t/a) e^{i\Delta\omega_0 t} e^{-i\omega_0 t/a} \hat{E}_{0,1}(\omega_0) \hat{E}_{0,2}^*(\omega_0) + \text{c.c.} \right). \quad (2.60)$$

Hence, the total interference term is given by:

$$I_{\text{exp,itf}}(t) = I_{\text{exp,IGM}}(t) + I_{\text{exp,FID}}(t) \quad (2.61)$$

$$\begin{aligned} &= \frac{e^{i\Delta\omega_0 t}}{2\pi} \left[\int_{-\infty}^{+\infty} d\omega \hat{E}_{0,1}(\omega) \hat{E}_{0,2}^*(\omega) e^{-i\omega t/a} \right. \\ &\quad \left. - 2\pi SNL \left(\mathcal{A}(t/a) e^{-i\omega_0 t/a} \hat{E}_{0,1}(\omega_0) \hat{E}_{0,2}^*(\omega_0) \right) \right] + \text{c.c.} \end{aligned} \quad (2.62)$$

$$= e^{i\Delta\omega_0 t} I_{\text{exp,itf, without c/e phase}}(t) \quad (2.63)$$

At a larger time scale (more than one interferogram), like in subsection 2.1.5:

$$S_{\text{tot}}(t) = e^{i\Delta\omega_0 t} \sum_{n=-\infty}^{+\infty} S_{\text{without c/e phase}}(t - n/\Delta f_{\text{rep}}) \quad (2.64)$$

The presence of the relative carrier-envelope phase makes the interferograms different. Indeed, on the contrary of the integral, this new oscillating term is not necessarily periodic with period $1/\Delta f_{\text{rep}}$. Thus consecutive interferograms present a relative phase. Experimentally, this prevents a straightforward averaging of the interferograms.

Effect of the phase drift on the DCS spectrum

The Fourier transform of the previous spectrum gives, using Fourier transform properties [213]:

$$\hat{S}(\omega) = \hat{S}_{\text{without c/e phase}}(\omega + \Delta\omega_0) \quad (2.65)$$

Hence, the effect of the envelope/carrier phase on the spectrum of one interferogram is a global shift of the modelised radiofrequency spectrum.

At a larger time scale, the spectrum of a temporal trace with all the interferograms is given by:

$$\hat{S}_{\text{tot}}(\omega) = \frac{1}{2\pi\Delta f_{\text{rep}}} \hat{S}(\omega + \Delta\omega_0) \sum_{n=-\infty}^{+\infty} \delta(\omega + \Delta\omega_0 - 2\pi n\Delta f_{\text{rep}}) \quad (2.66)$$

Hence, compared with the case without envelope/carrier phase, the effect of the envelope/carrier phase is to shift the comb structure on the radiofrequency spectrum.

To validate this modelisation and visualise the influence of other parameters than the envelope/carrier phase, especially Δf_{rep} fluctuations, a simulation was developed and will be discussed in section 2.3. Beforehand, the retrieval of optical spectra from radiofrequency spectra will be treated.

2.2 Retrieval of optical spectrum from radiofrequencies spectrum

The result obtained in subsection 2.1.6 is that the experimental radiofrequency DCS spectrum is a frequency comb that corresponds to the optical frequency comb stretched and shifted. The goal of this section is to develop the formalism to link the radiofrequency comb and the optical comb. This link is needed to retrieve the optical spectrum from the radiofrequency spectrum.

2.2.1 Notations and general case

As shown in subsection 2.1.6, the frequencies from the two combs can be noted:

$$\begin{cases} f_1 = f_{0,1} + n f_{\text{rep},1} & n \in \mathbb{N} \\ f_2 = f_{0,2} + m f_{\text{rep},2} & m \in \mathbb{N} \end{cases} \quad (2.67)$$

which can be rewritten as:

$$\begin{cases} f_1 = f_0 + n f_{\text{rep}} & n \in \mathbb{N} \\ f_2 = f_0 + \Delta f_0 + m f_{\text{rep}} + m \Delta f_{\text{rep}} & m \in \mathbb{N} \end{cases} \quad \text{with} \quad \begin{cases} f_0 = f_{0,1} \\ \Delta f_0 = f_{0,2} - f_{0,1} \\ f_{\text{rep}} = f_{\text{rep},1} \\ \Delta f_{\text{rep}} = f_{\text{rep},2} - f_{\text{rep},1} \end{cases} \quad (2.68)$$

$\Delta f_{\text{rep}} > 0$ is assumed without loss of generality.

The interference between two comb teeth gives the sum and difference of their respective frequencies. Thus, the interference between the two combs give the frequencies:

$$\begin{cases} f_{n,m} = f_0 \pm f_0 + (n \pm m) f_{\text{rep}} \pm \Delta f_0 \pm m \Delta f_{\text{rep}} & m, n \in \mathbb{N} \\ f'_{n,m} = -f_{n,m} \end{cases} \quad (2.69)$$

However, not all of these frequencies are within the bandwidth of our detection setup. In particular, the presence of the low-pass filter restricts the latter list to the frequencies that verify $|f_{n,m}| < f_{\text{rep}}/2$. This filtering allows to isolate one single beating envelope and to avoid aliasing, as will be seen on next subsection.

2.2.2 Isolating a single beating envelope

Eliminating the sum of frequencies

In equation 2.69, f_0 is smaller than f_{rep} by construction, Δf_0 is smaller than $2f_{\text{rep}}$, and Δf_{rep} is much smaller than f_{rep} . Thus, the $f_{n,m}$ that lie within the bandwidth of the filter correspond to those where the $(n \pm m)f_{\text{rep}}$ term remains under $3f_{\text{rep}}$. This can only be the case in the negative case ($\pm \rightarrow -$), that is to say that the beating of the two combs, and not their frequency sum, is considered.

Thus, $k = n - m \in \{-3, \dots, 3\}$ can be fixed, and equation 2.69 can be rewritten:

$$\begin{cases} f_{n,m} = \Delta f_0 + m \Delta f_{\text{rep}} - k f_{\text{rep}} & m \in \mathbb{N}, k \in \{-3, \dots, 3\} \\ f'_{n,m} = -f_{n,m} \end{cases} \quad (2.70)$$

Radiofrequency spectral bandwidth and aliasing

Equation 2.69 allows to retrieve the bandwidth of the spectrum in the radiofrequency region Δf_{rf} . Indeed, among the listed frequencies, the only ones whose power spectrum has a non-zero amplitude are the ones corresponding to teeth of the original combs with non-zero amplitude. The number Δn of corresponding indices n is given roughly by:

$$\Delta n = \Delta f_{\text{opt}}/f_{\text{rep}} \sim 10^4 \text{ in our case} \quad (2.71)$$

and thus, combining the latter equation with equation 2.69:

$$\Delta f_{\text{rf}} = \Delta n \Delta f_{\text{rep}} \sim 10^1 \text{ to } 10^3 \text{ kHz in our case} \quad (2.72)$$

This quantity is very small compared with f_{rep} and the bandwidth of our filter ($f_{\text{rep}}/2 \sim 59 \text{ MHz}$). This ensures that for a given pair of spectra, the teeth within the bandwidth of the filter correspond to a single value of $k = n \pm m$, as explained in figure 2.5.

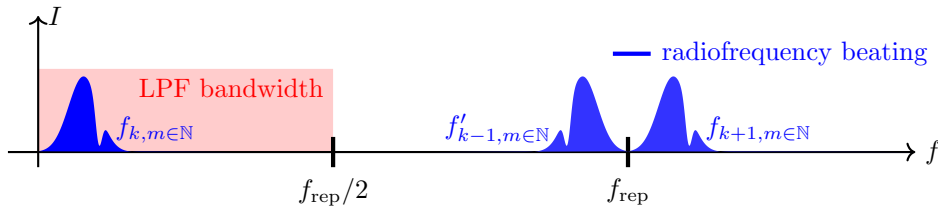


Figure 2.5: Illustration of the retrieved DCS radiofrequencies spectra: when no aliasing occurs, only one envelope is included in the low-pass filter bandwidth.

Moreover, if the spectrum is aliased (that is to say, if two teeth with non-zero amplitude are found around $f = 0$ or $f = f_{\text{rep}}/2$) (see figure 2.6), it will be aliased only once.

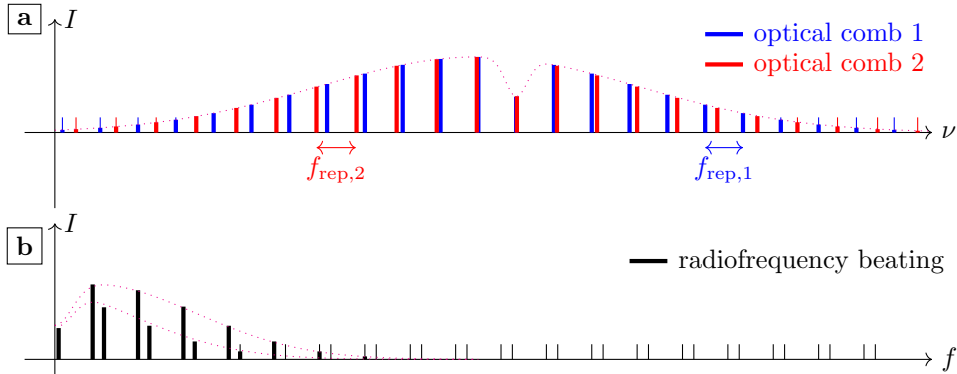


Figure 2.6: Illustration of the retrieved DCS spectrum in case of strong aliasing

In this latter case, the retrieval of the optical frequencies is not possible (unless a very high spectral resolution). Δf_{rep} and Δf_0 values must be set to avoid it. Since this case did not occur during this work, it will be neglected in the following.

2.2.3 Direct or reverse mapping of optical frequencies on radiofrequencies

Equation 2.70 shows that depending on the value of Δf_0 (that can range from $-f_{\text{rep}}$ to f_{rep}), either $f_{k,m}$ or $f'_{k,m}$ can be within the bandwidth of the low-pass filter, so the mapping of optical frequencies on radiofrequencies can be "direct" (higher optical frequencies being mapped on higher

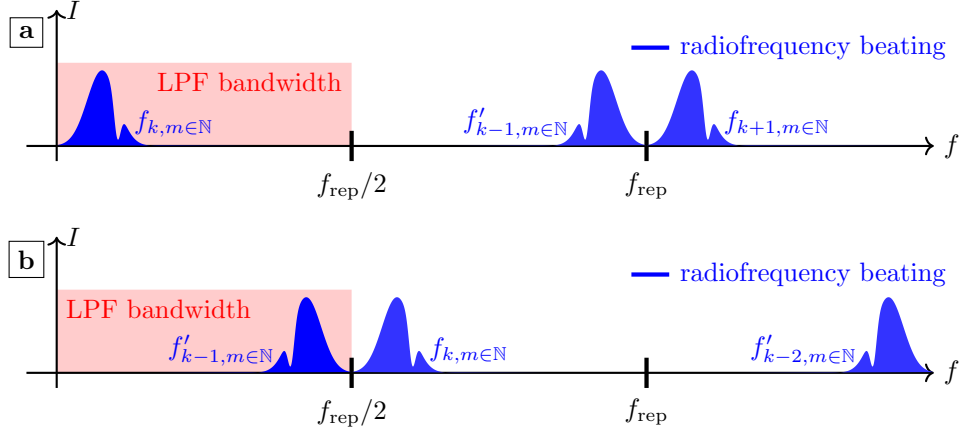


Figure 2.7: Illustration of two cases of retrieved DCS radiofrequencies: (a) "direct" case, where only frequencies $f_{k,m}$ are in the bandwidth of the filter; (b) "reverse" case, where only frequencies $f'_{k,m}$ are in the bandwidth of the filter

radiofrequencies) or "reversed" (higher optical frequencies being mapped on lower radiofrequencies). These two cases are illustrated in figure 2.7.

These two cases correspond to two different formulas to retrieve optical frequencies from radiofrequencies.

To establish the relation between optical and radio frequencies, one can isolate an expression of n in the expression of radiofrequencies (equation 2.70) and inject it in the expression of optical frequencies (equation 2.67).

Direct case

The direct case is illustrated in figure 2.8.

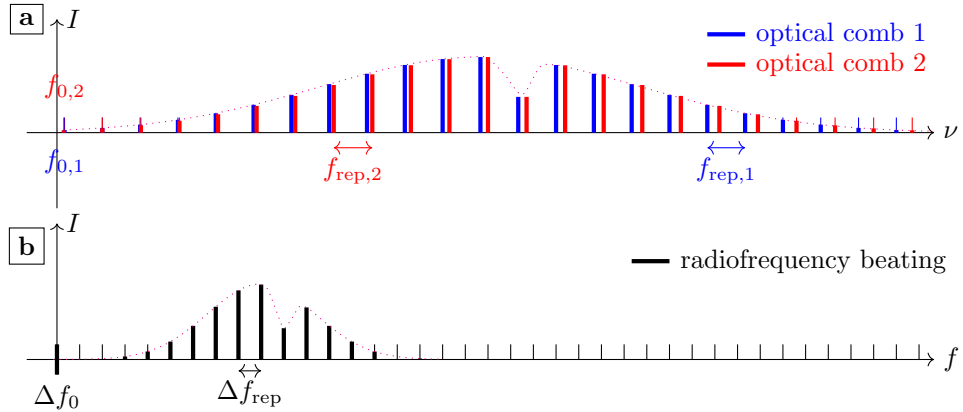


Figure 2.8: Illustration of the "direct" retrieval case : higher optical frequencies correspond to higher radiofrequencies

In this case, the optical frequencies f_{opt} are linked with radiofrequencies $f_{k,m}$ by the relation:

$$f_{\text{opt}} = f_{0,2} + a f_{\text{rf}} + a(k f_{\text{rep}} - \Delta f_0) \quad (2.73)$$

$$\text{with } a = \frac{f_{\text{rep}} + \Delta f_{\text{rep}}}{\Delta f_{\text{rep}}} \simeq \frac{f_{\text{rep}}}{\Delta f_{\text{rep}}}.$$

Reverse case

The reverse case is illustrated in figure 2.9.

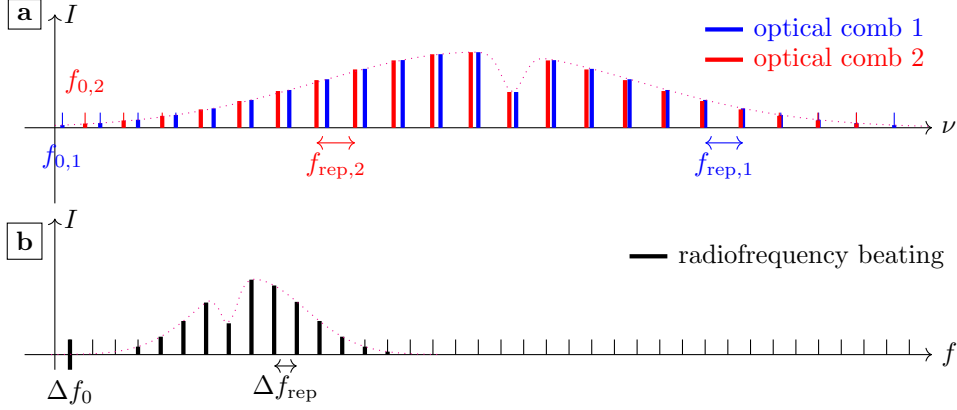


Figure 2.9: Illustration of the "direct" retrieval case : higher optical frequencies correspond to lower radiofrequencies

In this case, the optical frequencies f_{opt} are linked with radiofrequencies $f'_{k,m}$ by the relation:

$$f_{\text{opt}} = f_{0,2} - a(\Delta f_0 - k f_{\text{rep}}) - a f_{\text{rf}} \quad (2.74)$$

$$\text{with } a = \frac{f_{\text{rep}} + \Delta f_{\text{rep}}}{\Delta f_{\text{rep}}} \simeq \frac{f_{\text{rep}}}{\Delta f_{\text{rep}}}.$$

Summary

In the general case, the link between optical and radio frequencies can be written as:

$$f_{\text{opt}} = f_{\text{offset}} \pm m a \Delta f_{\text{rep}} \quad (2.75)$$

with the sign of $\pm$ depending on whether the case is direct or reverse, and f_{offset} containing Δf_0 and $k f_{\text{rep}}$. The f_{offset} is a global offset of the retrieved spectrum, while $a = f_{\text{rep}}/\Delta f_{\text{rep}}$ is the stretch factor of the radiofrequency spectrum.

Finding the right case experimentally

Experimentally, without any control on Δf_0 , the choice of a direct or a reverse retrieval requires an asymmetry on the radiofrequency. This asymmetry can be an absorption line (like in the graphs of this subsection) or an asymmetry of the emission spectrum.

Up to now, a first model of the DCS methodology, from the fs pulses to the retrieved optical spectra, has been developed analytically. However, should some hypotheses be challenged, the analytical modelisation would become much more difficult. To modelise experimental data in this case, a simulation was developed.

2.3 Simulation

The goal of the simulation is to simulate the DCS methodology, from the fs scale of the pulses to the measured experimental rf signal, based on the temporal modelisation of section 2.1. Hence, the simulation can not only validate the modelisation, by comparing its results with experimental data, but also visualise the effect of other parameters. In particular, the effect of a variation of Δf_{rep} on the spectra has not been computed because of the complexity of modelising it analytically, but such variations, that are observed experimentally, can be simulated.

The first subsection summarises the model on which the simulation is based. The second studies the effect of a Δf_{rep} variation on the retrieved spectra.

2.3.1 Model for the simulation

The analytical modelisation of section 2.1 holds for any pulse shape and spectral line shape that respects the hypotheses that are detailed along the development. To do actual computation, one pulse shape and line shape must be chosen. This subsection describes the ones that were chosen for the visualisation of the effect of Δf_{rep} variation. However, the simulation allows to fix arbitrarily all the parameters from section 2.1.

Pulse shape

A gaussian shape with characteristic temporal width τ was chosen. This choice was motivated by the fact that experimental data showed a good consistence with a gaussian model (autocorrelator traces, see section 3.4), and that it makes analytical computations easier.

The hypotheses that both combs share the same carrier frequency and output power are also made. They are motivated by experimental observations (on grating-based spectrometers and powermeters respectively, see section 3.1.3) and make the analytical computation easier.

Hence, formally, the pulse shapes that are used on the simulation are:

$$\mathcal{E}_1(t) = \mathcal{E}_2(t) = \mathcal{E}(t) = \mathcal{E}_0 e^{-\left(\frac{t}{\tau}\right)^2}. \quad (2.76)$$

Thus, equation 2.44 simplifies into:

$$S(t) = \mathcal{E}_0^2 \left(\sqrt{2\pi}\tau + \sqrt{2\pi}\tau e^{-\left(\frac{t/a}{\sqrt{2}\tau}\right)} - 4\pi\tau^2 SNL\theta(0)\Re(\mathcal{A}(0)) - SNL\pi\tau^2 \left[\mathcal{A}(t/a)e^{-i\omega_0 t/a} + \text{c.c.} \right] \right). \quad (2.77)$$

The simulation of the response of the medium (that appears in the last term of this equation) also needs an expression for the line shape $\mathcal{A}$.

Line shape

By taking into account both collisional and Doppler line width in a semi-classical model (developed in appendix A.2), the spectral shape can be written as:

$$\mathcal{A}(t) = \frac{1}{2\pi} e^{-\gamma_{\perp}|t|} e^{-(\Gamma_d t)^2/4} \quad (2.78)$$

with $\gamma_{\perp}$ the characteristic collisional spectral linewidth and Γ_d the characteristic Doppler broadening spectral linewidth. This model is broadly used in literature and accounts well for molecular lines such as the ones studied experimentally in this work [214, 215]. Models beyond this one have been developed and used [216–218], but are beyond the scope of this work.

2.3.2 Effects of Δf_{rep} fluctuations on the spectrum

The fluctuations of Δf_{rep} are observed experimentally, but they are difficult to track (see subsection 3.3). Thus, a simulation can help to visualise their effect on the temporal shape and spectrum of the experimental signal.

Parameters of the simulation

The time-based simulation that was developed during this work enables to make f_{rep} vary during the acquisition of an interferogram, which is equivalent to a shift of Δf_{rep} .

The order of magnitude that was chosen for these Δf_{rep} fluctuations corresponds to a 10^{-7} fs shift over the course of the centerburst, as it was enough to obtain a visible effect on the spectra.

The other parameters that were used for the following simulation are summed up in table 2.1. They were chosen as close to measured experimental parameters as possible.

A single oxygen line was modelised to reduce the computation time. The results obtained on this line can be generalised to the case where multiple lines are considered, due to the linearity of the model.

	parameter	value
Pulses	pulse duration	150 fs
	Central wavelength	768 nm
	Repetition rate	118 MHz
	Δf_{rep}	40 Hz
	Carrier-envelope phase	0 rad
O ₂ line	Central wavelength	761 nm
	Line strength	$4 \cdot 10^{-30}$ Hz/mol · cm ²
	Temperature	295 K
	Pressure	10^5 Pa
	Mixing ratio	0.2
	Lorentz coefficient	$502 \text{ s}^{-1} \text{ bar}^{-1}$
	Propagation length	7 m

Table 2.1: Parameters of the simulation

Results

A linear variation of Δf_{rep} is considered, with an amplitude corresponding to a 10^{-5} Hz shift over the course of the centerburst. The results on the spectrum are presented in figure 2.10.

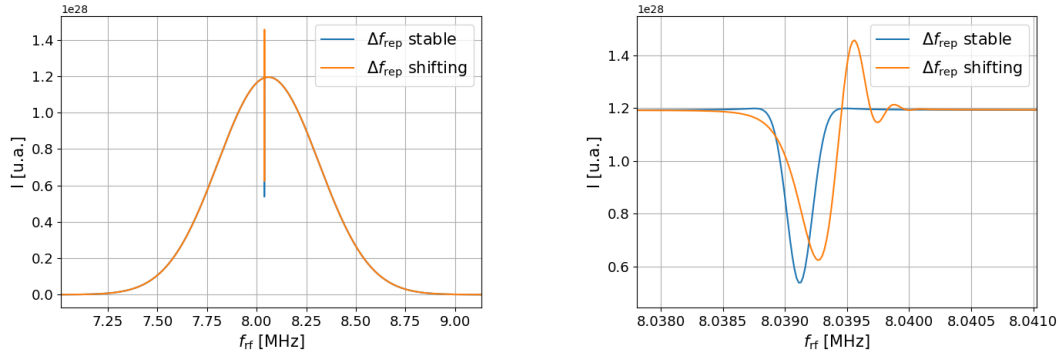


Figure 2.10: Simulated radiofrequency spectrum in the case of a linear variation of Δf_{rep} : spectrum and absorption line (left) and zoom on the absorption line (right), for the stable Δf_{rep} (blue) and the varying Δf_{rep} (orange). The value of the simulated stable Δf_{rep} is 40 Hz. The simulated fluctuations of Δf_{rep} are linear in time, with a variation corresponding to a $\sim 10^{-5}$ Hz shift during the centerburst (variation rate of $66.7 \text{ } \mu\text{Hz/ps}$).

It appears on the result of this simulation that the envelope of the spectrum with drifting Δf_{rep} is unchanged by the fluctuations. On the other hand, the absorption line is distorted and slightly shifted. In particular, it presents an asymmetric profile, that goes over the baseline.

This effect will disturb the retrieval of spectroscopic informations from the spectrum. Indeed, with this line distortion, the link between the optical depth and the concentration of the corresponding molecule is no longer given simply by Beer-Lambert's law. A more precise modelisation,

including the variations of Δf_{rep} , would have to be used in this case to link the optical depth with the concentration. More realistically, the Δf_{rep} fluctuations would be corrected experimentally using a stabilisation process, an adaptive clock for the detection, or post-acquisition correction algorithms.

Up to now, a first model of the DCS methodology, from the fs pulses to the retrieved optical spectra, has been developed. However, nothing has been said on how these pulses are produced. In particular, in our case, UV-DCS requires the production of UV femtosecond pulses, by second harmonic generation (SHG) on the IR pulses that are produced by our laser source. The characterisation of our laser source is the topic of chapter 3, while the next section presents the SHG process we are using.

2.4 Second harmonic generation (SHG) to reach the UV range

The emission spectrum of our Ti:Sa laser is between 750 nm and 820 nm. Thus, to reach the UV range, Second Harmonic Generation (SHG) can be used on this spectrum, to obtain a doubled spectrum between 375 nm and 410 nm.

The aim of this section is not to give a complete presentation of SHG, but to recall theoretical concepts that will be useful in the presentation of experimental results in chapter 4.

2.4.1 Principle of SHG

Second Harmonic Generation (SHG) is a three-wave mixing nonlinear process during which two photons with the same frequency are absorbed by a molecule and one is emitted, the emitted one containing the energy of both incoming ones. Since the energy of a photon is proportional to its frequency, the emitted photon's frequency is twice the incoming ones'.

Formally, this effect is described by the order two dielectric susceptibility of the medium. This component vanishes if the medium has a central symmetry: thus, SHG can only occur on surfaces or in the bulk of non-centrally symmetric media, namely composed of chiral molecules [219, 220].

2.4.2 Phase-matching condition

To be coherently produced, the emitted ("doubled") light must oscillate in phase with the incoming light, giving a phase-matching condition on the optical index of the medium : $n(\omega) = n(2\omega)$ [219, 221]. This condition can practically only be met in some birefringent materials (LBO, BBO, etc.) and custom-made layered materials. Moreover, in these non-isotropic media, the optical index highly depends on the propagation direction: thus, the incoming angle of the light on the SHG medium is a crucial parameter to meet the phase-matching condition. Deviation from this phase-matching angle results in a lower conversion efficiency. This condition is illustrated in figure 2.11.

2.4.3 SHG efficiency optimisation

The efficiency of the frequency doubling, defined as:

$$\alpha = \frac{\mathcal{P}(2\omega)}{\mathcal{P}(\omega)} \quad (2.79)$$

is proportional to the incoming intensity $I(\omega)$. Thus, to optimize this efficiency, one should concentrate the beam in the smallest possible section in the SHG medium.

However, if the beam is too focused, it will enter the crystal under a wide range of angle, which rules out the phase-matching angle condition. Thus, the frequencies for which the phase-matching condition is the most unmet will be doubled with very little efficiency. This tends to reduce the spectral width of the doubled spectrum and eventually hamper the overall efficiency.

Thus, a compromise must be found between these two phenomena to optimise the output power. This compromise was found using a simulation software in our case. A more detailed analytical

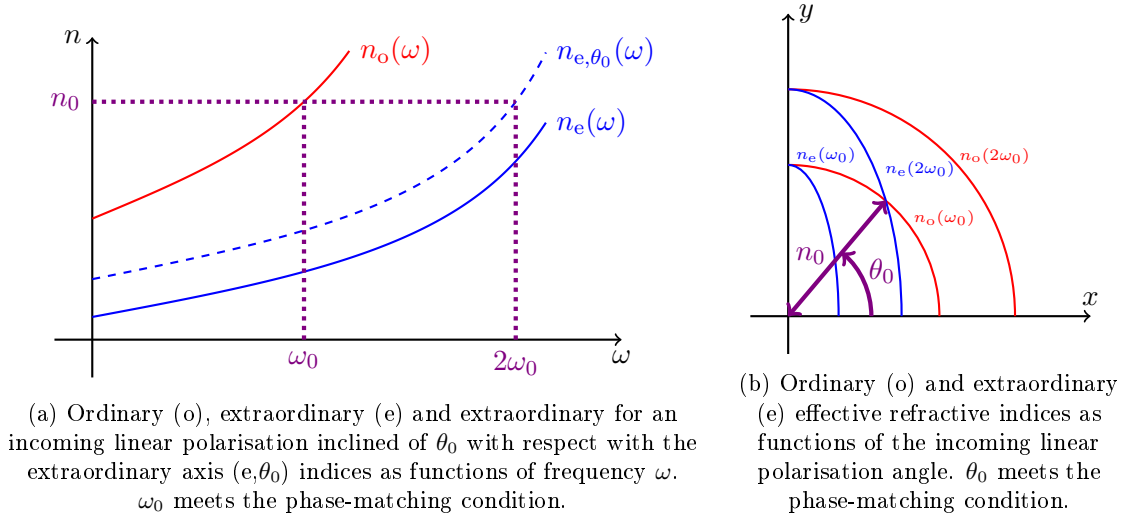


Figure 2.11: Illustration of a case of phase-matching in a birefringent medium.

approach can be found in Bourzeix's PhD thesis [222], continuing the work of Boyd and Kleinman [223, 224].

The length of the crystal is also one crucial parameter: the longer the crystal, the greater the efficiency, provided that the phase-matching condition is met; but this phase-matching condition is all the more precise on the incoming angle as the crystal is long. Thus, another compromise must be found on the length of the crystal.

Conclusion

The developed modelisation of the DCS methodology from the electrical field of a single pulse to the detected, filtered intensity at the temporal scale of the lab, and the associated spectra, allow to predict the different components of the detected experimental signal. It can be used to predict analytically the effect on the retrieved data of some deviations from the ideal case.

Some deviations from the ideal case, such as Δf_{rep} fluctuations, are not treated analytically, but can be simulated numerically. The qualitative effects that are observed are to be linked with experimental data.

The retrieval of the optical frequencies from the radiofrequency signal is non-trivial, and requires an asymmetry on the radiofrequency spectrum to be unambiguous (absorption line or emission spectrum asymmetry).

Reaching the UV range from our NIR Ti:Sa source can be done via Second Harmonic Generation. To optimise the efficiency of this method, one must find a balance between the overall conversion efficiency and the spectral width of the UV spectrum, which implies to choose carefully the different experimental components (focusing lens, SHG crystal).

Chapter 3

Characterisation of the Ti:Sa bidirectional cavity emission

Introduction

As discussed in chapter 1, the laser source that we use for dual comb spectroscopy is a unique free-running bidirectional ring Ti:Sa laser cavity [15]. This chapter aims at characterising the emission of this cavity.

In a first section, the design of the cavity and the characterisation setup are described. Bidirectional cavity were first introduced in 1996 [202], and evolution of their design followed to serve different purposes [205,225]. Originally, in the scope of rotation measurements, the coupling between the two counterpropagating beams was seen as a parasitic effect. To avoid it, the gain medium and the Kerr medium were separated. In our case, based on the design of previous cavities [204,226], this coupling is wanted, as the source of the mutual coherence between the two emission directions, which is needed to perform DCS (see section 2.1.4). Hence, the same Ti:Sa crystal was set to serve as both the gain and Kerr medium. The choice to use a Ti:Sa cavity was motivated by the need of high output power and low noise level, to achieve as good a signal/noise ratio as possible on spectroscopic measurements [180,227].

To reach Kerr-lens mode-locked emission regime (KLM), intracavity dispersion must be managed, hence the presence of a prism compressor, whose effect is detailed.

Finally, the setup used to quantitatively characterise the emission of the cavity, in terms of output power, pulse duration, spectral width and central wavelength, and especially repetition rate difference between the two emission directions (Δf_{rep}) is presented.

This Δf_{rep} is a central parameter for DCS, as seen in chapter 2.1. In our case, it is retrieved from the temporal trace of the interference between the two directions (namely interferograms (IGM)) using an algorithm. Thus, the contrast of the interferograms must be optimised. To do so, a wavefront adaptation strategy is set up [228], based on beam size measurements, that are detailed in a second section.

The third section focuses on Δf_{rep} . Its retrieval protocol is detailed. This retrieval protocol allows to sample the fluctuations of Δf_{rep} : they are quantified as well, and compared with those of the absolute repetition rate of the two directions ($f_{\text{rep},1}$ and 2). Finally, the variation of Δf_{rep} is quantitatively linked to experimental variations of cavity parameters (pump power, insertion of the prisms of the prism compressor).

The fourth section focuses on the characterisation of the emitted pulses, whose interferences form the aforementioned interferograms. They are quantitatively characterised in terms of pulse duration,

dispersion, central wavelength and spectral width. The link of these latter quantities with Δf_{rep} is also investigated.

Finally, the fifth and sixth sections aim at proposing a microscopic explanation for the existence of Δf_{rep} . Section five focuses on the self-steepening effect, a non-linear effect that takes places in the Ti:Sa crystal [204, 229], while section six focuses on chromatic dispersion in the cavity to explain the presence of Δf_{rep} . Both hypotheses are based on experimental measurements.

3.1 Description of the setup and parameters

3.1.1 Design of the homemade bidirectional cavity

To produce the two mutually coherent frequency combs lasers that are necessary to use DCS, a homemade, freerunning bidirectional ring cavity is used [15]. A schema of this cavity and the associated detection setup is given in figure 3.1.

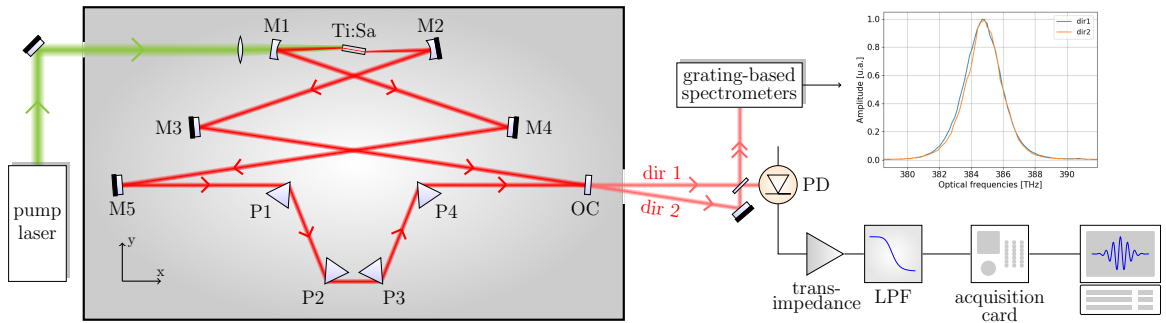


Figure 3.1: Schema of the bidirectional cavity and the detection setup. The pump laser is focused inside a Ti:Sa crystal, which produces the laser radiation. The light propagates in both directions within the cavity. The mirrors are numbered from 1 to 5 (M1, M2, etc.). The compressor's prisms are numbered from 1 to 4 (P1, P2, etc.). The light is emitted out of the cavity through an output coupler (OC). The two outcoming beams (directions 1 and 2, dir1 and dir2) are then superimposed on a beamsplitter and focused on a photodiode (PD). The temporal trace of the output electrical signal of this photodiode is amplified by a transimpedance, filtered by a low-pass filter (LPF) and sampled by an acquisition card, before being visualised on a computer. Part of the beams is also sent to grating-based spectrometers, that allow to visualise the spectra of the two directions live. One round-trip inside the cavity is approximately 3 m long, corresponding to a 118 MHz repetition rate in Kerr-lens mode-locked emission.

A green pump laser (Coherent®Verdi V5, 5 W output power at 532 nm central wavelength) is focused by a 100 mm focal length convex lens inside a 10 mm long titanium sapphire (Ti:Sa) crystal. The crystal is cut at Brewster angle to avoid unwanted reflections. The focus point of the pump laser is slightly off from the center of the crystal; this asymmetry is strongly linked with the repetition rate difference between the two directions, as will be seen further on in subsection 3.5. The ring cavity allows a bidirectional laser emission: the beams will be called dir1 and dir2 in the following. The cavity emits through a wedged output coupler with reflectivity 90%.

The two beams share two waist locations, one in the middle of the curved M1 and M2 mirrors and one in the middle of the P2 and P3 prisms.

A small tilt on the pump lens corrects for the pump laser's astigmatism due to the M1 mirror. The tilt on the M1 and M2 mirrors corrects for the astigmatism due to the Brewster cut of the crystal.

A prisms compressor is used for dispersion management and Kerr-lens mode-locked regime (KLM) triggering. Its effect is detailed in the next subsection (3.1.2).

The optical table is put within a laminar air flow to minimize the presence of dust. The cavity is inside a foam box to minimize acoustic noise. Both the pump laser and the Ti:Sa laser are thermalised.

3.1.2 Adjustable prisms compressor for dispersion management

To generate frequency combs, a Kerr-lens mode-locked (KLM) emission from the cavity is used [230–232]. This regime must be triggered, and its stability must be ensured. Its stability highly depends on the net dispersion of the cavity. Moreover, it can be triggered through soft-aperture mode-locking by the movement of an element in the cavity. Thus, an adjustable prisms compressor was mounted in the cavity, to manage the net dispersion and add a movable component.

The dispersion of the cavity pulses is quantified by the group delay dispersion GDD, defined as the second derivative of the spectral phase with respect to pulsation ω :

$$\text{GDD} = \frac{\partial^2 \varphi}{\partial \omega^2} \quad (3.1)$$

The adjustable part of the prism compressor is the two bottom prisms of the compressor (P2 and P3 in figure 3.1), that are mounted on a translation stage. The latter can be translated along the compressor's symmetry axis (y axis on figure 3.1). This translation is used to trigger Kerr-lens mode lock regime (KLM) by a short mechanical translation, with the motorised stage or by hand, at a rate of ~ 1 to 10 Hz. Once the KLM regime is triggered, this translation is also used to change the negative dispersion induced by the compressor.

The negative dispersion of the compressor is related to the position of the prisms t (shown in figure 3.2) by the formula 3.2 [233]:

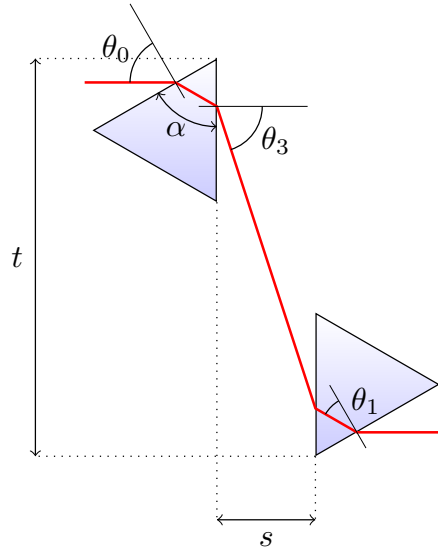


Figure 3.2: Schema of half of the compressor. Both prisms are identical, their shape is an isosceles triangle, characterised by their apex angle α . The angles θ_0 , θ_1 and θ_3 are defined with respect with the line perpendicular to the prism surface. The red line corresponds to the trajectory of the laser beam.

$$\text{GDD} = \frac{\lambda_l^3}{2\pi c^2} \left[L_g \frac{d^2 n}{d\lambda^2} \Big|_{\lambda_l} - \left(4L + \frac{L_g}{n^3} \right) \left(\frac{dn}{d\lambda} \Big|_{\lambda_l} \right)^2 \right] \quad (3.2)$$

with n the refractive index of the prism, λ_l the spectrum's central wavelength, L the average distance traveled though air and L_g the average distance traveled though glass, given by:

$$\begin{cases} L_g = (t - s \tan \theta_3) \frac{\sin \alpha}{\cos \theta_1} \\ L = s / \cos \theta_3 \end{cases} \quad (3.3)$$

Moreover, Snell-Descartes laws give:

$$\begin{cases} \theta_1 = \arcsin(\sin(\theta_0)/n) \\ \theta_3 = \arcsin(n \sin(\alpha - \arcsin(\sin(\theta_0)/n))) \end{cases} \quad (3.4)$$

In our case (SF₁₀ prisms), $\theta_0 \simeq 60^\circ$ and $\alpha = 59.7^\circ$ (Brewster angle of SF₁₀ at 800 nm [234]). The parameter s is worth 11.2 cm and the parameter t is around 20.1 cm; it is varied over a few millimeters.

Our setup does not allow to retrieve precisely the absolute value of this negative GDD because θ_0 cannot be measured accurately (up to a few degrees). Moreover, θ_0 is very likely to vary when realigning the cavity. However, the variation of the distance t is accurately (up to 10^{-4} m) changed and monitored with a motor-driven translation stage that is monitored on the local computer. Combined with the formula above (equation 3.2), this control allows to retrieve the variation of the compressor's negative GDD.

3.1.3 Characterisation setup

To characterise the emission of the cavity, several elements were set up. A schema of this detection setup is presented in figure 3.3.

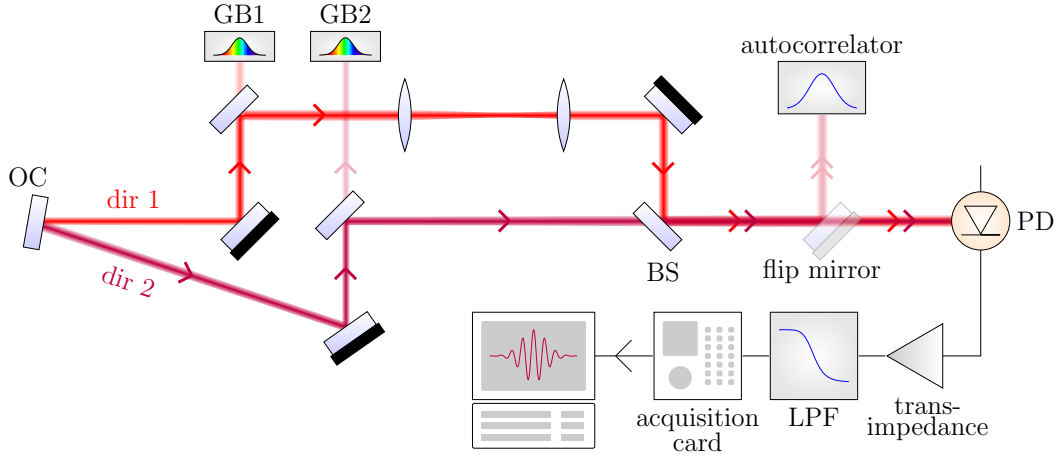


Figure 3.3: Schema of the detection setup that was used to characterise the emission of the cavity. The two beams (dir1 and dir2, for direction 1 and direction 2) are emitted in different direction through the cavity's output coupler (OC). They are later superimposed on a 50/50 beamsplitter (BS) and focused on a photodiode (PD). The output signal of this photodiode is amplified by a transimpedance, filtered by an electronic low-pass filter (LPF) and sampled by an acquisition card, connected to a local computer. Mirror losses of both directions are sent to grating-based spectrometers (GB1 and GB2). Lenses are used to adapt direction 1's wavefront, as will be seen on section 3.2. A flip mirror allows to divert both beams to an autocorrelator to measure their pulse duration, as will be seen on section 3.4.

The quantities that were measured and the instruments that were used are summarized in table 3.1.

observable	notation	instrument	order of magnitude
pump power	$\mathcal{P}_{\text{pump}}$	pump laser interface	5 W
output power (dir1/dir2)	$\mathcal{P}_1/\mathcal{P}_2$	powermeter	200 mW
power spectra shape	$\hat{S}_1/\hat{S}_2$	grating-based spectrometers	5-10 nm-wide
pulses duration	τ_1/τ_2	commercial autocorrelator	100-300 fs
P2+P3 prisms position	x_{prisms}	translation stage interface	5 mm
repetition rate difference	Δf_{rep}	see next subsection	1-100 Hz

Table 3.1: Measured output parameters of the cavity

The output power of the pump laser is monitored through a user interface up to a 0.01 W precision. Inside the cavity, the position of prisms P2 and P3 (fixed on the same translation stage) can be changed both manually and via a motor, which is monitored by a user interface up to a precision of 100 μm . Outside the cavity, the power of each direction is measured with a powermeter with a ~ 3 mW precision. The losses of two mirrors are analysed by grating-based spectrometers (Avaspec-3648 and Avaspec-ulS4096cl-evo from Optoprim) to give the output spectra of both directions with a $\simeq 1$ nm resolution. The pulse durations are measured using an autocorrelator (APE Pulsecheck). To record the temporal trace of the beating of the two combs, the beams are focused on a fast photodiode (FDS010 from Thorlabs), whose signal is amplified by a transimpedance (DHPCA-100 from FEMTO), filtered by a homemade 50 MHz 4th order low pass filter and numerised by an acquisition card (ATS9440 from ACQUYSIS). To optimise the contrast of this temporal trace, the wavefronts of both directions were matched, as detailed in section 3.2. Finally, the Δf_{rep} parameter can be retrieved from the temporal trace of the beating of the two combs, as detailed in section 3.3.

3.2 Wavefront management

As seen in chapter 2, the DCS methodology relies on the detection of the beating between two combs. Thus, the contrast of the interference patterns on the photodetector is crucial. To optimise this contrast, the wavefronts of the two interfering beams must be as similar as possible [228], both in terms of curvature and beam size. To ensure this, the waist positions of both directions and their wavefront as a function of space were both retrieved, before adapting the wavefronts to optimise the contrast on our photodetector.

3.2.1 Wavefront retrieval protocol

Assuming a circular (not elliptical) gaussian shape for the beam, the evolution of its wavefront during a free-space propagation is completely characterised by only two parameters (waist position z_0 and minimum waist size w_0) [235]. In particular, the evolution of its waist is given by:

$$w(z) = w_0 \sqrt{1 + \left(\frac{z - z_0}{z_R} \right)^2} \quad (3.5)$$

with z the distance on the direction of propagation and $z_R = \pi w_0^2 n / \lambda$ the Rayleigh length (with n the refractive index of the propagation medium and λ the central wavelength of the carrier).

Thus, given the knowledge of the beam waist as a function of space for several points, the propagation parameters z_0 and w_0 can be retrieved through a numerical fit.

The underlying assumption of a gaussian shape for the beam was verified experimentally during the process, as will be seen in this same subsection.

3.2.2 Setup of beam retrieval measurement experimental setup: chopper and retrieval algorithm

The waist of the beam is measured for several propagation distances z . At a given position, to retrieve the waist of the beam, the following protocol is followed.

A chopper (opaque wheel with regularly spaced holes, as illustrated in schema 3.4), whose rotation rate $\dot{\theta}$ and distance to the center of the beam r are well-known is placed on the trajectory of the beam. The temporal evolution of the light intensity that goes through the chopper $I(t)$ is recorded thanks to an amplified photodiode.

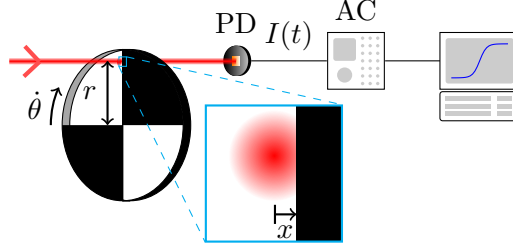


Figure 3.4: Schema of the setup used for waist measurement. The beam goes through the rotating wheel, the output intensity depends on the position x of the blade. The output intensity is recorded by an amplified photodiode (PD). The output signal $I(t)$ of this photodiode is sampled by an acquisition card (AC) and analysed on a computer through a Labview interface.

With the hypothesis that the waist is small compared with the distance to the center of the chopper ($w \ll r$), one can show that the light intensity depends on the position of the chopper as:

$$I(x) \propto \text{erf} \left(\frac{x}{w(z)} \right) = \int_{-\infty}^x \int_{-\infty}^{+\infty} e^{-\frac{x'^2 + y'^2}{w(z)^2}} dx' dy'. \quad (3.6)$$

Thus, for a given rotation rate $\dot{\theta}$ and radius r , while the chopper is passing through the beam, the intensity as a function of time is given by:

$$I(t) \propto \text{erf} \left(\frac{r\dot{\theta}t}{w(z)} \right) \quad (3.7)$$

from which $w(z)$ (the only unknown parameter) can be retrieved thanks to a numerical fit.

This method enables to perform many measurements within a short period of time, provided that the rotation rate is high enough. In our case, the use of a rotation rate of the order of 50 turns/s and a chopper with 10 holes allowed to perform several hundreds of measurements, allowing to correctly evaluate the statistical uncertainty.

3.2.3 Results of waist measurements

A chopper with 10 holes was used, rotating at an angular speed of $\dot{\theta} \sim 50$ rotations/s, with the beam at a distance $r \sim 10$ cm of the center of the chopper. A Labview interface allowed to perform real-time measurement and averaging, and gave a statistical uncertainty on the measure, as presented in figure 3.5.

The Labview algorithm performs a derivative of the measured signal, before fitting the derivative with a gaussian function. This method allows to retrieve the beam size w , linked with the temporal width of the gaussian σ_t and the setup parameters via the relation:

$$w = \sigma_t r \dot{\theta}. \quad (3.8)$$

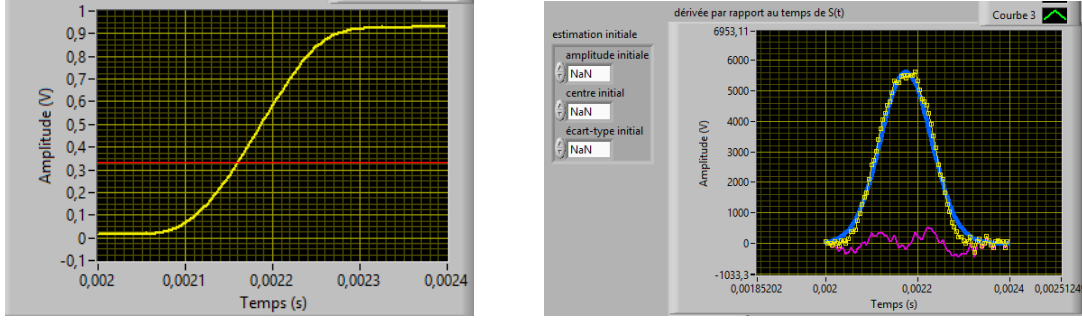


Figure 3.5: Experimental signal obtained with the experimental setup from figure 3.4 (left), numerical derivative of this signal (yellow), gaussian fit (blue) and fit residues (pink) (right)

The uncertainties of the fit parameters are very small compared to the statistical uncertainties.

The retrieved waists ranged from 0.7 mm to 2 mm, with a typical relative uncertainty of the order of 10^{-2} , over a distance of the order of 1 meter. This range is compatible with the ABCD matrix-based modelisation of the beam. Moreover, the qualitative good adjustment of the fit and the small relative incertitude allow to validate the gaussian beam shape assumption a posteriori.

3.2.4 Beam propagation parameters retrieval

These measurements allowed to retrieve the propagation parameters of the two beams (waist position with respect with the output coupler z_0 and waist size w_0). The results of the measurements are presented in figure 3.6 and the retrieved parameters are given in table 3.2.

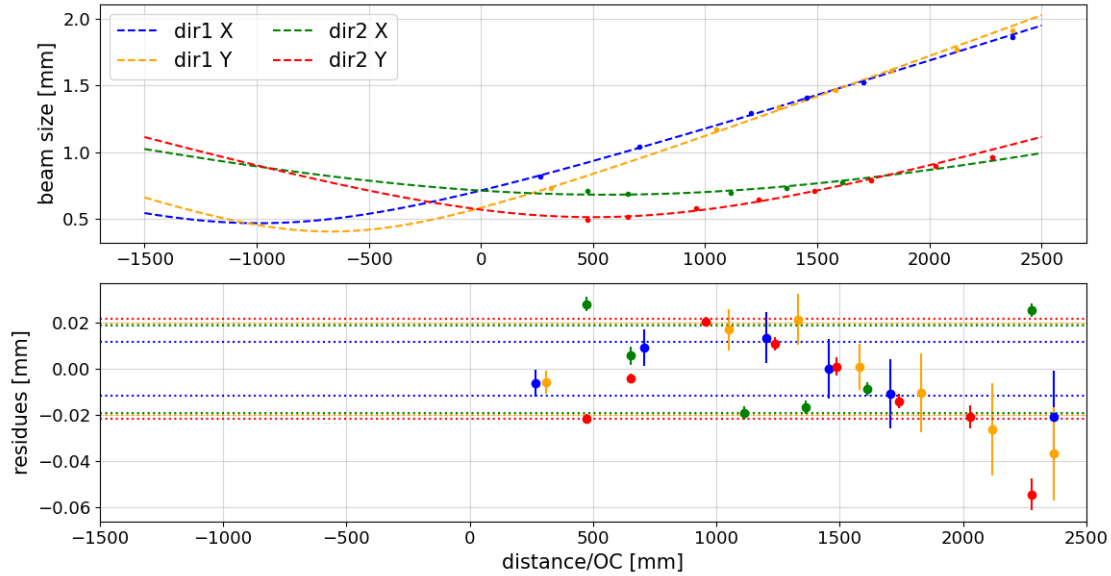


Figure 3.6: Beam size measurements on the X and Y axis for directions 1 and 2 as functions of the distance with respect with the output coupler (OC) and fit with the gaussian propagation model. (Top) Beam size measurements using the aforementioned method (points) and fit with the gaussian propagation model function (equation 3.5) (dashed curves). (Bottom) Residus of these fits. The error bars correspond to the statistical uncertainty retrieved with the aforementioned algorithm. The dotted lines give the standard deviation of each set of data. The retrieved propagation parameters are

w_0 [μm]	x axis	y axis	z_0 [mm]	x axis	y axis
dir1	466 ± 6	403 ± 6	dir1	-990 ± 21	-670 ± 21
dir2	680 ± 15	511 ± 9	dir2	560 ± 73	500 ± 33

Table 3.2: Retrieved output parameters of the cavity

The propagation parameters were retrieved with a relative uncertainty of the order of 10^{-2} . In the case of the waist position, it corresponds to an absolute uncertainty of the order of a centimeter. This corresponds to the order of magnitude of the precision that we can reach on the position of the lenses that will be used to adapt the wavefront. Thus, this precision is high enough for our use of these data.

The difference between the x and y axis can be attributed to the astigmatism of the pump laser after going through the M1 mirror (see figure 3.1). It is relatively small and will be neglected in the following. To treat it without modifying the cavity setup, anisotropic optics would be needed.

The residus of all four series of measures show a clear tendency, which seems to show that the gaussian model must be refined to describe our system at this level of precision. To quantify this distance to the gaussian model, M^2 formalism could be used [236]. This, however, is beyond the scope of this work.

The retrieved propagation parameters allowed to proceed with the wavefront adaptation protocol, which is presented along with the associated spectroscopy results in chapter 4.

3.3 Δf_{rep} measurement and dependance

The repetition rate difference between the two directions Δf_{rep} is the central parameter for DCS.

3.3.1 Δf_{rep} retrieval with interferometry

To retrieve Δf_{rep} from the temporal trace of the interferograms, the property (mentioned in section 2.1.5) that the repetition rate difference corresponds to the inverse of the duration between two consecutive interferograms (IGMs) was used.

To acquire the temporal trace of the interferograms, the two beams are combined on a photodiode thanks to a 50/50 beamsplitter, as illustrated on figure 3.3.

To retrieve $1/\Delta f_{\text{rep}}$ from this temporal trace, an algorithm is used. This algorithm first performs a basic peak finding to find the approximate position of each interferogram. Then, each interferogram is fitted with a test function (gaussian envelope multiplied with a dephased sinusoid), from which the central position of the interferogram is retrieved (as the central position of the envelope term).

An example of this process is given in figure 3.7.

The difference between centerbursts of consecutive interferograms allows to sample $1/\Delta f_{\text{rep}}$ (see section 2.1.5) and its fluctuations, at the acquisition rate of Δf_{rep} (one point per interferogram).

The retrieved values of Δf_{rep} range from 0 to approximately 200 Hz, depending on several parameters (see subsection 3.3.3). For a given regime, its fluctuations are typically 1 Hz large, as presented in figure 3.8.

The top graphs show that the centerburst position varies linearly with the interferogram number, which shows that Δf_{rep} is globally stable. The residues of their linear fit, presented in the bottom graphs, allow to quantify Δf_{rep} fluctuations. As can be seen on these graphs, Δf_{rep} fluctuations are one or two order of magnitude lower than the average Δf_{rep} value.

The experimental consequences of these fluctuations in the optical spectra retrieval process is a stretch of the radiofrequency spectra, different for each interferogram, that must be corrected, as will be further detailed in section 4.1.

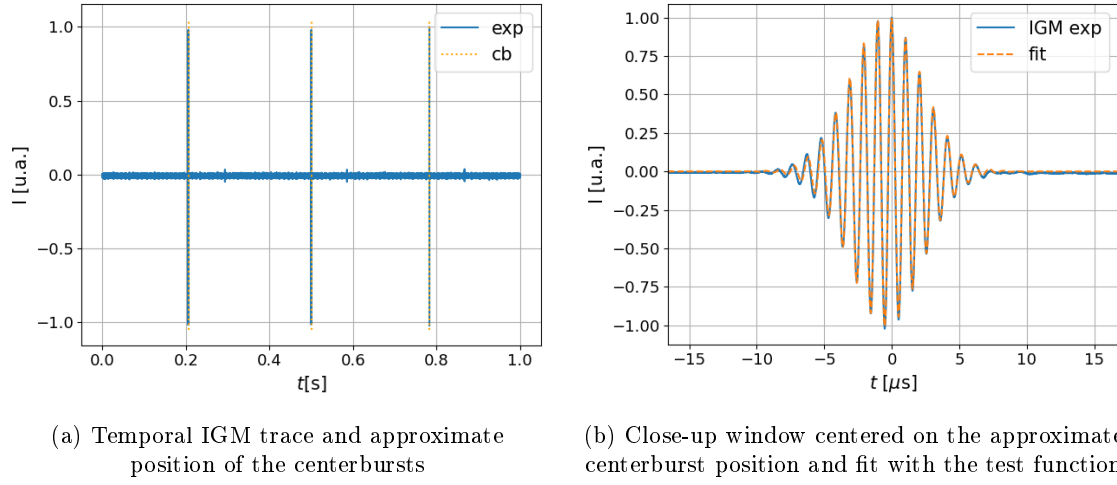


Figure 3.7: IGM experimental temporal trace and results of the two steps of the centerburst retrieval algorithm. (Left) A first peak finding step allows to retrieve the centerbursts (orange vertical lines) with a precision of the order of one IGM oscillation. (Right) A second step fits the interferogram with the product of a gaussian envelope and a dephased cosinus (orange dashed line). The central position of the retrieved envelope term corresponds to the centerburst.

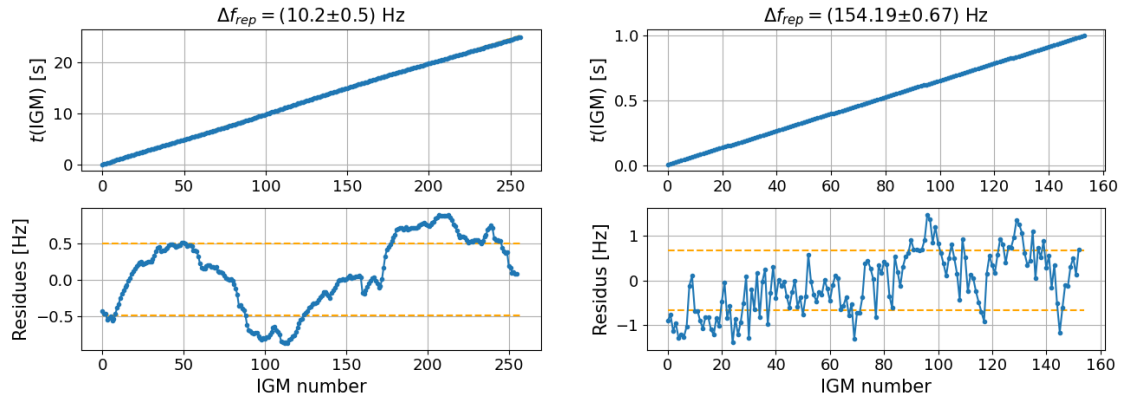


Figure 3.8: Examples of retrieved Δf_{rep} fluctuations in low Δf_{rep} regime (left) and high Δf_{rep} regime (right). In both subfigures, the temporal position of the centerbursts with respect with their number is given on the top graph, and residuals from linear fit are given on the bottom graph. The orange dashed line correspond to the standard deviation.

3.3.2 Δf_{rep} stability compared with f_{rep} stability

In the retrieval process of optical spectra from DCS temporal traces, not only Δf_{rep} but $f_{\text{rep},1}$ and $f_{\text{rep},2}$ are also involved (see section 2.2). Thus, the fluctuations of $f_{\text{rep},1}$ and $f_{\text{rep},2}$ were quantified.

To measure the fluctuations of f_{rep} in both directions, a frequency counter (FCA3000 from Tektronix) with several input ports was used. The two beams were sent each into a input port and measured simultaneously. The result is shown in figure 3.9.

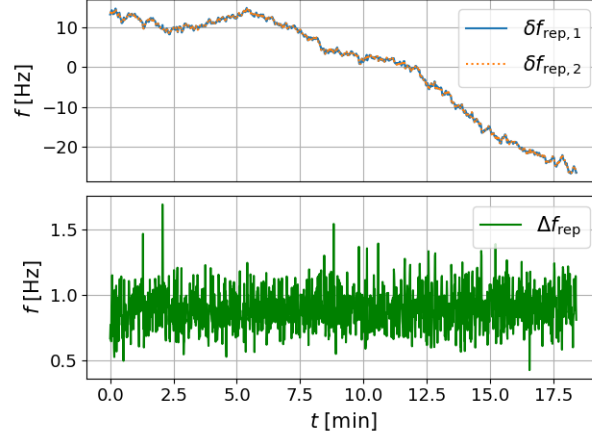


Figure 3.9: Fluctuations of $f_{\text{rep},1}$ and $f_{\text{rep},2}$ around their mean value (top) and Δf_{rep} value given by the difference $|f_{\text{rep},2} - f_{\text{rep},1}|$ over the course of ~ 18 minutes, measured with a frequency counter. The average value of f_{rep} is ~ 118 MHz, the average of Δf_{rep} is ~ 0.9 Hz.

It appears clearly that Δf_{rep} fluctuations are at least one order of magnitude lower than f_{rep} fluctuations. This tends to show that more than 90 % of f_{rep} fluctuations are shared by both directions. Hence, Δf_{rep} is much more stable than f_{rep} . This can be attributed to the fact that both beams share the same pump laser, gain medium and cavity, thus some of the fluctuations due to these elements.

However, the relative fluctuations of f_{rep} (of the order of 10^{-5}) are much smaller than those of Δf_{rep} (of the order of 10^{-1} in this case). As a result, Δf_{rep} is likely to be a greater source of uncertainty in the process of optical frequencies retrieval.

3.3.3 Dependence with respect with cavity parameters

The dependence of Δf_{rep} with respect with experimental parameters of the cavity (namely prisms insertion and pump power) was quantified. To do so, Δf_{rep} was measured (through the aforementioned protocol) while varying these parameters, without breaking the KLM regime. The results are shown in figure 3.10.

Qualitatively, these results show clear tendencies: Δf_{rep} increases with the prisms insertion (which corresponds to a greater negative dispersion induced by the compressor) and decreases with the pump power.

The increase of Δf_{rep} with the prism insertion can be interpreted as such: a greater prisms insertion induces more negative GDD. This additional negative GDD tends to shorten the pulses without decreasing their energy, thus increasing their peak power. Hence, nonlinear effects in the crystal, that depend on the peak power and that may be the cause of the existence of Δf_{rep} (as will be seen later on section 3.5), have a qualitatively greater effect on the emission.

The decrease of Δf_{rep} with the pump power is more difficult to interpret, because it has an effect on the emission through the stimulated emission in the Ti:Sa crystal, which is a complex, nonlinear effect.

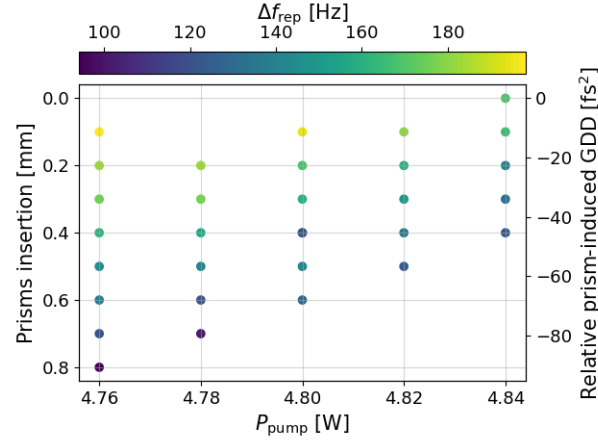


Figure 3.10: Retrieved Δf_{rep} values vs pump power and relative prisms insertion. The prisms insertion correspond to the variation of the t parameter of figure 3.2 from an arbitrary start ((20.1 ± 0.2) cm). The corresponding relative negative GDD induced by the prisms compressor is indicated on the right y axis.

Quantitatively, the fact that a variation of the order of 1 % on these parameters result in a factor 2 change on Δf_{rep} show the sensitivity of the setup.

The limits of the variations of the parameters were chosen because of the instability of the regime: too big a decrease of power would have caused the KLM regime to drop, just like too big or too little a prism insertion. However, within these bounds (that are not controlled experimentally), Δf_{rep} can be controlled through these experimental parameters.

3.3.4 Baseline depletion

The temporal trace from which Δf_{rep} is retrieved presents a baseline. This baseline is removed during the algorithmic process that allows to retrieve Δf_{rep} . However, its removal implies to quantify its value and fluctuations. In particular, a depletion appears once per IGM cycle on the measured temporal trace baseline, as can be seen on figure 3.11.

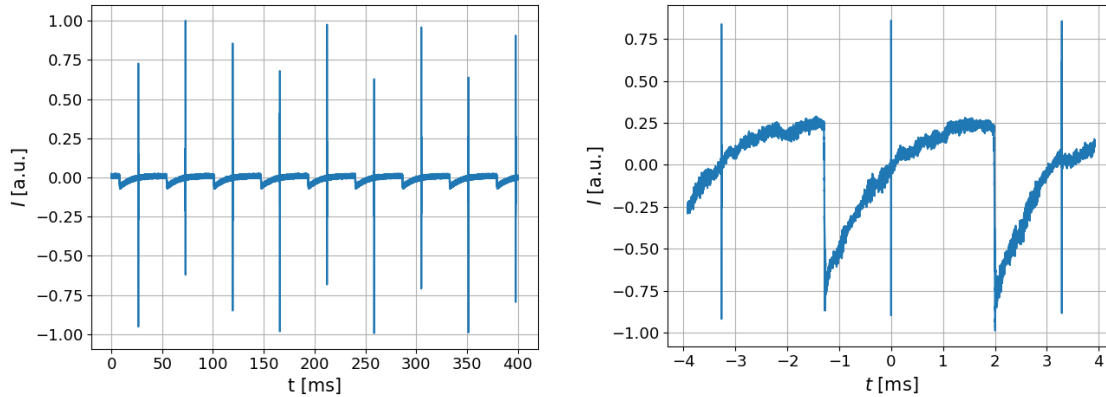


Figure 3.11: Temporal IGM traces presenting baseline depletions in cases of low (left, ≈ 20 Hz) and high (right, ≈ 150 Hz) Δf_{rep}

The amplitude of the depletion compared with the normalised interferogram increases with Δf_{rep} , as can be seen on figure 3.11. It appears with the same periodicity that the IGM (once every

$1/\Delta f_{\text{rep}}$), which shows it is linked with an intracavity phenomenon. More precisely, it appears when the two counterpropagating pulses meet inside the Ti:Sa crystal. It can be interpreted by a gain competition in the crystal that eventually lowers the pulses gain, thus the pulses' power and the temporal trace baseline. A quantitative approach of this phenomenon would require accurate modelisation of the pulses' propagation inside the Ti:Sa crystal, which is beyond the scope of this work. Up to our knowledge, no result has been published yet about such a depletion effect in bilinear cavities.

3.4 KLM pulses characterisation

As seen on chapter 2.1, the pulses that are emitted by the cavity are directly linked with the experimentally measured DCS temporal trace. Likewise, their spectra is directly linked with the spectral envelope on which absorption lines appear. Thus, the characterisation of the pulses is necessary.

3.4.1 Pulse duration

Autocorrelation measurements

To measure the duration of the pulses, autocorrelation measurements were carried out using a commercial autocorrelator (pulseCheck NX from APE). The principle of the measure is presented here.

The principle of an autocorrelator is presented in figure 3.12, along with the specific method our autocorrelator uses (SHG).

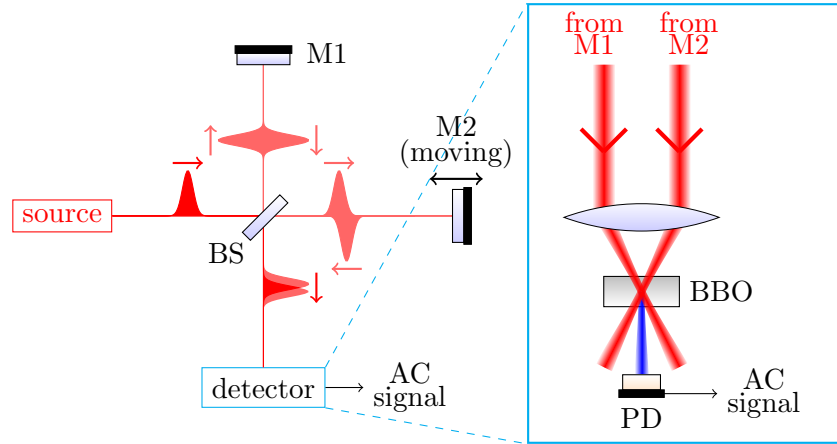


Figure 3.12: Principle of SHG autocorrelator. *Left*: the pulse is split into two arms using a beam-splitter (BS), then recombined and detected. The length of one of the two arms can be controlled thanks to a moving mirror, changing the delay between the two recombined pulses. Knowing the speed of the moving mirror, the analysis of the output signal of the photodetector (autocorrelation (AC) signal) allows to retrieve the pulse duration. *Right*: the detector of our autocorrelator detects the second harmonic intensity produced by the interference of the two recombined pulses.

To modelise our pulses, experimental measurements (detailed later on this same subsection) tend to show that a "sech" temporal shape for the field is a good approximation (more than a gaussian shape):

$$E_s(t) = \text{sech}\left(\frac{\pi t}{2\tau}\right) e^{-i\omega t} = \frac{1}{\cosh\left(\frac{\pi t}{2\tau}\right)} e^{-i\omega t} \quad (3.9)$$

This pulse is split in two by a beamsplitter. The two new pulses travel back and forth through their own interferometer arm and are recombined into a BBO crystal with symmetrical angles (see figure 3.12). As seen in section 2.4, a second harmonic field is produced for an angle satisfying the phase-matching condition (see subsection 2.4.2), namely the average of the two incoming angles in this case. For a given optical length difference between the two arms δ and $k = \omega/c$ the wave vector, the produced SHG instantaneous field is given (up to a constant) by:

$$E_{AC,ist}(t) = e^{-2i\omega t} \text{sech}\left(\frac{\pi t'}{2\tau}\right) \text{sech}\left(\frac{\pi t' - \delta/c}{2\tau}\right) e^{ik\delta} \quad (3.10)$$

which corresponds to the instantaneous intensity:

$$I_{AC,ist}(t) = \text{sech}\left(\frac{\pi t'}{2\tau}\right)^2 \text{sech}\left(\frac{\pi t' - \delta/c}{2\tau}\right)^2. \quad (3.11)$$

The experimental output signal of the photodiode corresponds to the average of this instantaneous signal (see subsection 2.1.3) [237]:

$$I_{AC,s}(t) = \int_{-\infty}^{+\infty} \text{sech}^2\left(\frac{\pi t'}{2\tau}\right) \text{sech}^2\left(\frac{\pi t' - \delta/c}{2\tau}\right) dt' = \frac{4}{\sinh^2\left(\frac{\pi \delta/c}{2\tau}\right)} \left(\frac{\cosh\left(\frac{\pi \delta/c}{2\tau}\right)}{\sinh\left(\frac{\pi \delta/c}{2\tau}\right)} \frac{\pi \delta/c}{2\tau} - 1 \right) \quad (3.12)$$

and in the case of a linearly moving mirror M2 ($\delta = v_0 t$):

$$I_{AC}(t) = \frac{4}{\sinh^2\left(\frac{\pi v_0 t}{2c\tau}\right)} \left(\frac{\cosh\left(\frac{\pi v_0 t}{2c\tau}\right)}{\sinh\left(\frac{\pi v_0 t}{2c\tau}\right)} \frac{\pi v_0 t}{2c\tau} - 1 \right). \quad (3.13)$$

Thus, provided that the speed of the mirror v_0 is known, one can retrieve the duration of the pulse τ from this signal with a numerical fit.

The general autocorrelation protocol, allows to measure short pulses (compared with the response time of our photodetectors) by "stretching" them, by a factor v_0/c that appears in equation 3.13. However, it does not provide any information on the phase of the pulse, nor can it account for any temporal asymmetry of the pulse. Moreover, an hypothesis on the pulse shape (sech here) is needed to retrieve the pulse duration from the autocorrelation function duration.

Compared with other autocorrelation methods [238], the SHG autocorrelation method allows to directly measure the autocorrelation function of the intensity of the pulse, making it immune to any temporal phase of the pulses (see equation 3.11). However, SHG is more energy-demanding than regular beating between the two arms, and needs precise alignment of the focus points of both arms inside the crystal and accurate phase-matching [233].

The assumption of a sech shape for the field is motivated by the fact that it corresponds to the shape of a soliton propagating inside a Kerr medium [219,220] such as the Ti:Sa crystal in our cavity. In our case, the hypothesis of a perfectly solitonic propagation all along the cavity is not justified, as will be seen in subsection 3.4.2. However, as can be seen on figure 3.13 for example, the fit of our autocorrelation function by a "sech²" function give smaller residues than its fit with a gaussian, suggesting that the sech² model describes our pulses more accurately than the gaussian model.

Our commercial autocorrelator allowed to measure autocorrelation traces continuously, which allowed to estimate a statistical uncertainty on the retrieved pulse durations. This statistical uncertainty was much larger than the numerical uncertainty of the fits. This uncertainty was all the more important as the baseline was perturbed by the depletion described in subsection 3.3.4. It ranged from 1 to 10 % of the retrieved pulse duration.

The absolute retrieved pulse durations ranged from 100 to 300 fs, depending on various parameters such as Δf_{rep} .

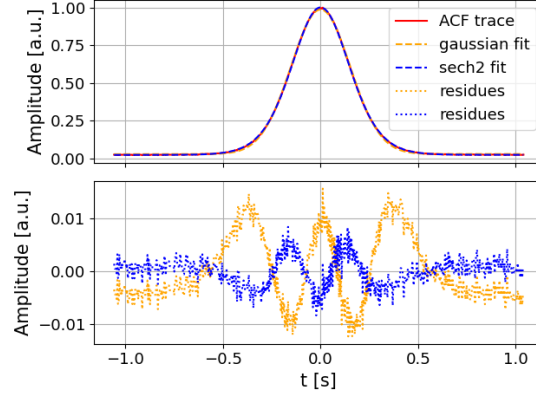


Figure 3.13: Autocorrelation trace of a pulse, gaussian and sech^2 fits. The result of the gaussian fit is given in orange and the result of a sech^2 fit in blue, along with their corresponding residues. The std of the residues was 0.0062 for the gaussian fit and 0.0026 for the sech^2 fit.

Dependence with respect with Δf_{rep}

To quantify the dependence of the pulse durations with respect with Δf_{rep} , the pulse durations were retrieved with our autocorrelator while making Δf_{rep} vary. To make Δf_{rep} vary, variations of pump power and prisms insertion were used, as described in subsection 3.3.2. The results are presented in figure 3.14.

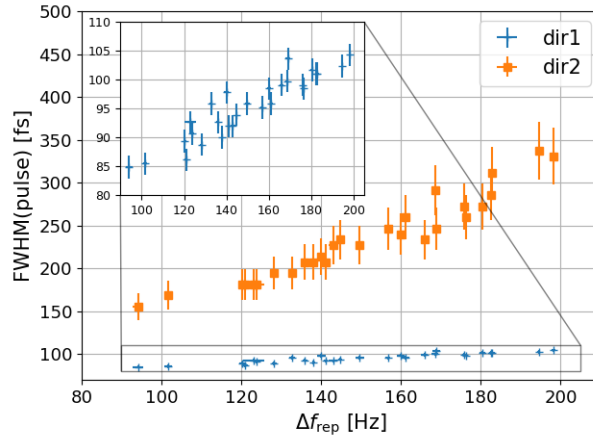


Figure 3.14: Retrieved FWHM of the pulses, from a sech^2 model numerical fit on the autocorrelation traces. The parameters that were changed to make Δf_{rep} vary were the pump power (from 4.76 W to 4.84 W) and the prisms compressor position (over the course of 0.6 mm), without losing the bidirectional KLM regime.

Pulse durations are systematically greater in direction 2 than in direction 1. Moreover, an increasing dependency with respect with Δf_{rep} is observed for both directions. Direction 1 presents a much greater variation and relative variation than direction 2: direction 2 duration variation is ~ 250 fs, from 150 fs to 350 fs, which is more than a 100% increase, while direction 1 varies from 85 to 105 fs, which corresponds to a $\sim 20\%$ increase.

These differences can be interpreted in terms of global delay dispersion (GDD). As will be seen further on, positive GDD tend to broaden the pulses' duration. The direction 1 beam is taken from the cavity right after coming out of the prisms compressor, which minimises its dispersion, whereas

the direction 2 beam is taken after its amplification and reflection on two (slightly dispersive) mirrors. Thus, not only is the direction 2 beam likely to have a higher absolute value of GDD than direction 1, but it also is more likely to be sensitive to GDD fluctuations. Hence, direction 2 presents a greater pulse duration than direction 1, and a greater pulse duration variation for the same change of experimental parameters.

The interpretation of these pulse duration measurements showed the need for a quantification of the pulses GDD inside the cavity.

3.4.2 Dispersion inside the cavity

To better quantify the pulses durations after exiting the cavity, their dispersion inside the cavity was quantified. To do so, a semi-theoretical model was confronted to our experimentally retrieved pulse duration data.

Qualitative behaviour

During their roundtrip around the cavity, the pulses undergo temporal shape variations. These changes can be characterised in terms of global delay dispersion (GDD). Normal GDD tends to broaden the pulse duration, while anomalous GDD reduces it.

During one round-trip around the cavity, the total GDD accumulated by the pulse is 0, to ensure the stability of the regime. Thus, the normal dispersion must be compensated by the anomalous dispersion. In our cavity, the normal dispersion is due to the reflections on the mirrors, propagation in the air, transmission through the prisms and the crystal, and is compensated by the prisms compressor and the non-linear effects inside the crystal (self-phase modulation especially). A quantitative study on the GDD induced by each element this particular cavity was carried out in Clément Pivard's PhD thesis [15].

The value of the anomalous dispersion induced by the prisms compressor was detailed in subsection 3.1.2.

Semi-theoretical description

The deformation of the pulse during its roundtrip in the cavity can be quantified using the indicator developed by Brabec et. al. [230]. This indicator quantifies the difference from the perfectly solitonic regime, where the pulse propagates without any deformation, which corresponds to the case $r = 0$. On the contrary, the greater the value of r , the more deformed the pulse during its round-trip. The formula for this indicator is:

$$r = \frac{1}{2\pi} \frac{(\Phi W)^2}{|D|} \quad (3.14)$$

with D the net round-trip linear GDD (namely the round-trip GDD without the part due to non-linear effects in the crystal), $W = \int P(t)dt$ the pulse energy, $P(t)$ the instantaneous pulse power, $\Phi P(t)$ the round-trip nonlinear phase shift due to Kerr effect in the crystal. This expression can be interpreted as the ratio between the phase due to non-linear effects in the crystal and the phase due to the rest of the cavity. In the case where they perfectly cancel out, the pulse duration is stable (soliton); otherwise, non-linear effects induce non-negligible pulse duration change (solitary wave).

In our case, this indicator is $r \simeq 7$. This corresponds to a solitary wave regime, where the pulses are deformed along their roundtrip around the cavity.

More quantitatively, in the case where the pulse is deformed, the same team gave an empirical formula for the duration of the intracavity pulse τ depending on the point of the cavity [239]:

$$\tau = \frac{3.53|D|}{\Phi W} + \alpha \Phi W \quad (3.15)$$

with D the net round-trip GDD, $W = \int P(t)dt$ the pulse energy, $P(t)$ the instantaneous pulse power, $\Phi P(t)$ the round-trip nonlinear phase shift due to Kerr effect in the crystal, and α a numerical factor depending on the point in the round-trip.

Hence, depending on the point of the roundtrip where the pulse meets the output coupler, the dispersion measured outside the cavity will change. This allows to explain the pulse duration disparity between direction 1 and 2, that are not taken from the cavity at the same point in their roundtrip.

Quantitative measurements

To describe more quantitatively the dispersion of the pulses, measurements using negative-GDD mirrors were carried out.

The duration of the pulses within the cavity is given, on the one hand, by the aforementioned formula 3.15. On the other hand, under the assumption that the pulses have a gaussian shape, their duration is also given by:

$$\tau = \tau_0 \sqrt{1 + \left(\frac{\text{GDD}}{2\tau_0^2} \right)^2}. \quad (3.16)$$

with GDD the group delay dispersion and τ_0 the dispersion-free pulse duration. These values were retrieved for both pulses.

To do so, the strategy was to add supplementary negative GDD to the pulses using dispersive mirrors (Layertech n°142012, inducing $\Delta\text{GDD} = -150\text{fs}^2$ per reflection), while measuring their duration. A numerical fit using the formula 3.16 would then give τ_0 and the value of GDD before our GDD addition. The experimental setup is schematised in figure 3.15.

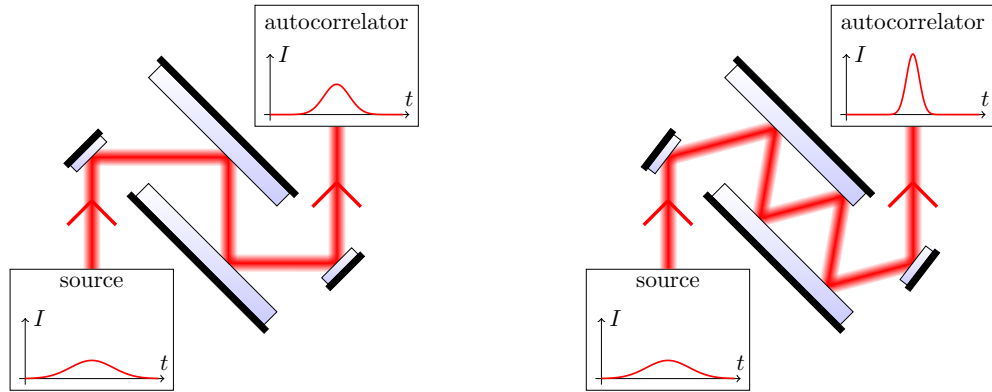


Figure 3.15: Schema of the experimental setup for the GDD retrieval protocol. The pulse is reflected n times on chirped mirrors (left: $n = 2$, right: $n = 4$), which adds a corresponding negative GDD of $n\Delta\text{GDD}$ and thus shortens the pulse duration. The pulse duration is then retrieved thanks to an autocorrelator. This allows to retrieve the duration of the pulse as a function of the number of reflections.

The results are presented in figure 3.16.

The retrieved values allow to say that the pulses are not Fourier-limited, but still in a low-dispersion regime ($\text{GDD}/\tau_0^2 \sim 10^{-1}$). This is consistent with what could have been expected from a Kerr-lens mode-locked Ti:Sa cavity [240].

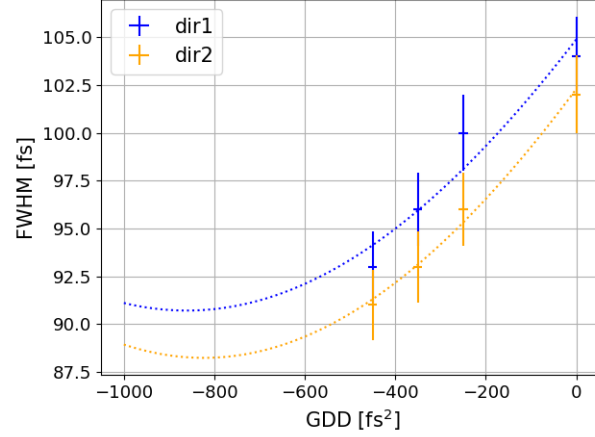


Figure 3.16: Retrieved FWHM of the pulses from the autocorrelation measurements as a function of the added negative GDD. The dotted lines correspond to the fit with the temporal envelope of a free-propagating gaussian beam as a function of its GDD (cf equation 3.16). Retrieved parameters: for dir1 $\tau_0 = (90.7 \pm 2.9)$ fs, $\text{GDD} = (861 \pm 97)$ fs²; for dir2 $\tau_0 = (88.2 \pm 0.9)$ fs, $\text{GDD} = (823 \pm 31)$ fs². These results were obtained with a different KLM regime than the previous graphs, with $\Delta f_{\text{rep}} = 0$.

3.4.3 Spectral characterisation

The power spectra of both directions were measured with grating-based spectrometers with resolution ~ 1 nm, using mirrors losses outside the cavity. Numerical fit on this data allowed to retrieve the central wavelengths and spectral widths, as presented in figure 3.17.

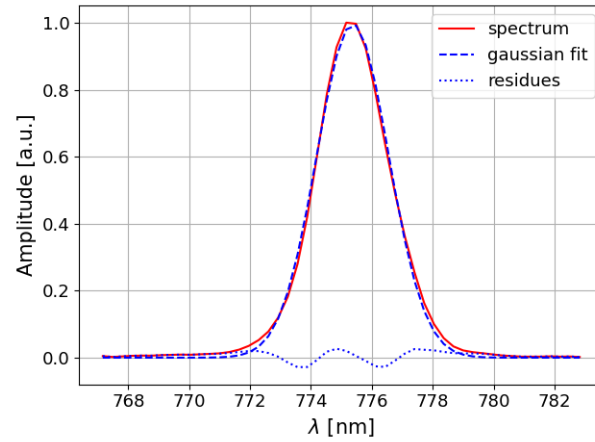


Figure 3.17: Power spectrum from a grating-based spectrometer, gaussian fit and residues. Retrieved parameters for the fit: central wavelength = (775.37 ± 0.02) nm, $\text{FWHM} = (2.7 \pm 0.3)$ nm

As can be seen on this graph, the gaussian shape fits qualitatively well, allowing to retrieve the central wavelength and spectral width of the spectra. The uncertainty given by the fit algorithm is much smaller than the spectral resolution of the spectrometers (~ 1 nm), which can be taken as an upper bound for the actual uncertainty.

The spectra's central wavelengths range from 760 nm to 810 nm, depending on the alignment of the elements of the cavity, which corresponds to the gain of the Ti:Sa crystal [240].

Dependence with experimental parameters

The spectral width (FWHM) of the spectra range from 2 to 10 nm. The experimental parameters that allow to make it vary are the same as for Δf_{rep} : cavity mirrors alignment, prisms insertion, pump power. Since the dependence of Δf_{rep} in these experimental parameters was studied in section 3.3, only the correlation between the spectral width and Δf_{rep} is presented here.

To investigate the correlation between the spectral width and Δf_{rep} , both these parameters were measured, as prisms insertion and pump power were changed. The result is presented in figure 3.18.

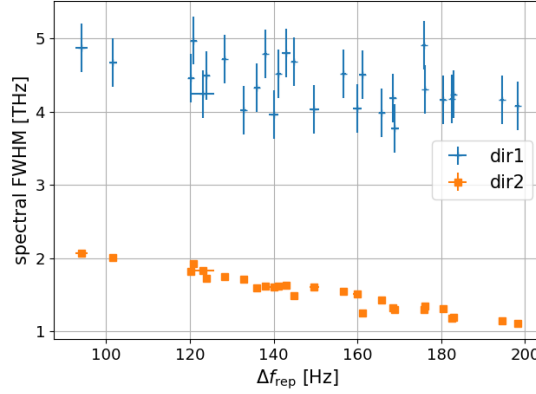


Figure 3.18: Variation of the spectral width of both power spectra as functions of Δf_{rep} value. The parameters that were changed were the pump power (from 4.76 W to 4.84 W) and the prisms compressor position (over the course of 0.6 mm). The measurement uncertainties are much larger for direction 1 because the mirror loss that was used had a lower power, resulting in a lower signal/noise ratio on the spectra.

A decreasing tendency with Δf_{rep} can be observed for direction 2, while the measurement uncertainties do not allow to distinguish a variation for direction 1. The relative variation of direction 2 (factor 2, from 2 to 1 THz) is much larger than direction 1. Like for pulse duration measurements (figure 3.14), this can be interpreted in terms of GDD.

Distance to the Fourier limit

These spectral width measures, combined with the independant pulse duration measures, allow to quantify how far from the Fourier limit are the pulses.

The Fourier limit, in the case of gaussian pulses, can be expressed in terms of power spectral width $\Delta\nu$ (FWHM) and pulse duration Δt (intensity FWHM) [233] by:

$$\Delta t \geq \Delta t_{\text{Fourier-limited}} \text{ with } \Delta t_{\text{Fourier-limited}} = \frac{2 \ln(2)}{\pi} \frac{1}{\Delta\nu}. \quad (3.17)$$

Hence, by comparing the pulse duration measurements with the Fourier-limited durations obtained thanks to the spectral width measurements, the distance from the Fourier limit can be quantified. This comparison is given in figure 3.19.

The pulses are found to be relatively close to the Fourier limit: $\Delta t \leq 1.6 \Delta t_{\text{Fourier-limited}}$ for direction 1 and $\Delta t \leq 1.4 \Delta t_{\text{Fourier-limited}}$ for direction 2. This is consistent with the low-dispersion regime result observed in subsection 3.4.2.

Upper bound for Δf_{rep}

The spectral measurements allowed to show a qualitative behaviour of the spectra, that is illustrated in figure 3.20 and that can be used to predict an upper bound for Δf_{rep} .

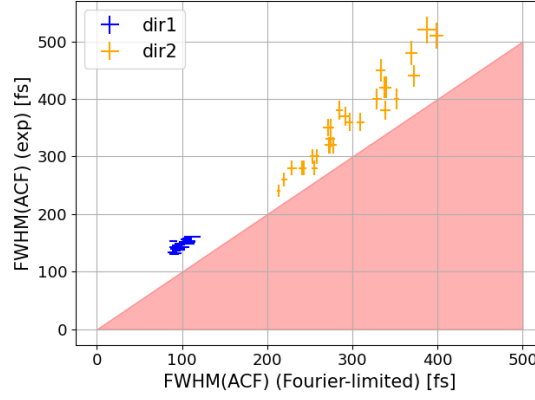


Figure 3.19: Pulse duration retrieved from autocorrelation measurements as a function of Fourier-limited pulse durations, retrieved from spectral width measurements using grating-based spectrometers using the formula 3.17. The zone forbidden by the Fourier limit is colored in red.

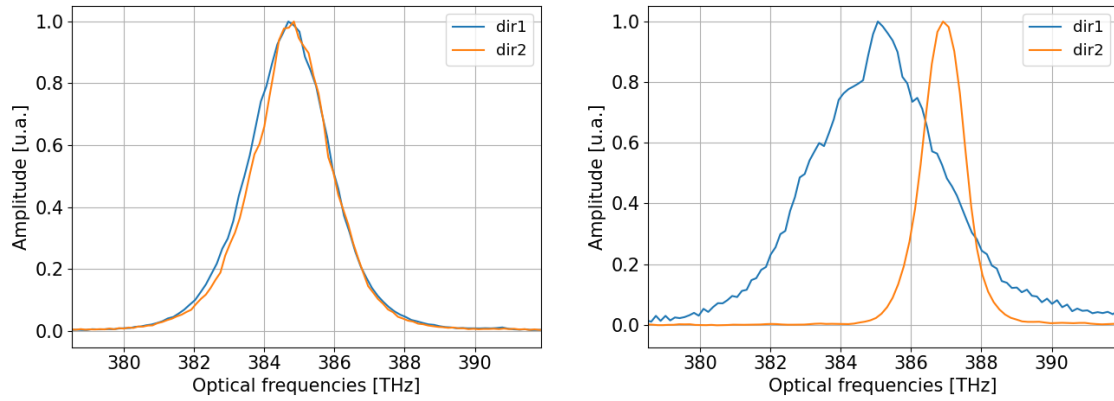


Figure 3.20: Normalised spectra in the cases of $\Delta f_{\text{rep}} = 0$ (left) and high Δf_{rep} (168 Hz) (right)

Qualitatively, the spectrum of direction 2 tend to drift in central wavelength and get thinner as Δf_{rep} increases, while the spectrum of direction 1 remains more stable. This gives an upper bound for Δf_{rep} values exploitable in DCS. Indeed, as seen in subsection 2.1.6, the amplitude of the DCS spectrum is proportional to the product of the spectra of both directions, so if the spectra drift so much apart that they don't overlap anymore, no signal will be observed. This point was not reached experimentally, but an upper bound of several hundreds of Hz can be extrapolated for our cavity from our measurements, at least in the known regimes. Up to now, the highest observed Δf_{rep} was 425 Hz [241].

3.5 Self-steepening effect

To characterise the cavity and its Δf_{rep} parameter, tries were made to understand the origin of Δf_{rep} at the microscopic scale.

In bidirectional cavities, the Δf_{rep} can originate in several phenomena: Sagnac effect [242, 243], polarisation multiplexing [194, 195, 244], wavelength multiplexing [200, 201, 245, 246], asymmetric cavity configuration [197, 199, 203, 204, 247, 248]. In the case of free running bidirectional ring cavities, the microscopic origin of Δf_{rep} is still widely unknown.

In 2016, several hypotheses were proposed by Ideguchi et. al. for the origin of Δf_{rep} in bidirectional ring cavities [204], one of them being the self-steepening effect.

3.5.1 Theoretical description

The self-steepening effect is a non-linear effect (of order 3 in the electric field), that tends to increase the slope of the pulses, as illustrated in figure 3.21 [249–251], and that modifies the group velocity of the pulse depending on its peak intensity.

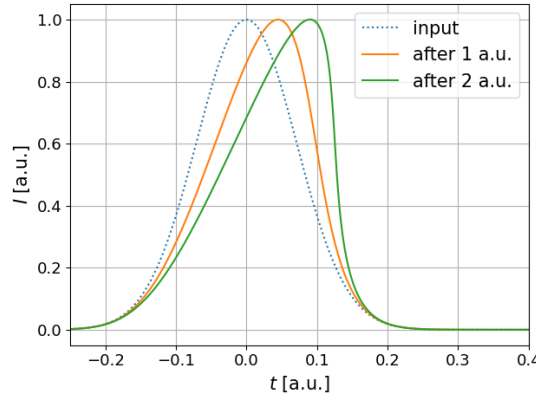


Figure 3.21: Deformation of a gaussian pulse by self-steepening effect. t is the time in the comoving frame. blue: temporal input; orange: after a 1 arbitrary unit propagation; green: after a 2 arbitrary units propagation.

Formally, it appears in the non-linear Schrödinger equation, written in the comoving frame of the pulse under the assumption of slowly-varying envelope:

$$\underbrace{\partial_z \mathcal{A}}_{\text{evolution}} = \underbrace{\frac{i}{2} k'' \partial_t^2 \mathcal{A} + \frac{1}{6} k''' \partial_t^3 \mathcal{A}}_{\text{dispersion terms}} - i\gamma \underbrace{\left(\overbrace{|\mathcal{A}|^2 \mathcal{A}}^{\text{Kerr}} - \overbrace{\frac{i}{\omega_0} \partial_t (|\mathcal{A}|^2 \mathcal{A})}^{\text{self-steepening}} - \overbrace{T_R \mathcal{A} \partial_t (|\mathcal{A}|^2)}^{\text{Raman}} \right)}_{\text{non-linear response of medium}} \quad (3.18)$$

with z and t the direction of propagation and time in the comoving frame, $\mathcal{A}(z, t)$ the envelope of the electrical field, k'' and k''' the second and third derivative of the wave vector with respect with the pulsation ω around the carrier pulsation ω_0 , γ a third-order non-linear coefficient of the material and T_R its Raman scattering coefficient.

The group velocity difference inside the crystal between the two directions due to the self-steepening effect causes a round-trip time difference in the cavity between the two directions, and thus a repetition rate difference.

3.5.2 Influence on the repetition rate

In a simulation paper from 2010, Sander et. al. [229] predicted quantitatively a dependency of the round-trip duration inside a cavity with respect with the pulse energy due to the self-steepening effect.

To verify this prediction experimentally, Δf_{rep} was retrieved for different bidirectional regimes with different pulse energy differences between the two directions.

Δf_{rep} was retrieved from the temporal interferogram trace, as described in subsection 3.3.1. Intracavity pulse energy was retrieved from extracavity average pulse power measurements: the intracavity pulse energy W begin linked with the extracavity pulse power P_{avg} , the transmission of the output coupler T and the repetition rate f_{rep} through the equation:

$$W = \frac{1}{T} \frac{P_{\text{avg}}}{f_{\text{rep}}}. \quad (3.19)$$

To make the extracavity average pulse power vary, the pump power, prisms configuration and cavity mirrors alignments were changed. The result is presented in figure 3.22.

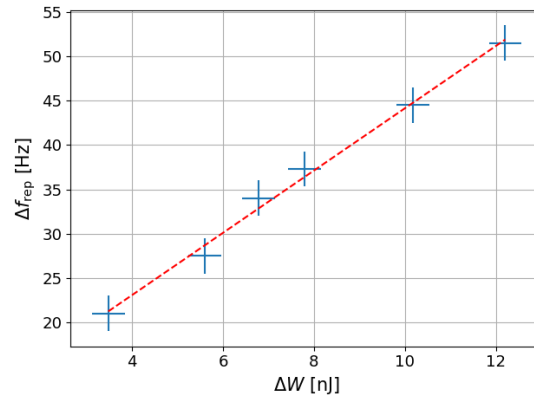


Figure 3.22: Δf_{rep} as a function of the intracavity pulse energy difference (blue) and linear fit (red). The retrieved slope corresponds to a variation of roundtrip time of (0.25 ± 0.03) fs/nJ.

Reproductions of these measurements in other KLM regimes (with higher Δf_{rep}) have given bigger slope coefficients, as presented in figure 3.23, but they remain in the same order of magnitude.

Our result is consistent with the (0.1 ± 0.2) fs/nJ slope prediction of Sander et. al.. This can be interpreted in favor of the self-steepening effect being the source of Δf_{rep} in our cavity.

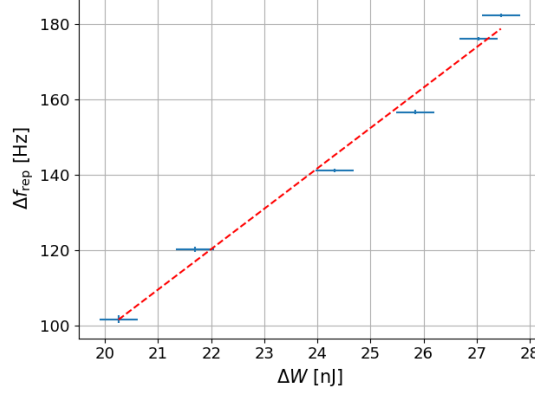


Figure 3.23: Δf_{rep} as a function of the intracavity pulse energy difference (blue) and linear fit (red) in a high- Δf_{rep} regime. The retrieved slope corresponds to a variation of roundtrip time of (6.3 ± 0.4) fs/nJ.

3.6 Chromatic dispersion

An other hypothesis to explain the origin of Δf_{rep} is the chromatic dispersion within the Ti:Sa crystal.

Since the spectra of the two directions do not have the same central wavelength, the non-linear variation of refractive index with respect with the wavelength within the crystal causes a difference of group velocity in the crystal Δv_g between the two directions, thus a difference of duration for crossing the crystal, thus a difference in repetition rate.

3.6.1 Quantitative formulation

To quantify this theoretical difference, Sellmeier coefficients were used [234] to compute the group velocity at the central wavelength of the two spectra (which were retrieved numerically from grating-based spectrometers measurements as described in subsection 3.4.3).

With the hypotheses of a small group velocity difference compared with the absolute group velocity ($\Delta v_g \ll v_g$) and a constant group velocity across the crystal (which is not true anymore if the non-linear effects are taken into account), the repetition rate difference is linked with this group velocity difference by:

$$\Delta f_{\text{rep}} = f_{\text{rep}}^2 \Delta v_g l_{\text{crystal}} / v_g^2 \quad (3.20)$$

To derive $v_g(\lambda)$, starting from Sellmeier formula:

$$n(\lambda) = \sqrt{1 + f(\lambda)} \quad \text{with} \quad f(\lambda) = \sum_{i=1}^3 \frac{a_i \lambda^2}{\lambda^2 - b_i^2} \quad (3.21)$$

which is derived with respect with λ (the derivative is noted $\xi'(\lambda) = \frac{d\xi}{d\lambda}(\lambda)$):

$$n'(\lambda) = \frac{f'(\lambda)}{2n(\lambda)} \quad \text{with} \quad f'(\lambda) = - \sum_{i=1}^3 \frac{2a_i b_i^2 \lambda}{(\lambda^2 - b_i^2)^2} \quad (3.22)$$

to be able to compute the group velocity:

$$v_g = \frac{c}{n - \lambda_0 n'(\lambda_0)}. \quad (3.23)$$

3.6.2 Results

In our case, f_{rep} is 118 MHz, the length of the crystal is 1 cm, and the coefficients of Ti:Sa (for λ expressed in μm) are [234]:

$$\begin{cases} a_1 = 1.041 & b_1^2 = 0.0004 \\ a_2 = 1.03 & b_2^2 = 0.0141 \\ a_3 = 3.55 & b_3^2 = 123.8 \end{cases} \quad (3.24)$$

The retrieved theoretical Δf_{rep} values, compared with the measured Δf_{rep} values for a single KLM regime, are presented on figure 3.24

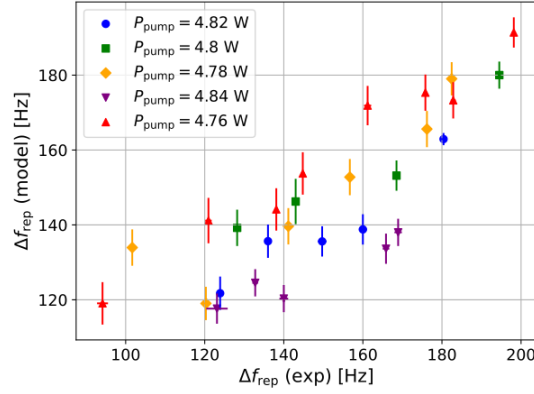


Figure 3.24: Theoretical Δf_{rep} retrieved from the dispersion model vs measured Δf_{rep} . The parameters that were changed to make Δf_{rep} vary experimentally were the pump power and the position of the prisms.

The two set of values are in good agreement, which tends to confirm the chromatic dispersion as a source of Δf_{rep} .

However, this very good agreement with the experimental data does not generalise to every regime. For instance, another comparison with another set of measures using a lower- Δf_{rep} regime is presented in figure 3.25.

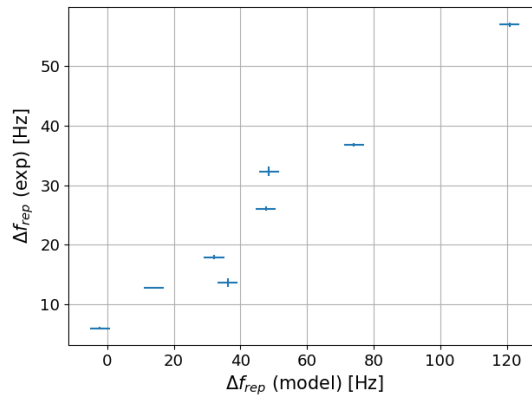


Figure 3.25: Δf_{rep} retrieved from the dispersion model vs measured Δf_{rep} in a lower Δf_{rep} regime.

In this case, although a correlation is still clearly visible, the absolute values differ by a factor

2 approximately. This shows that this dispersive model fails to predict Δf_{rep} accurately in every regime.

3.6.3 Limits of the model

This model only takes into account the dispersion within the crystal. It neglects the dispersion in the air and in the prisms.

In the air, similar calculations using Sellmeier coefficients allow to quantify a predicted repetition rate difference with the same method.

The travel length in the air during a roundtrip is approximately 3 m, which is two orders of magnitude greater than the travel length in the crystal; however, the group velocity difference is four orders of magnitude lower, giving a theoretical repetition rate difference two orders of magnitude lower, thus negligible at our level of precision.

In the prisms, the predicted repetition rate difference is difficult to quantify precisely, because the length of the path of the beams within the prisms (of the order of magnitude of cm) is difficult to measure accurately. However, an order of magnitude can be calculated, using the same method as for the crystal and the air.

The Sellmeier coefficients of the SF₁₀ prisms are given by the same source as the crystal and the air [208]. The order of magnitude of travel length inside the prisms is given by hand measurements and is of the order of the centimeter. The computations give the same order of magnitude as for the prisms for the predicted value of Δf_{rep} .

Hence, taking into account the dispersion inside the prisms precisely may greatly improve the accuracy of the model.

Conclusion

Our homemade free-running bidirectional ring Ti:Sa cavity ensures an intrinsically high degree of mutual coherence between its two emission directions, enough to observe high-SNR interferograms without the need for any feedback-based stabilisation, adaptive detection or post-acquisition correction. To modify its emission, intracavity mirrors orientation, pump power variation, and insertion variation of prisms from a prisms compressor can be used.

A detection setup allows to measure several output quantities.

The cavity can emit from 760 to 800 nm central wavelength. The produced pulses, close to Fourier limit, range from 85 to 350 fs pulse duration, which corresponds to several THz wide spectra. This important spectral width will be needed to retrieve optical spectra from DCS measurements in the next chapter (chapter 4).

An important quantity for DCS is the repetition rate difference between the two directions Δf_{rep} . It is retrieved numerically from the detected temporal trace of the beating of the two directions (interferograms). It ranges from 0 to 400 Hz. Its fluctuations were sampled and will be used in DCS methodology in the next chapter (chapter 4).

Δf_{rep} is strongly correlated with other emission parameters of the cavity, namely pulse duration, spectral width, intracavity dispersion and pump power.

A depletion in the baseline of the experimental interferogram trace is also observed, due to the gain competition between the two directions when they meet inside the Ti:Sa crystal. It is all the more important as Δf_{rep} increases, which makes the DCS methodology more difficult.

The source of Δf_{rep} in bidirectional ring cavities is still a disputed subject. Several hypotheses were emitted, one being the self-steepening effect, a third-order non-linear effect happening in the crystal. A simulation work later gave a simulation-based numerical value of the expected effect of the self-steepening effect on f_{rep} . Our experimental results are in good agreement with this value,

which tends to support the hypothesis of the self-steepening effect being the source of Δf_{rep} in our cavity. Further investigation would need a theoretical or simulation work that is beyond the scope of this work.

Another hypothesis for the source of Δf_{rep} is chromatic dispersion within the Ti:Sa crystal. The two beams, having different spectra, have a different group velocity in the crystal, thus a different round-trip duration in the cavity, thus a different repetition rate. A modelisation using Sellmeier coefficients predicts Δf_{rep} values that are the same order of magnitude than the measured ones, but fail to be more accurate in some cases. Upgrading the model would need taking into account the dispersion within the prisms, which implies to measure accurately the length of the optical path inside them.

Chapter 4

Dual comb spectroscopy in the IR and UV ranges

Introduction

This chapter presents the DCS results that were obtained using our bidirectional cavity during this work. This work follows previous development of this DCS experiment in the research group [15]. The experimental protocol is described in section 4.1.

Section 4.2 presents IR-DCS results. O_2 spectroscopy results that were obtained before this work [241] were reproduced, and their signal/noise ratio (SNR) was improved.

Post-acquisition averaging on the retrieved optical spectra was used to improve their SNR. Phase noise correction algorithms were tried on our data, but did not work due to the phase noise varying too fast.

Section 4.3 presents proof-of-concept UV-DCS measurements that were carried out on cesium (Cs) atomic lines and NO_2 molecular lines. The UV combs were produced using second harmonic generation (SHG) from the IR combs in a BBO crystal. The produced output power is of the order of 30 mW, which is enough to perform UV-DCS on a km-long atmospheric path [180]. The experimental setup allows to perform both IR and UV DCS simultaneously.

Asymmetric, Fano-type lineshapes were observed on the retrieved absorption lines of the experimental spectra, in both the IR and UV ranges. This lineshape prevents accurate retrieval of the gas concentration. Comparison of this lineshape with the results of the DCS simulation described in 2.3 suggests that Δf_{rep} fluctuations may be the origin of the asymmetric lineshape.

4.1 Experimental protocol

4.1.1 Experimental setup

General presentation

Simultaneous IR and UV spectroscopy is realised with the setup presented in figure 4.1.

To maximise the contrast of the interferograms, the wavefronts of the beams are modified to make them similar on the photodetector using convex lenses, as detailed further in this paragraph. The IR beams are focused on a 1 mm long BBO crystal to produce the UV beams by second harmonic generation (SHG). The IR and UV beams are then separated using a dichroic mirror. The IR beams follow a ~ 7 m long optical path and are superimposed on a 50/50 beamsplitter before being

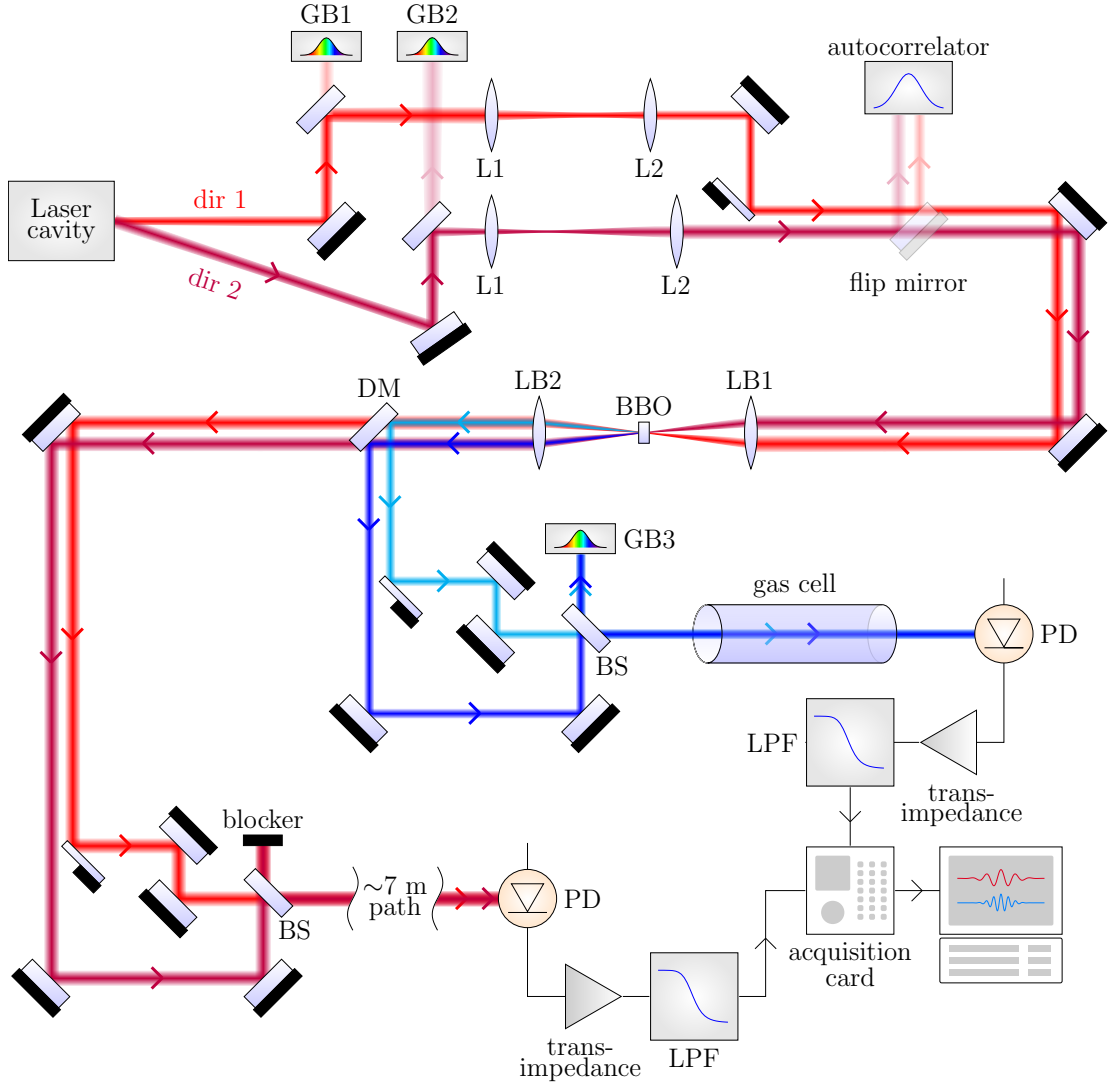


Figure 4.1: Schema of the complete detection setup for simultaneous IR and UV spectroscopy. Both beams (dir1 and dir2, for direction 1 and 2) are emitted from the output coupler of the laser cavity. Mirror losses are detected by grating-based spectrometers (GB1 and GB2) to measure the spectral power density of the beams. The wavefront of both beams are adapted with lenses, as detailed later in this paragraph. A flip mirror allows to deflect the beams to an autocorrelator to measure their pulse duration. Both beams are focused into a BBO crystal where second harmonic generation (SHG) takes place, creating two UV beams. The SHG setup is further detailed in subsection 4.3.1. The IR and UV beams are separated by a dichroic mirror (DM) that reflects the UV beams but is transparent to IR beams. The IR beams go through an approximately 7 m long optical path, before being superimposed on a 50/50 beamsplitter (BS) and focused on a photodiode (PD). The output signal of this photodiode is amplified by a transimpedance and filtered by a low-pass filter (LPF) before begin sampled by an acquisition card and treated on the local computer. The UV beams are superimposed on a 50/50 beamsplitter (BS). One of the output directions of the beamsplitter is detected by a grating-based spectrometer (GB3). The other output of the beamsplitter is sent through a gas cell before being focused on a photodiode (PD). The output signal of this photodiode is amplified by a photodiode and filtered by a low-pass filter (LPF) before being sampled by the same acquisition card as the IR signal and treated on the same computer. This setup is placed on an optical table.

focused on a photodiode (FDS010 from Thorlabs). The output signal of this photodiode is amplified with a transimpedance (DHPCA-100 from FEMTO), filtered by a homemade 50-MHz bandwidth 4th order low-pass filter and sampled by an acquisition card (ATS9440 from ACQUYSIS) to be treated on the local computer. As for the UV beams, they are superimposed on a beamsplitter and sent through a gas cell, before being focused on a photodiode. The photodiode, like the rest of the UV detection setup, uses similar devices as the IR detection setup. The output signal of the photodiode is amplified with a transimpedance and filtered by a low-pass filter, before begin sampled by the same acquisition card as the IR signal, to be treated on the same local computer.

The setup still allows to measure every quantity that was used to characterise the cavity emission (see table 3.1). Two supplementary quantities are detected: the UV interferogram temporal trace, using the same detection setup as the IR interferogram trace, and the UV beams power spectra, with resolution ~ 1 nm, using a third grating-based spectrometer (Avaspec-3648 from Optoprim).

Our autocorrelator does not allow to measure the UV pulses durations. This is due to the phase-matching condition that must be met in the autocorrelator crystal to perform duration measurements (see figure 3.12). The crystal can only strike the phase-matching condition between 700 nm and 1250 nm, which does not include UV.

Wavefront adaptation

To maximise the contrast of the interferograms, the wavefronts of both directions were adapted to be similar on the photodetector [228]. The wavefront adaptation protocol that was used relies on the same ABCD matrix model [235] and experimental measurements presented in section 3.2.

In the case without the SHG setup, the simulated beam size of both directions from the output of the cavity to the photodetector is presented on figure 4.2.

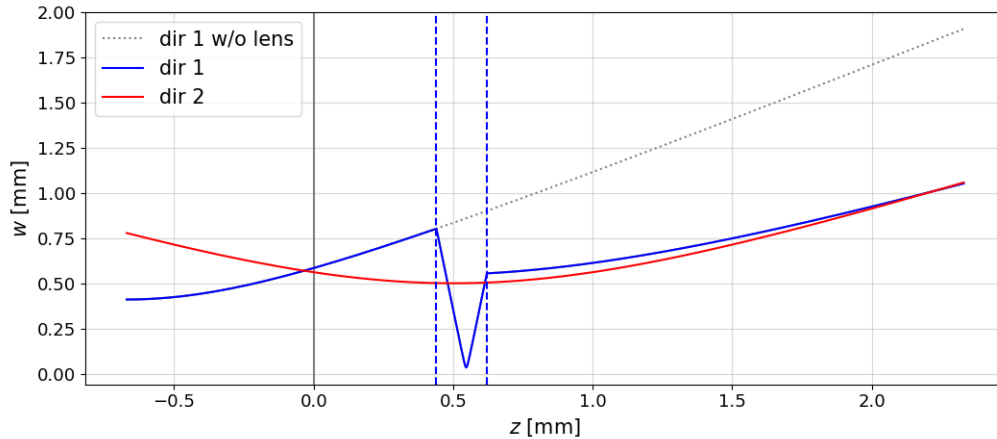


Figure 4.2: Beam size of both directions outside the cavity with respect with the position, computed with ABCD model from the retrieved propagation parameters. The output of the cavity is marked by the grey vertical line. Lenses positions are marked with colored dashed vertical lines. The beam size of direction 1 without the lenses is plotted in grey.

Two lenses, of respective focal lengths 100 mm and 75 mm, located respectively 44 cm and 62 cm after the output coupler, are used to superimpose the wavefront of direction 1 with the one of direction 2. The corresponding optical path is schematised in figure 3.3 at the beginning of chapter 3.

Using this setup, the interferograms contrast is up to 70%.

In the case with the SHG setup, two more lenses are added to the optical path to focus the beams inside the BBO crystal and recollimate them after the crystal. This modifies their wavefront. Thus,

two more lenses are added, to ensure their wavefront is still similar on the photodetector, using the same model as in the previous case. Moreover, the SHG process has an effect on the wavefront of the IR beams. This effect is found experimentally small, so it is neglected in the modelisation, but it will be discussed in section 4.3.

The modelised beam size evolution with respect with the propagation distance is presented in figure 4.3 and the positions and focus lengths of the lenses that were used are presented in table 4.1. The reasoning for choosing these particular values is given right after in this subsection.

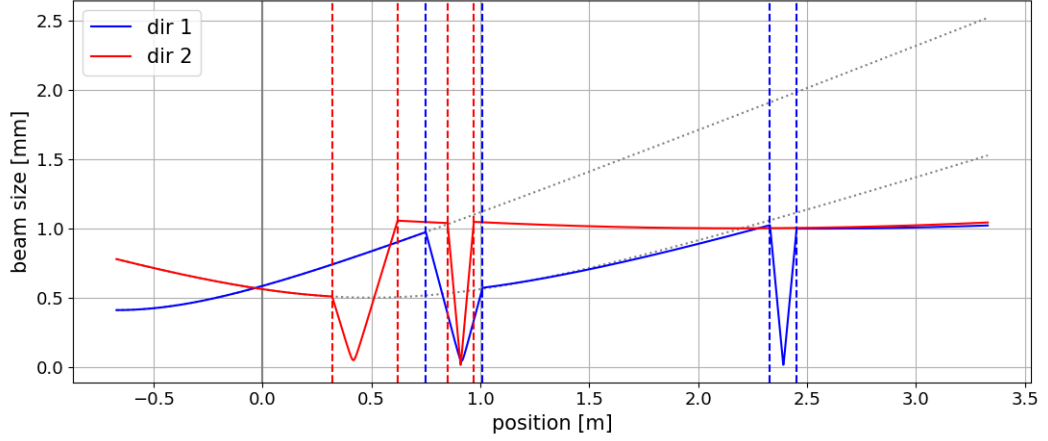


Figure 4.3: Simulated beam size as a function of the propagation length for both IR beams (dir1 and dir2). Simulated beam sizes are in blue (dir1) and red (dir2). Positions of the lenses are marked by dashed vertical lines. Simulated evolution of the beam size without the last two lenses (corresponding to the lenses around the BBO crystal) are given in dotted gray lines. The model is based on the ABCD matrix model [235] and the experimentally measured beam size data from section 3.2.

	lens number	position/OC	focal length
	#	[mm]	[mm]
dir1	L1	750	150
	L2	1010	100
	LB1	2330	60
	LB2	2450	60
dir2	L1	320	100
	L2	620	200
	LB1	850	60
	LB2	970	60

Table 4.1: Lenses positions with respect with the output coupler and focal lengths in our detection setup. Lenses number 3 and 4 of both direction correspond to the same physical pair of lenses, around the BBO crystal (see figure 4.1).

The positions and focus lengths of the lenses were determined in the following order:

- Lenses 1 and 2 of direction 1 served to adapt its wavefront for IR interferometry without the SHG setup. To ensure the reproducibility of the results that were obtained before the UV measurements, these lenses remained untouched.
- Lens 3 of direction 1 was then set up to optimise the UV output power.

To optimise the UV output power, a compromise must be found between the phase matching condition and the beam focus, as explained in section 2.4.3. The experimental parameters that

allow to fix this compromise are the focal length of the lens that focuses the beams inside the BBO crystal (see figure 4.1) and the beams sizes when they reach this lens [235]. To find the values of these experimental parameters that lead to the highest possible UV output power, a simulation was carried out, as will be detailed in subsection 4.3.1. The third lens position and focal length were then set up to match the results of this simulation.

- Lens 4 of direction 1 was then set up to recollimate the beam. The distance between lenses 3 and 4 is slightly higher than the sum of their focal lengths (see table 4.1), which allows to recollimate the beam better in this case.
- Lenses 3 and 4 of direction 2 correspond to lenses 3 and 4 of direction 1.

Directions 1 and 2 were initially meant to produce UV beams in separate crystals, to be able to optimise the SHG processes separately in each direction. However, it was found experimentally that higher IGM contrasts were reached when both directions were doubled inside the same crystal. Thus, the same BBO crystal served for both directions, as shown in figure 4.1. As a consequence, lenses 3 and 4 of direction 1 are also lenses 3 and 4 of direction 2, and their positions and focal lengths were already set up.

- Lenses 1 and 2 of direction 2 were then set up to optimise the interferograms contrast.

To optimise the contrast on the interferograms, the two directions' wavefronts must be as similar as possible on the photodetector, as explained in section 3.2. This holds for IR and UV interferograms. To ensure both directions' UV wavefronts are similar, the propagation parameters of the UV beams in the BBO crystal (waist and Rayleigh length) are set to be the same. To do so, the propagation parameters of the IR beams that produce the UV beams are set to be the same in the crystal. To do so, a telescope is set up in direction 2 to widen the beam size before it reaches lens 3, to make it as wide as the direction 1 beam when it reaches lens 3.

Experimentally, the lenses were placed with an uncertainty of the order of 1 mm. The uncertainty of their focal length is of the order of 1 % according to the builder (Thorlabs, EKSMA).

This setup allowed to acquire simultaneously UV and IR interferograms with contrasts up to 60 %, as illustrated on figure 4.4.

To measure the contrast of the interferograms, the interferogram temporal trace was measured using an amplified fast photodiode (PDA10A2 from Thorlabs) linked to a fast oscilloscope (10 GHz acquisition rate). This high acquisition rate allows to resolve each pulse, making it easier to measure the absolute value of the output signal of the photodetector, which is necessary to compute the value of the contrast.

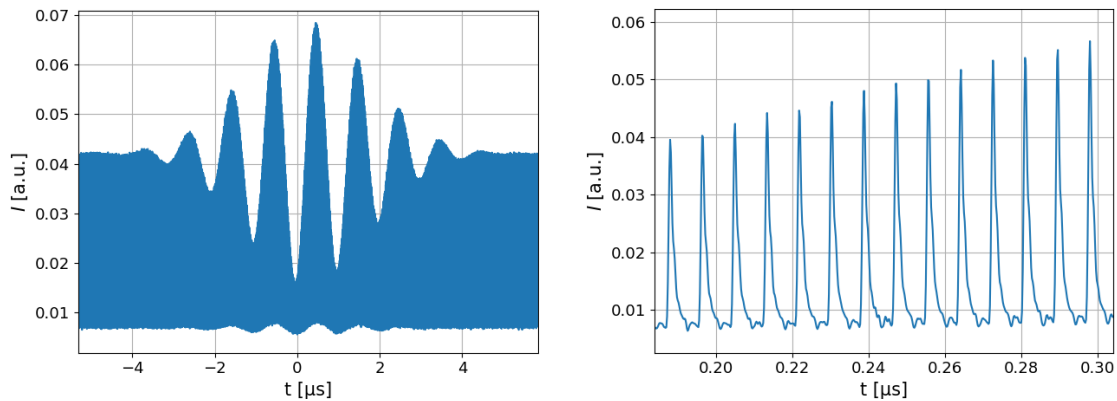


Figure 4.4: UV interferogram temporal trace, acquired with a fast commercial amplified photodiode linked to a fast oscilloscope. Centerburst (left) and close-up (right).

To further increase the contrast, the main limit is the phase matching condition in the BBO crystal. Experimentally, it is optimised by fine-tuning the position and angles of the BBO crystal mount. As illustrated on figure 4.1, both directions go through this crystal, so it is difficult to optimise the phase matching for both directions at the same time. Moreover, the SHG process also has an impact on the IR beams and their contrast, as will be detailed in subsection 4.3. The result of the phase-matching being different for each beam is a loss of mutual coherence, hence a loss of contrast on the interferograms.

4.1.2 Analysis

To retrieve spectroscopic information from the interferogram temporal trace, an analysis algorithm was developed. Its aim is to retrieve one transmission spectrum from each interferogram temporal trace.

Interferograms temporal windowing

As detailed in chapter 2.1, each interferogram allows to retrieve one optical spectrum. Thus, the first step of the algorithm is to locate the interferograms. To do so, the same steps as in the Δf_{rep} retrieval algorithm (subsection 3.3.1) are followed. A basic peak finding algorithm allows to retrieve the approximate temporal position of the interferograms, then a numerical fit allows to locate precisely the central position of the envelope. Finally, they are extracted, with a tunable length window, and their spectra are computed using an FFT function [252] on this window.

The length of this temporal window is critical regarding the retrieved spectroscopic data. The criterion to set the length of the window is that a tradeoff must be found between the signal/noise ratio (SNR) and the resolution of the retrieved optical spectra.

The longer the window, the better the spectral resolution on the rf spectra, but the lower the spectral SNR per spectral element, as the temporal SNR decreases far from the centerburst.

This process is illustrated in figure 4.5. In this case, it appears that a 6 ms window give a more noisy spectrum than a 1 ms window, while a 0.2 ms window does not allow to properly resolve the spectral lines that the spectrum presents, as can be inferred from the lower amplitude of the absorption line. Thus, the choice in this case would be a 1 ms window.

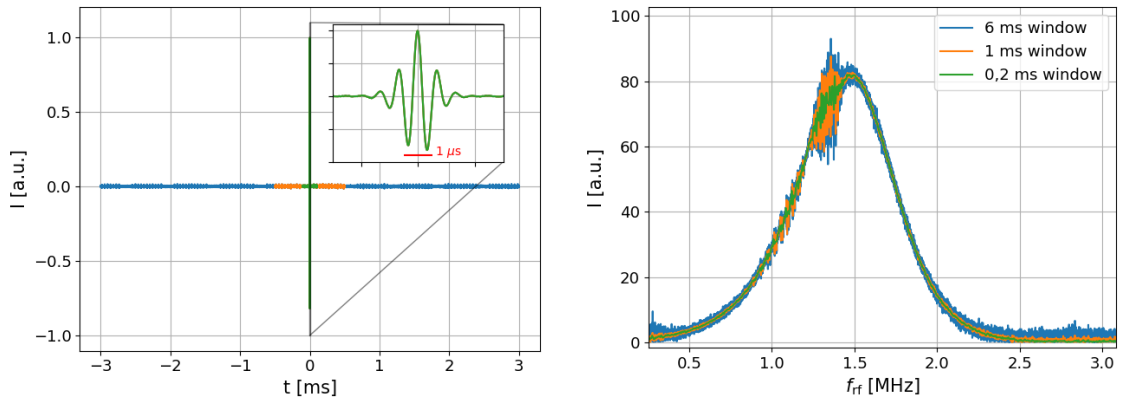


Figure 4.5: Temporal interferogram traces with different window lengths (left) and corresponding power spectra (right). The spectra on the right were obtained by FFT of the corresponding temporal window. Δf_{rep} average value was 13 Hz. The featured fast variations correspond to absorption lines of O_2 (A band).

The FFT window tradeoff between the resolution and the SNR depends on the value of Δf_{rep} . Indeed, the resolution of the FFT spectrum of one windowed interferogram (rf spectrum) scales

with the inverse of the FFT window duration. Besides, as seen in section 2.1 (equation 2.54), the rf spectrum corresponds to the optical spectrum compressed by a factor $1/a = \Delta f_{\text{rep}}/f_{\text{rep}}$, thus for a given optical resolution the corresponding rf resolution scales with Δf_{rep} . Hence, for a given window duration, the resolution that is retrieved on the optical spectrum scales with $1/\Delta f_{\text{rep}}$. Formally, with τ_{IGM} the window duration, f_{rf} the rf resolution and ν_{opt} the optical resolution:

$$f_{\text{rf}} \propto \frac{1}{\tau_{\text{IGM}}} \quad \text{and} \quad \nu_{\text{opt}} = \frac{f_{\text{rep}}}{\Delta f_{\text{rep}}} f_{\text{rf}} \quad \text{so} \quad \nu_{\text{opt}} \propto \frac{f_{\text{rep}}}{\Delta f_{\text{rep}}} \frac{1}{\tau_{\text{IGM}}} \quad (4.1)$$

Thus, experimentally, the length of the FFT temporal window is tuned for each set of interferograms, as a function of its Δf_{rep} .

For the sake of clarity, this latter discussion does not distinguish between spectral resolution and spectral increment. This is equivalent to do the hypothesis of a Fourier-limited situation. This hypothesis does not hold rigorously in our case, as the spectral resolution of our spectrometer is inferior to the linewidth of the detected absorption lines, as will be seen on next sections. However, the discussion remains relevant about the spectral envelope retrieval process, as the ones we detect are well-resolved by our spectrometer. It remains relevant as well in the case where the FFT window corresponds to an optical spectral increment superior to the spectral resolution of the spectrometer.

Complementary to this windowing, a Hann window was added to the UV interferogram to remove spurious digital noise [253].

Spectral baseline removal

To extract the transmission spectra from the retrieved DCS spectra, the envelope term is removed (see equation 2.54). This spectral envelope corresponds to the baseline of the spectra, on which the absorption lines are imprinted. Several methods were tried to extract this baseline: numerical fit with standard lineshapes, polynomial fit, and interpolation from the Fourier transform of a shorter window. As described hereafter, the latter turned out to be the most accurate.

Numerical fit with gaussian and sech^2 shapes were tried on our experimental DCS spectra to retrieve their baseline.

This choice of lineshapes was motivated by the fact that gaussian and sech^2 shapes fitted well the spectra of direction 1 and 2 (see subsection 3.4.3), that are directly linked with the DCS spectra (see equation 2.54).

The numerical fits were done with least square methods, using preexisting algorithms [254]. The results of these fits is presented in figure 4.6.

The residues of the fit of the experimental spectrum are of the order of 10^{-1} arbitrary units, which is one order of magnitude less than the amplitude of the spectrum. Hence, the gaussian model fits well the experimental spectrum. However, the residues of the fit are of the same order of magnitude as the optical depth of the absorption lines. Thus, the baseline fluctuates a lot, which prevents to identify the retrieved transmission as the real transmission, and thus prevents further spectral analysis.

To retrieve the baseline of the experimental spectra with more accuracy, fit with a polynomial function was considered.

This choice was motivated by the generality of polynomes as a fitting function. To avoid the absorption lines perturbing the fit, only points without absorption lines were used to build the baseline. The degree of the polynome was determined by a compromise. If the degree is too low, the retrieved baseline is oversimplified compared with the real baseline. If the degree is too high, the fit algorithm is too much influenced by the noise, which biases the retrieved baseline. The results of these fits is presented in figure 4.7.

The baseline is well fitted by the polynomial model. The residues are one order of magnitude lower than for the gaussian or sech^2 fit. However, a bias is still visible on the baseline on the points that present absorption lines.

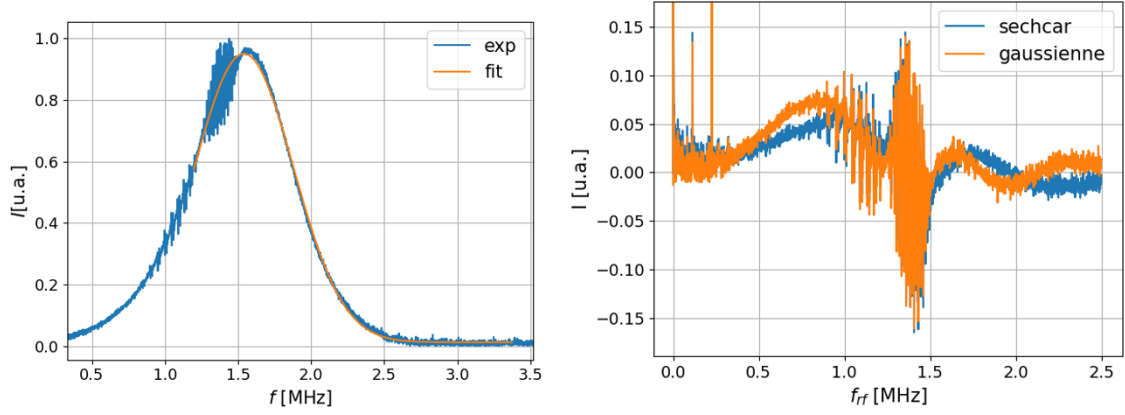


Figure 4.6: DCS spectrum obtained with the FFT of a 1-ms window around an interferogram and retrieved baseline using a gaussian fit (left) and corresponding transmission (right). Δf_{rep} average value was 13 Hz. The featured fast variations correspond to absorption lines of O_2 (A band).

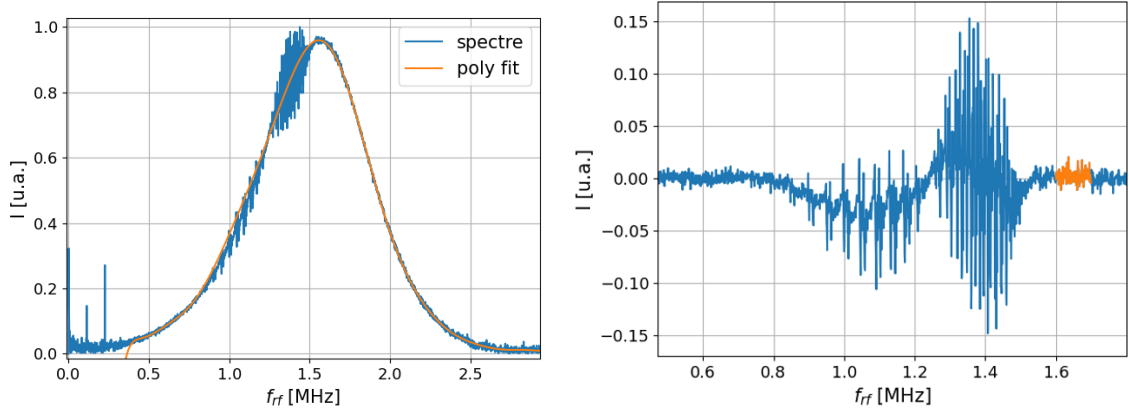


Figure 4.7: DCS spectrum obtained with the FFT of a 1-ms window around an interferogram and retrieved baseline using a degree 30 polynomial fit (left) and corresponding transmission (right). The orange points on the left graph are the points that were used to build the polynomial fit. Δf_{rep} average value was 13 Hz. The featured fast variations correspond to absorption lines of O_2 (A band).

To retrieve the baseline of the experimental spectra with more accuracy, a baseline retrieval protocol based on Fourier analysis was tried.

This choice was motivated by the fact that this model allows to adapt more to a general spectral shape than the gaussian or sech^2 model, while still taking into account every point of the spectrum, rather than restricting to the points without absorption lines like the polynomial model. The model is based on the fact that the free-induction decay is much longer in time than the interferogram, which is equivalent to say that the absorption lines are much smaller in spectral width than the DCS spectrum envelope. Thus, the Fourier transform of a temporal window whose size is of the order of the size of the centerburst will not feature the absorption lines, that correspond to the Fourier transform of the free induction decay. This corresponds to applying a low-pass filter to the spectrum, removing its fast components, namely the absorption lines.

Numerically, the linewidth of the retrieved spectral lines is of the order of 1 GHz, while the spectral envelope of the DCS spectrum is of the order of 1 THz: hence the small linewidth hypothesis is correct.

The baseline corresponds to the FFT of a small window including only the centerburst. The length of the window is redefined for each set of interferograms, depending on the centerburst duration. The baseline is interpolated from the FFT of the small window on a grid with the same spectral increment as the full spectrum to be able to divide it.

The result of this process is presented in figure 4.8.

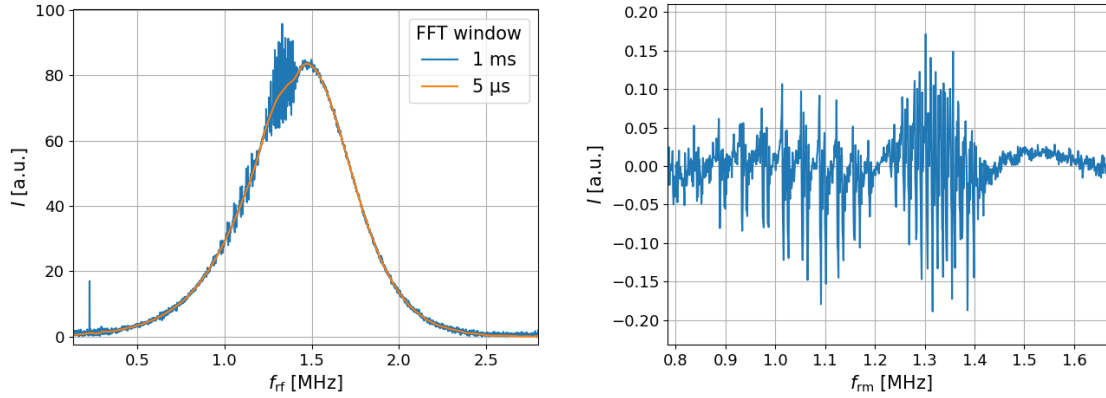


Figure 4.8: DCS spectrum obtained with the FFT of a 1-ms window around an interferogram and retrieved baseline using a FT-based fit (left) and corresponding transmission (right). The length of the window that served to obtain the spectral baseline was 5 μs . The baseline was interpolated from the points of the FFT of this small window. Δf_{rep} average value was 13 Hz. The featured fast variations correspond to absorption lines of O_2 (A band).

The retrieved baseline, though it still presents some erroneous tendencies, fits qualitatively and quantitatively better than the other models.

This latter method was thus kept to retrieve the baseline of the experimental spectra.

4.1.3 Averaging

To increase the signal/noise ratio of our transmission spectra, averaging processes were set up. In this subsection, the main difficulties that hindered the averaging are presented, and the implemented averaging processes are described.

Temporal description of phase noise

As seen in subsection 1.4.1, in our freerunning cavity, the emission is not stabilised. In particular, Δf_0 is non zero, which causes a carrier-envelope offset on the interferograms that shifts regularly from one interferogram to another (see equation 2.64). Moreover, Δf_0 and Δf_{rep} are free to fluctuate

in time. These fluctuations modify the interferograms, thus preventing straightforward averaging of the interferograms. Their effect is illustrated in figure 4.9.

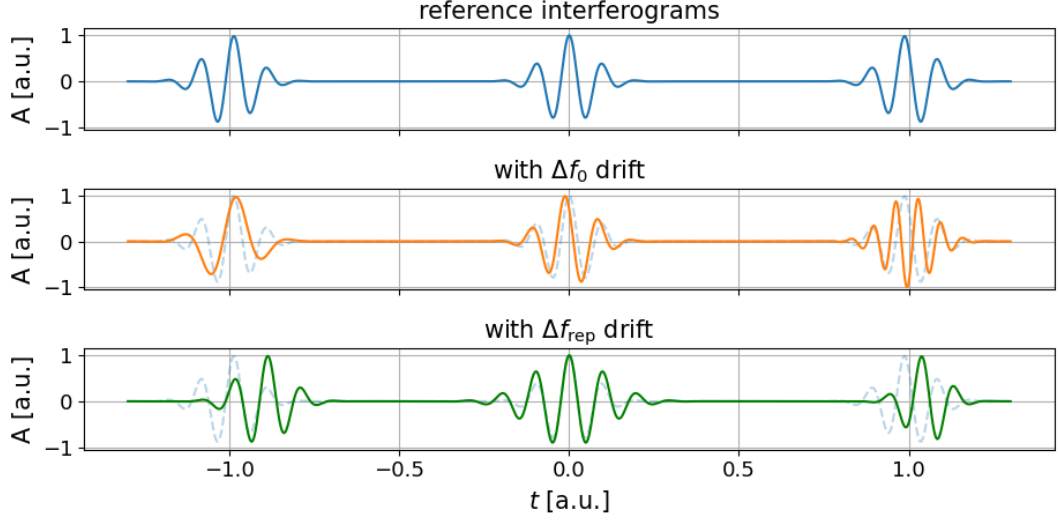


Figure 4.9: Simulated interferograms without Δf_0 and Δf_{rep} fluctuations (up), with only Δf_0 fluctuations (middle) and with only Δf_{rep} fluctuations (bottom).

Qualitatively, Δf_0 fluctuations modify the oscillation frequency of the interferograms temporal trace, while Δf_{rep} fluctuations modify the duration of the interferograms and shift them in time with respect with a perfect $1/\Delta f_{\text{rep}}$ period.

Quantitatively, the Δf_0 fluctuations $\delta\Delta f_0(t)$ and the Δf_{rep} fluctuations $\delta\Delta f_{\text{rep}}(t)$ appear in the expression of the temporal trace of the interferograms as [192] (the fluctuation terms are in blue):

$$S_{\text{noisy}}(t) = e^{2i\pi\Delta f_0 t} e^{2i\pi \int_0^t \delta f_0(t') dt'} \sum_p A_p e^{2i\pi p \Delta f_{\text{rep}} t} e^{2i\pi p \int_0^t \delta \Delta f_{\text{rep}}(t') dt'} \quad (4.2)$$

with p summing over the comb teeth, each tooth having an amplitude A_p .

Interferogram averaging

To be able to average the temporal interferograms straightforwardly, the fluctuations and the shifting phase/carrier offset (corresponding to the $e^{2i\pi\Delta f_0 t}$ term if equation 4.2) of the interferograms temporal trace must be experimentally corrected. To do so, a powerful averaging algorithm was developed by Hébert et. al. in 2017 [192]. Its main steps are briefly described here and its application to our data is discussed.

The algorithm aims at sampling $\delta\Delta f_0(t)$ and $\delta\Delta f_{\text{rep}}(t)$ at the rate of one point per interferogram, to then remove them numerically from the interferograms temporal trace.

To sample $\delta\Delta f_0(t)$ and $\delta\Delta f_{\text{rep}}(t)$ on an interferogram, it is first isolated using a peak finding algorithm. Then, it is compared with a reference interferogram, arbitrarily chosen as the first of the set. To compare two interferograms time traces, the algorithm uses ambiguity maps. This allows to retrieve the time shift between the centerbursts of the two interferograms and their relative phase, thus sampling $\Delta f_0 + \delta\Delta f_0(t)$ and $\Delta f_{\text{rep}} + \delta\Delta f_{\text{rep}}(t)$ on the centerbursts.

The time shift and relative phase between the two interferograms are retrieved as the argmax of the ambiguity function $\chi_{i,j}(\tau, f)$:

$$\chi_{i,j}(\tau, f) = \int_{-\infty}^{\infty} \text{CB}_i(t) \text{CB}_j^*(t + \tau) e^{2i\pi f t} dt \quad (4.3)$$

This function compares the time traces of two centerburst CB_i and CB_j , by computing the overlap of the two traces shifted in time and in spectral domain. This function reaches its maximum for τ corresponding to the time shift between the two centerbursts and f corresponding to the oscillating frequency difference between the two centerbursts. This allows to retrieve these quantities.

An ambiguity map of experimental interferograms is presented in figure 4.10.

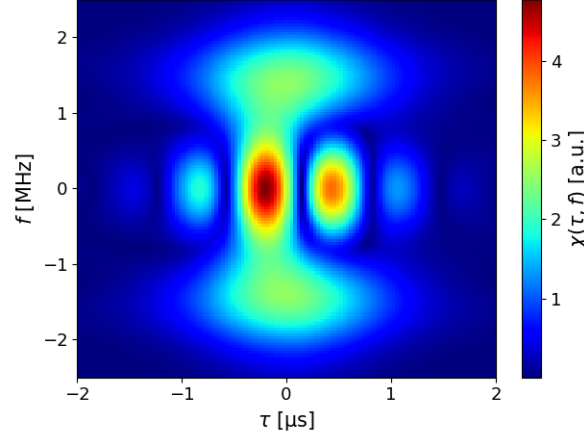


Figure 4.10: Ambiguity map of two experimental interferograms: ambiguity function χ as a function of the time shift τ (average value was subtracted for the sake of clarity) and spectral offset f . For this set of interferograms, $\Delta f_{\text{rep, average}} = 20$ Hz. The retrieved arguments of the max are $\tau = 0.2$ μs and $f = 0$ Hz

$\delta\Delta f_0(t)$ and $\delta\Delta f_{\text{rep}}(t)$ for every t are then extrapolated from these sampled points. Finally, a set of "corrected" interferograms is built, namely evenly-spaced and in phase interferograms. These can be averaged in time.

This method relies on a frequent enough sampling of $\delta\Delta f_0(t)$ and $\delta\Delta f_{\text{rep}}(t)$ that these fluctuations can be completely extrapolated from the sampled points. Since the algorithm samples one point per interferogram, this condition corresponds to a high enough Δf_{rep} value, compared with the fluctuations' amplitude. A quantitative criterion to distinguish if the fluctuations are sampled correctly is to check if the retrieved phase, that appears in the blue terms of equation 4.2, varies over more than π between two consecutive interferograms. If it does, it can be assumed that $\Delta f_0(t)$ and $\delta\Delta f_{\text{rep}}(t)$ are not sampled fast enough and that the algorithm cannot correct phase noise on this dataset.

In our case, the retrieved phase varies over more than π between two interferograms, as presented in figure 4.11.

Hence Δf_{rep} is not high enough to sample the fluctuations of our system efficiently and correct phase noise.

The algorithm has also been tried on interferograms temporal traces with higher Δf_{rep} (more than 150 Hz). In these cases, the treatment was prevented by the baseline depletion (see subsection 3.3.4). Indeed, the algorithm includes Hilbert transforms of the whole interferogram trace, which is much perturbed by the presence of this depletion.

To tackle the issues that prevent the algorithm from working on our interferograms temporal traces, a baseline correction algorithm could be set up. This investigation is among the perspectives of this work.

Spectral description of phase noise

To average the transmission spectra without relying on temporal interferograms averaging, the averaging was done directly on the transmission spectra.

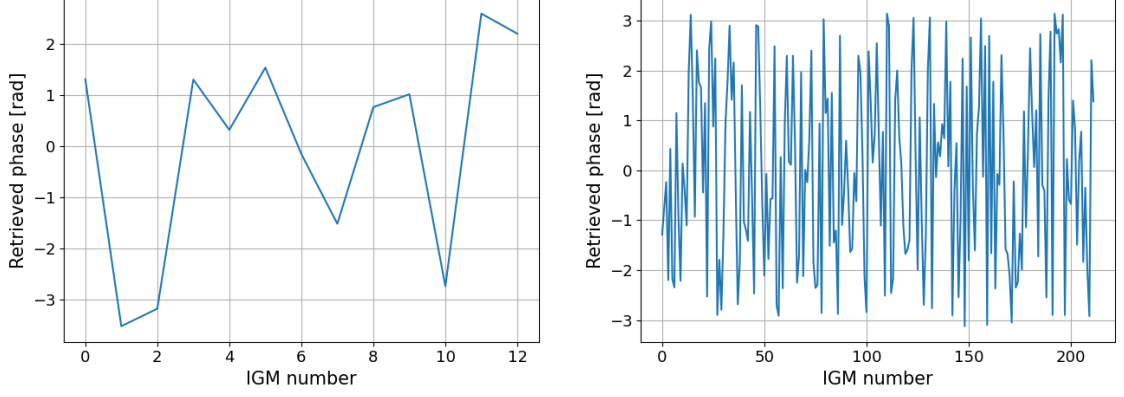


Figure 4.11: Retrieved $\int_0^t \delta f_0(t) dt$ for a experimental interferogram temporal traces in a low Δf_{rep} (~ 12 Hz) case (left) and a high Δf_{rep} (~ 106 Hz) case (right). Unwrapping algorithms were applied unsuccessfully.

To average the transmission spectra, both Δf_0 and Δf_{rep} fluctuations must be corrected. These two sources of fluctuations correspond to different deformations of the power spectra. Δf_0 fluctuations shift the whole spectrum, while Δf_{rep} shift teeth separately. These effects are illustrated on figure 4.12.

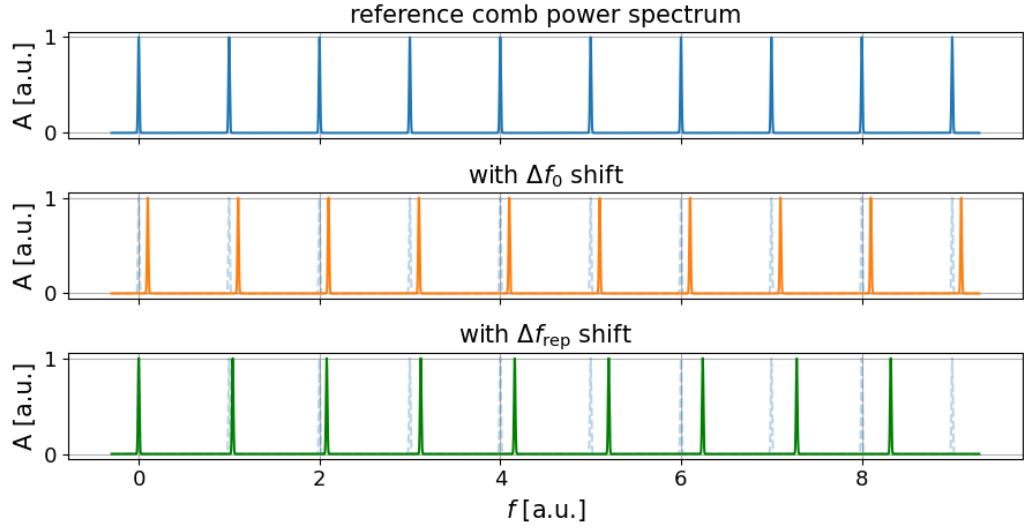


Figure 4.12: Simulated comb spectra without Δf_0 and Δf_{rep} fluctuations (up), with only a Δf_0 shift (middle) and with only a Δf_{rep} shift (bottom).

Thus, to average the spectra, both a global shift and a stretch of the spectra must be corrected.

Spectral averaging

To average our experimental spectra, both global shift and stretch with respect with a reference experimental spectrum must be retrieved for each spectrum.

The reference experimental spectrum is chosen arbitrarily. To retrieve the shift and stretch with respect with this spectrum, absorption lines are used as absolute references. Thus, experimentally, to be able to average a set of spectra with this method, they must feature at least two resolved

absorption lines. This constraint was easily met in the IR when working with the O₂ A band, that features many lines. This constraint is more challenging in the UV range, as we use cesium for spectral calibration, which features only two resolved absorption lines in a limited spectral range.

4.2 Infrared spectroscopy: O₂ spectroscopy

To quantify the spectral resolution of our spectrometer, O₂ detection experiment was carried out on a 7 m long optical path within the lab, as schematised on figure 4.1. A previous O₂ detection experiment had been carried out to obtain proof-of-concept results with this cavity [241]. This work aimed at refining these results in terms of signal/noise ratio. The choice of these absorption lines was motivated by the fact that they are intense (up to $6 \cdot 10^{-24}$ cm²/mol), correspond to a species that is abundant in the atmosphere ($5 \cdot 10^{18}$ molecules/cm³) and are within the gain bandwidth of Ti:Sa, around 760 nm. The absorption cross section is given in figure 4.13.

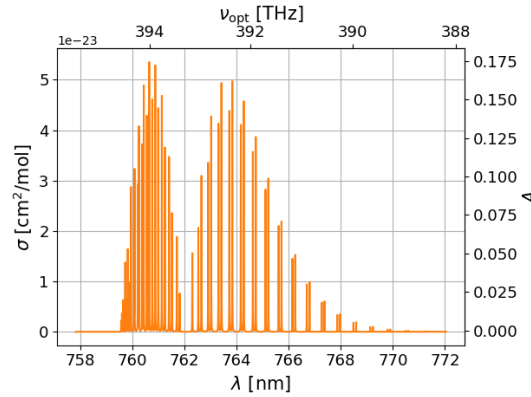


Figure 4.13: Absorption cross section of O₂ and corresponding absorbance A (given by an order 1 development of the Beer-Lambert law) for a 7 m path in room conditions of pressure and temperature, as a function of the wavelength and optical frequency. Data were retrieved from the HITRAN database [210].

4.2.1 Averaging

To increase the signal/noise ratio of the retrieved transmittance spectra, the averaging protocol described in subsection 4.1.3 was applied to our O₂ spectra. The main steps are shown here on a set of 10 interferograms with $\Delta f_{\text{rep}} \sim 20$ Hz.

First, the optimal FFT window duration was found to be 10 ms for a single interferogram acquisition. This windowing sets a maximum resolution of 200 Hz for the rf spectrum and 1 GHz for the optical spectrum. A one-shot spectrum is shown on figure 4.14.

The corresponding signal/noise ratio, defined as the ratio of the maximum amplitude of the retrieved absorption lines over the standard deviation of the noise of the baseline, is 11. The lineshape will be discussed later in subsection 4.2.2. The bias on the baseline level can be attributed to the baseline retrieval algorithm.

To use the averaging protocol, two absorption lines were chosen among the most intense ones to serve as absolute references. A comparison of the averaged 10 spectra and a one-shot spectrum is shown in figure 4.15, along with the lines retrieved from the HITRAN database [255].

The experimental absorption lines match with the absorption lines from HITRAN, since the latter were used to build the optical frequency axis. The amplitude of the lines match the theoretical result from Beer-Lambert law (see figure 4.13). The SNR of the experimental absorption lines is 34. This

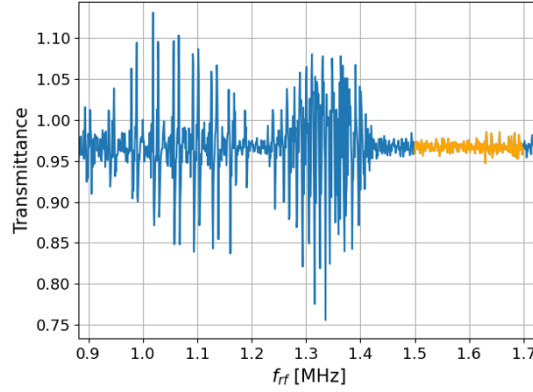


Figure 4.14: Experimental transmittance spectrum. The corresponding spectrum was obtained by FFT of a 10 ms window around an interferogram. The orange extract is the extract whose standard deviation was computed to calculate the signal/noise ratio. Δf_{rep} is 13 Hz. This windowing corresponds to a minimum optical resolution of 550 MHz.

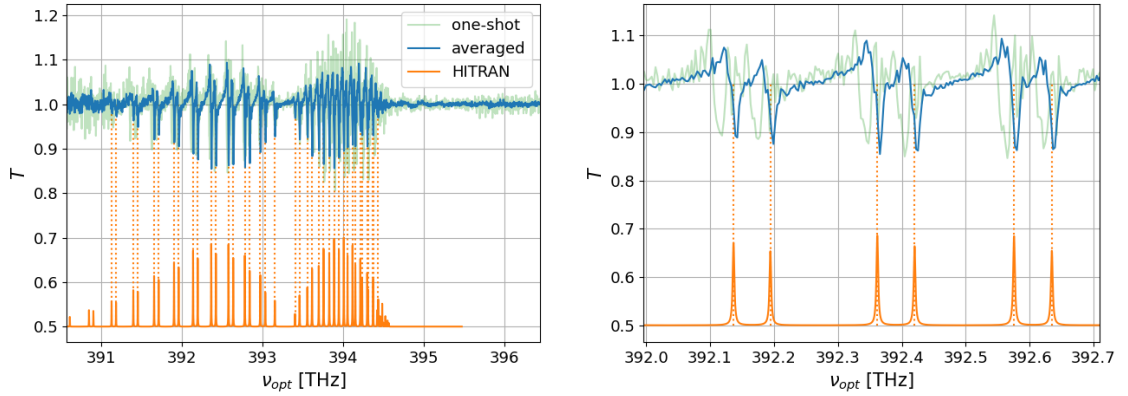


Figure 4.15: Transmission spectrum retrieved from the FFT of a 10 ms window around an interferogram (green), average of 10 spectra from this same acquisition (blue) and corresponding absorption lines from the HITRAN database (orange) (left) and close-up (right). Δf_{rep} is 13 Hz. This windowing corresponds to a minimum optical resolution of 550 MHz. The resolved O_2 lines are 1.5 GHz wide.

represents a factor 3 increase compared with the one-shot spectrum. This value was expected, since averaging on N acquisitions increases the SNR of a factor $\sqrt{N}$ and in our case $N = 10 \simeq 3^2$. This SNR also represents a factor 6 increase compared with the previously published O₂ detection results of the team, namely ~ 5 [241].

4.2.2 O₂ absorption lines lineshape analysis

The lineshape of the retrieved absorption lines does not match the expected lineshape. They are asymmetric and present a peak over the baseline. This lineshape is a Fano-type lineshape [256, 257].

This lineshape can be caused by the Fano effect, namely the non-linear coupling of discrete transitions with a continuum of transitions [256, 258]. This effect was first described quantitatively by Fano in 1961 and was observed in various domains, as reviewed in [257]. In particular, it was observed [163, 259] and treated [260, 261] in dual-comb systems.

A fit of an experimental absorption line with a Fano-type line is presented in figure 4.16.

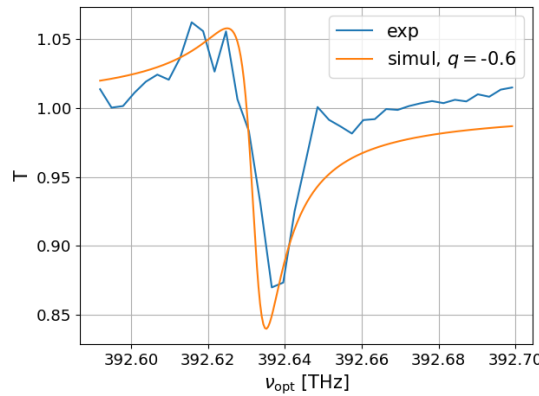


Figure 4.16: Transmission spectrum retrieved from the FFT of a 2 ms window around an interferogram, averaged over 10 spectra (same spectrum as in figure 4.16) (blue) and fit with a simulated Fano type line (orange). The parameters of the Fano line, as presented in [256], are $q = -0.6$, $\epsilon = (\nu - \nu_0)/\Delta\nu$ with $\nu_0 = 392.63$ THz, $\Delta\nu = 4.46$ GHz. These parameters are given as a coarse estimation, no optimisation was performed.

The SNR of the data does not allow accurate retrieval of the best fitting parameters. However, the experimental and simulated lines fit qualitatively well. This result does not allow to discriminate the nonlinear Fano effect as a source of the observed lineshape.

To further challenge this hypothesis, these spectroscopic results can be reproduced with a lower peak power. The pulses were chirped using chirped mirrors to lower their peak power, but the subsequent SNR decrease prevented significant observation. This investigation is among the perspectives of this work.

On the other hand, asymmetric DCS lineshape can be caused by Δf_{rep} fluctuations, as described in subsection 2.3.2. To test this hypothesis, our experimental data are compared with our simulation results.

A coarse fit of an experimental absorption line with a simulated line deformed by Δf_{rep} fluctuations is presented in figure 4.17.

The SNR of the experimental data and the low resolution of the simulated data do not allow precise retrieval of the best fitting simulated parameters. However, experimental and simulated data fit qualitatively well. This result does not allow to discriminate Δf_{rep} fluctuations as a source of the observed lineshape.

This hypothesis is difficult to discuss because it concerns fluctuations of Δf_{rep} at the time scale of one interferogram, while our experimental and algorithmic setup allows to sample these fluctuations

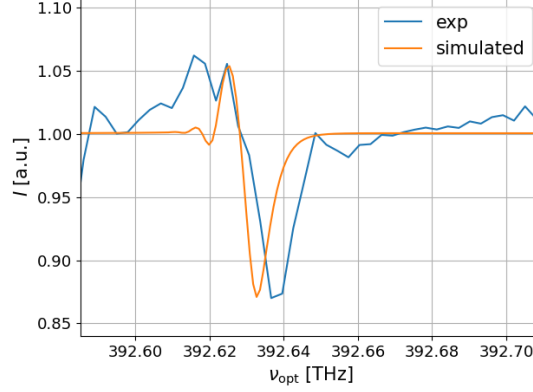


Figure 4.17: Transmission spectrum retrieved from the FFT of a 2 ms window around an interferogram, averaged over 10 spectra (same spectrum as in figure 4.16) (blue) and fit with a simulated line with fluctuating Δf_{rep} (orange). The average value of the simulated Δf_{rep} is 40 Hz. The simulated fluctuations of Δf_{rep} are linear in time, with a variation corresponding to a $\sim 10^{-5}$ Hz shift during the centerburst (variation rate of 66.7 $\mu\text{Hz}/\text{ps}$).

at a much slower rate of once per interferogram. Hence, to further challenge this hypothesis, a more temporally-resolved sampling of Δf_{rep} fluctuations is needed.

4.3 Ultraviolet spectroscopy: Cs and NO₂

4.3.1 Second harmonic generation setup

To produce UV frequency combs, second harmonic generation (SHG) is used on the IR combs produced by the cavity. The aim of the SHG setup is to produce enough UV output power to perform UV-DCS in the atmosphere over a km-long path. Theoretical calculations were carried out before this work to compute the corresponding average output power threshold, which is ~ 10 mW for our setup and frequency range [180].

The SHG setup is schematised in figure 4.1. The beams of both output directions from the cavity are focused by a convex lens inside a BBO crystal. The beams are then recollimated by another convex lens.

A BBO crystal was chosen because of BBO's capacity to reach high efficiency and strike the phase matching condition on broadband spectra (such as our cavity's) [262,263]. A 1 mm long crystal was chosen. The criterion to choose the crystal length is a tradeoff between the SHG bandwidth and the SHG efficiency: the longer the crystal, the higher the efficiency but the lower the SHG bandwidth. In our case, a 1 mm long crystal allows SHG with sufficient efficiency with a span that covers the emission spectra of our cavity.

To optimise the SHG efficiency, a compromise must be found about the focal length of the focusing lens, as mentioned in subsection 2.4.3. To determine the optimal focal length, a study was carried out using the SHG simulation software SNLO [264]. This software allows to compute the SHG efficiency ab-initio for tunable beam parameters at the surface of the crystal. These parameters at the surface of the crystal correspond to focal lengths of focusing lenses, the correspondance is given by ABCD matrix formalism [235] that SNLO also computes. Hence, the parameters that optimise the efficiency can be retrieved by trying different input parameters. The simulation results are displayed in figure 4.18.

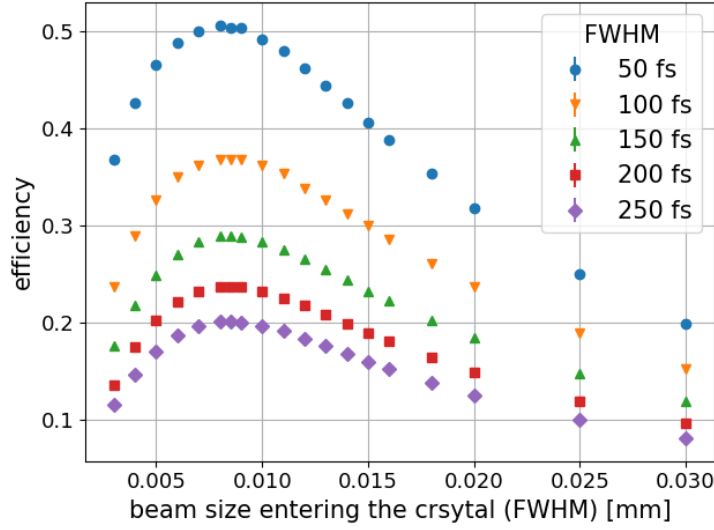


Figure 4.18: Simulated SHG efficiency as a function of the beam size entering a 1 mm long BBO crystal for different pulse durations, simulated by SNLO. The beam is focused at the center of the crystal. The pulse energy is set at 1.7 nJ. The central wavelength is set at 800 nm.

The simulated SHG efficiency decreases with the pulse duration, from 0.5 to 0.2. This is consistent with the fact that SHG increases with the peak power: since the pulse energy is kept constant, if the pulse duration increases, the peak power decreases, thus the efficiency decreases.

The simulated efficiency presents a maximum for a beam size entering the crystal of $\sim 80 \mu\text{m}$, independently of the pulse durations in the considered duration range (50 to 250 fs). This maximum is the compromise between phase-matching and high intensity described in subsection 2.4.3. Thus, the focusing lens was chosen to match this beam size. The closest commercially available focal length, that was chosen, was 60 mm.

Using this setup, UV beams with output power up to 80 mW, which corresponds to a $\sim 10\%$ efficiency, were generated. This power has decreased over time to stabilize around 30 mW, probably due to degradation and pollution of the BBO surface. However, this power level is largely sufficient to perform interferometric measurement using our Si-biased photodiode, so no further optimisation of the SHG efficiency was performed.

The output power is the same order of magnitude than the simulated process. The difference between the two can be attributed to misalignment, imperfections in the crystal or chirp, which was not considered in the simulation.

4.3.2 Cesium spectroscopy

To obtain a proof-of-concept result of the UV-DCS experiment, a cesium detection experiment was carried out.

Experimental setup

A gas cell containing cesium gas was put on the optical path, as illustrated in schema 4.1. The cell is 75 mm long and slightly wedged to avoid parasite reflections. At room temperature the Cs partly condensates on the walls of the cell, thus it was heated using a heat pistol before the measurements to sublime it and maximise the gas concentration in the cell. The absolute Cs concentration in the cell is unknown.

The detected Cs absorption lines are located at 387.615 nm (773.427 THz, $2.57987 \cdot 10^6 \text{ cm}^{-1}$) ($6S_{1/2} - 8P_{1/2}$ transitions) and 388.861 nm (770.949 THz, $2.57161 \cdot 10^6 \text{ cm}^{-1}$) ($6S_{1/2} - 8P_{3/2}$ tran-

sitions) [265]. Hence, to be able to detect them both simultaneously, a UV spectrum at least ~ 1.1 nm (2.2 THz, $6.9 \cdot 10^3$ cm $^{-1}$) large around 388 nm is needed, which corresponds to a doubled spectrum from a 2.2 nm (1.1 THz, $3.7 \cdot 10^3$ cm $^{-1}$) IR spectrum around 776 nm. This IR spectrum is achieved through precise alignment of the mirrors of the cavity, pump laser and prisms position. The simultaneous detection of two lines is necessary to be able to rebuild the optical frequency axis, as explained in subsection 4.1.3. The phase-matching angle of the BBO crystal is tuned empirically to maximise the contrast of the interferograms.

Raw spectra

The retrieved transmission spectrum is presented in figure 4.19 for an acquisition with $\Delta f_{\text{rep}} \sim 5$ Hz.

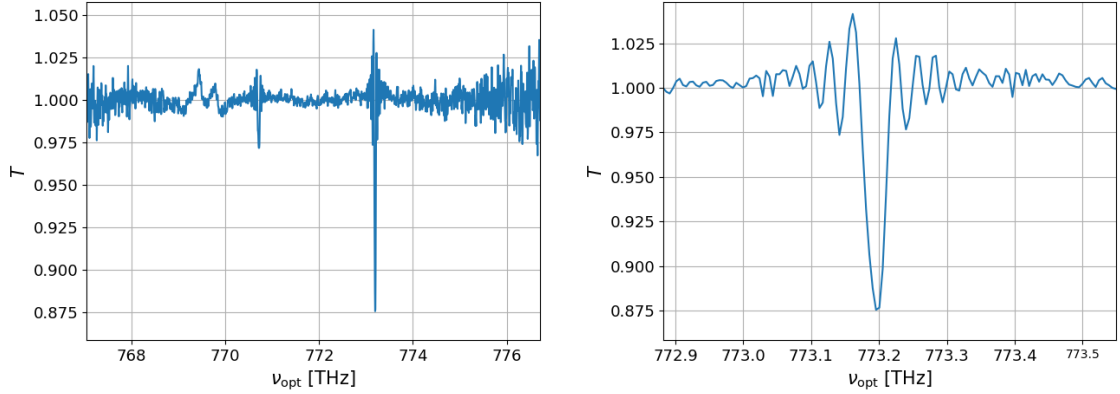


Figure 4.19: Retrieved transmission spectrum from a 5 ms FFT window around an interferogram (left) and close-up on an absorption line (right). The baseline was obtained with the FFT of a 30 μ s window around the interferogram and removed. This windowing corresponds to a maximum resolution of 400 Hz in the rf domain and 12 GHz in the optical domain. The optical frequency axis was rebuilt using the two cesium lines that appear in the spectrum.

The two Cs lines appear on the spectrum. Up to our knowledge, this is the first time both of these lines are retrieved in a single acquisition. This shows our spectrum spans over more than 2.5 THz in the UV range, with a raw SNR of the order of 10.

A coarse approximation of the resolution of our spectrometer in this $\Delta f_{\text{rep}} = 5$ Hz regime can be deduced from these measurements. Under the hypothesis that the retrieved Cs lines are very thin spectrally compared to the spectrometer resolution, the spectrometer resolution corresponds to the spectral width of the observed line. In this case, the line is approximately 50 GHz wide, while the Cs lines are GHz-wide [184]. Hence, the hypothesis can be validated a posteriori, and a coarse approximation of 50 GHz for the spectral resolution of the spectrometer in this regime can be given.

The absorption lines feature noise. In particular, an oscillating lineshape is observed. These oscillation can be due to Δf_{rep} fluctuations, as seen on section 2.3.

Cesium lineshape analysis

To test the hypothesis that oscillations on the transmission spectrum are caused by Δf_{rep} , predictions from the simulation described in section 2.3 about these oscillations are compared with the experimental data.

The simulation simulates the probing of one comb by the other, only the probed comb undergoing absorption. As mentioned in section 2.1.4, the temporal trace of this one-sided probing is a FID signal on one temporal side of the interferogram. Hence, to compare the predictions of the simulation with experimental results, the simulated spectrum must be compared with an experimental spectrum obtained by FFT of only one side of the interferogram.

The compared feature is that the simulated spectra show oscillations that are "inverted" when Δf_{rep} fluctuations change sign, as illustrated in figure 4.20. For the retrieved power spectrum, this change of sign of Δf_{rep} fluctuations is formally equivalent to a swap of the two combs, the probing becoming the probed and vice versa. This swap is equivalent to considering the other temporal side of the interferogram's centerburst: the first comb probes the second one before the centerburst and the second probes the first after the centerburst. Hence, to simulate a spectrum obtained by FFT of a temporal window on one side of the centerburst (resp. the other), the simulation can be run two times, only with Δf_{rep} fluctuations changing sign. The results are given in figure 4.20.

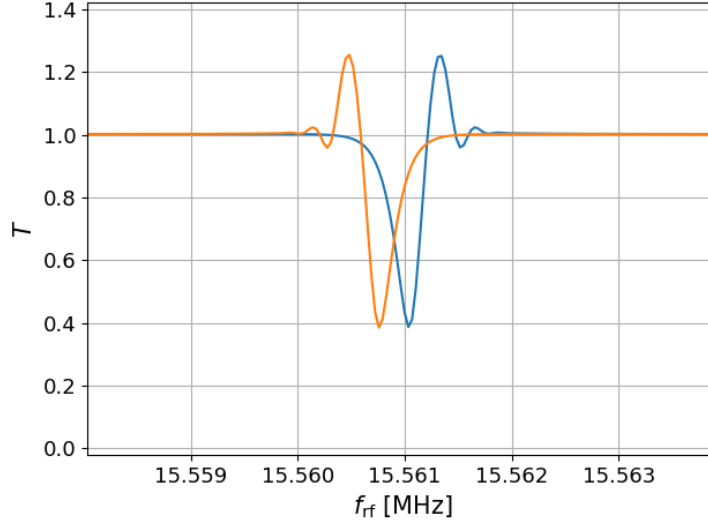


Figure 4.20: Simulated DCS transmission spectra, with Δf_{rep} fluctuations of opposite signs. The average value of the simulated Δf_{rep} is 40 Hz. The simulated fluctuations of Δf_{rep} are linear in time, with a variation corresponding to a $\sim 10^{-5}$ Hz shift during the centerburst (variation rate of 66.7 $\mu\text{Hz/ps}$).

The simulation predicts an inversion of the oscillations when considering opposite sides of the interferogram.

Spectra from opposite sides of the interferogram are presented in figure 4.21 with a close-up on an experimental absorption line.

An inversion such as the one predicted by the simulation is clearly visible. This tends to show that the oscillating lineshape can be attributed to Δf_{rep} fluctuations.

4.3.3 NO₂ detection

To obtain a proof-of-concept result of the UV-DCS experiment on atmospheric gases, a NO₂ detection experiment was carried out.

Experimental setup

A 50 cm long gas cell is set on the optical path, as illustrated in figure 4.1. To be able to set a known concentration of NO₂ in the cell, a pump, manometer and gas mixer are set up with a NO₂ gaz mixture bottle and a N₂ bottle. This setup allows to tune the absolute concentration in the cell from 0 to 1.5 bar. The NO₂ gaz mixture bottle is composed of a 1800 ppm concentration in NO₂, 1800 ppm concentration in N₂O, 4500 ppm in NO, diluted in N₂. Concentrations are given by the seller with a 10% uncertainty.

A NO₂ detector is also set up to detect any possible leak from the system, as NO₂ is a toxic gas [8].

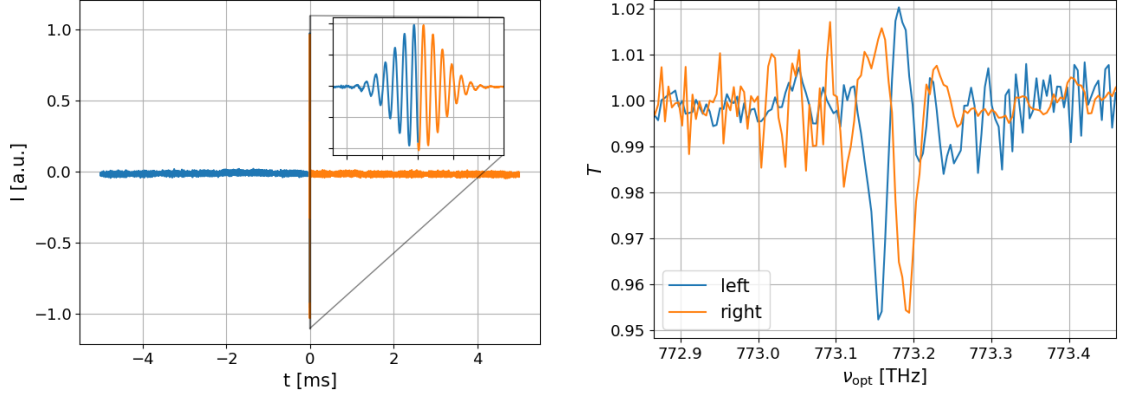


Figure 4.21: Retrieved transmission spectrum from a 10 ms FFT window left and right of an interferogram (right) and used windows (left). The baseline was obtained with the FFT of a $30\ \mu\text{s}$ window around the interferogram and removed. A Hanning window was applied to temporal traces before the FFT. The optical frequency axis was rebuilt using the two cesium lines that appear in the spectrum. Δf_{rep} is 5 Hz. This windowing corresponds to a minimum optical resolution of 470 MHz.

For security reasons, as NO_2 is a toxic gas, a NO_2 detector is also installed in the laboratory room. This detector is connected to a pneumatic valve that would close the gas circuit in the event of accidental diffusion of NO_2 in the laboratory.

To avoid parasitic reflections, the UV-fused silica windows of the cells are wedged at Brewster angle.

NO_2 detection protocol

A different protocol than the one used for O_2 and Cs must be applied for NO_2 detection, because of the large spectral width of its absorption lines.

The NO_2 absorption spectrum in the near UV features broad absorption lines, of the order of the spectral width of the emission spectrum, as illustrated in figure 4.22.

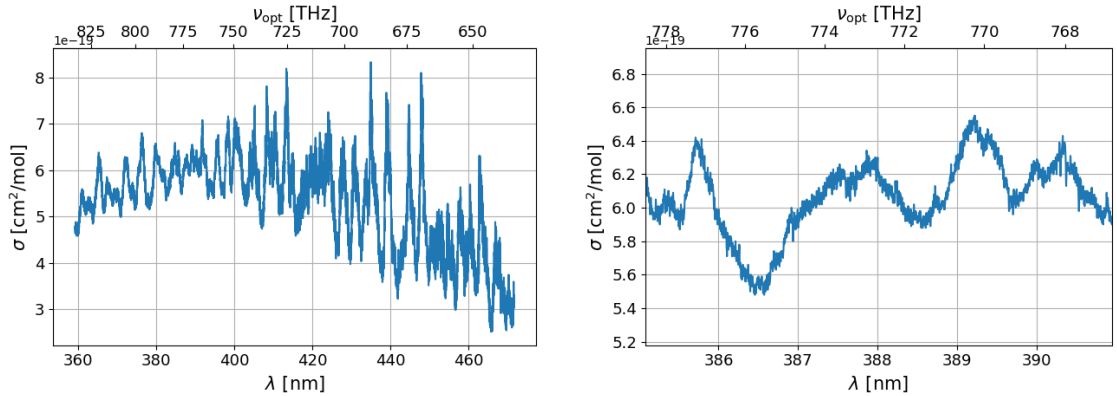


Figure 4.22: Absorption cross section of NO_2 for $T = 298.5\ \text{K}$, $P = 2\ \text{Torr}$ and close-up on a spectral window corresponding to the width of our emission spectrum for this series of measures. Data from Yoshino et. al. [266]. The cross section was retrieved with a resolution of $0.0025\ \text{nm}$ using a grating-based spectrometer.

Thus, the baseline retrieval process presented in subsection 4.1.2 cannot be used, since it relies

on the property that spectral absorption lines are much thinner than the spectral envelope of the emitted spectrum.

To retrieve the baseline, a spectrum is acquired with the cell containing only N₂. This spectrum serves as a baseline for following measurements. This method is more sensible to spectrum fluctuations than the previous one, because the baseline and the absorption spectrum are not recorded simultaneously: hence shifts in the laser emission between the two measurements can induce bias.

Moreover, the optical frequency axis retrieval protocol that uses narrow absorption lines as absolute references cannot be used anymore either with only NO₂ lines, as they are too broad. To tackle this problem, the cesium cell of subsection 4.3.2 is placed on the optical path: thus the spectra feature the narrow Cs absorption lines, that are used as absolute references to calibrate the frequency axis. This method is as reliable as for the Cs spectra measurements because the Cs lines are measured simultaneously with respect with NO₂.

NO₂ concentration is retrieved using a numerical fit of the transmission spectrum with Beer-Lambert's law:

$$T(\lambda) = 1 - \exp(-NL\sigma(\lambda)) \quad (4.4)$$

with T the transmission, N the gas concentration (molecules/m³), L the length of the optical path in the absorbing medium (m) and σ the absorption cross-section (m²/molecule). The used cross-section is from [266]. These cross-section data were obtained at low pressure, but the dependency of the cross section with respect with the pressure is negligible at our level of precision [267].

Results on NO₂

The retrieved spectrum is presented in figure 4.23.

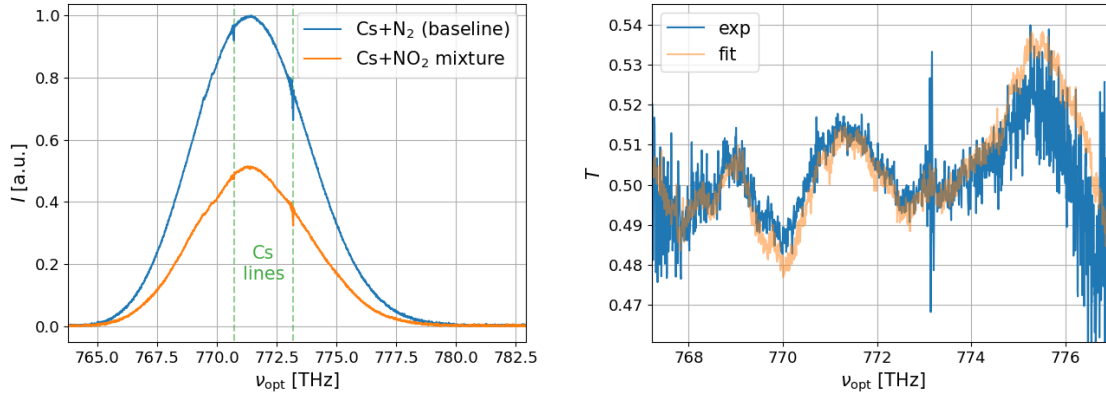


Figure 4.23: Spectra retrieved from a 5 ms window around an interferogram with and without NO₂ in the cell (left), and corresponding transmission spectrum and comparison with HITRAN database (right). Δf_{rep} is 5 Hz. This windowing corresponds to a minimum optical resolution of 3 GHz. The baseline spectrum measurement (without NO₂) was done 7 minutes before the measurement with NO₂. The NO₂ concentration in the cell was set to $4.85 \cdot 10^{17}$ molecules/cm³. The optical frequency axis was retrieved using the Cs absorption lines that appear on the spectra. The HITRAN fit is given by Beer-Lambert's law with a NO₂ concentration of $4.13 \cdot 10^{17}$ molecules/cm³.

The spectrum features the two Cs lines that allow to calibrate the frequency axis. This allows to show that the retrieved spectrum spans over 5 THz in the UV range. Its resolution can be estimated at 50 GHz, as explained in subsection 4.3.2.

The retrieved NO_2 concentration is $4.13 \cdot 10^{17}$ molecules/ cm^2 . It differs by 17% with the result expected from the set concentration: $4.85 \cdot 10^{17}$ molecules/ cm^2 . This difference is not understood yet. However, the spectrum clearly features spectroscopic signature from NO_2 . This constitutes a proof-of-principle UV-DCS result using our cavity. Up to our knowledge, this is one of the first UV-DCS atmospheric trace gas detection using a freerunning laser source, only preceded by the detection of formaldehyde by Frst et al. [182]. The state of the art of UV-DCS, including this work, is illustrated in figure 4.24.

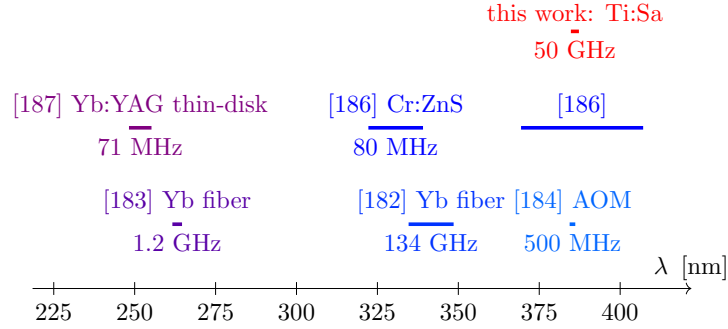


Figure 4.24: Current exhaustive state of the art of UV-DCS. Each bar represents a publication. The length of each bar represents the published spectral range. Above each bar is precised the used laser source. Below each bar is the spectral resolution. The only publications featuring atomic or molecular spectroscopic results are [184] (Cs spectroscopy), [182] (formaldehyde spectroscopy) and this work.

Conclusion

Proof-of-concept DCS experiments were carried out in the IR and the UV ranges, on atomic (for the UV) and molecular absorption lines of atmospheric gases, using our freerunning bidirectional Ti:Sa cavity. The retrieved IR spectra span over 20 THz in the near IR with a resolution of the order of the GHz, with single-shot measurement durations of the order of the ms around the centerbursts. The retrieved UV spectra span over several THz with a resolution of the order of 50 GHz in the near-UV, with single-shot measurement durations of 5 ms around the centerbursts.

The accuracy of the concentration retrieval process is limited by the observed asymmetric, Fano-type lineshape. The SNR improvement on the IR spectroscopic results, due to the better characterisation of the cavity, allowed to further detail this lineshape. Its presence on the UV spectra, and its analysis using the simulation, allowed to identify a possible cause for it, namely the fluctuations of Δf_{rep} . Hence, the Δf_{rep} fluctuations were identified as a limiting factor for spectroscopy using our cavity. The compensation of these fluctuations using an adaptive clock may greatly improve the sensitivity of our spectroscopy setup.

Conclusion and perspectives

A UV-DCS experiment was developed using our freerunning, bidirectional ring Ti:Sa laser cavity, leading to proof-of-concept results for trace gas measurement.

The development of this UV-DCS experiment required a characterisation of the used Ti:Sa bidirectional cavity.

Starting from previous work, the cavity's emission was characterised quantitatively. Output parameters such as spectral width, central wavelength, pulse duration and repetition rate difference between the two directions, were measured in relation to tunable parameters of the cavity such as mirrors alignment, pump power and negative dispersion induced by a prism compressor. The dependance of these output parameters on the tunable elements was systematically studied. The dispersion of the pulses inside and outside of the cavity was quantified, to find the fs pulses are in a low-dispersion regime.

The origin of the repetition rate difference between the two propagating directions was discussed. Experimental data tend to confirm the hypothesis that the repetition rate difference arises from the self-steepening effect of the pulses in the Ti:Sa crystal. Numerical calculations considering the dispersion inside the cavity, in the Ti:Sa crystal and the prisms, tend to foster that dispersion may also be the cause of the repetition rate difference.

Further investigation will require a numerical simulation of the Ti:Sa cavity, especially of the propagation of the pulses inside the Ti:Sa crystal, taking into account non-linear effects.

Spectroscopic results in the IR range were obtained using our cavity. The contrast of the interferograms was optimised by adjusting the wavefronts of the beams, to maximise their overlap on the photodetector. The retrieved interferograms temporal trace was treated numerically using an averaging protocol. This allowed to reproduce O₂ spectroscopy results from previous work, increasing the signal/noise ratio by a factor of 2. The use of post-acquisition correction algorithms failed, due to the level of phase noise in our freerunning cavity and baseline distortion effects arising from gain competition in the Ti:Sa crystal.

Considering the absorption spectroscopy experiments, asymmetric distorted lineshapes were systematically observed. The origin of these lineshapes was investigated and discussed.

A comparison with numerical simulation suggests that this distortion could be attributed to Fano effect. To further evaluate the possible origins of lineshape distortion, a DCS numerical simulation was developed during this work, based on a temporal model of DCS, that was developed during this work as well. The model allowed modelisation and simulation from the fs scale to the laboratory scale. The simulation produced results that tend to show that the lineshape distortion may be caused by repetition rate difference fluctuations.

To further investigate this issue, these hypotheses can be tested. The reproduction of the spectroscopic measurements with lower output power, by chirping the pulses, would give information about the dependance of the lineshape distortion with the non-linear Fano effect. The reproduction of the spectroscopic measurements with the repetition rate difference stabilised through an adaptive clock setup would give information about the dependance of the lineshape distortion with the repetition rate difference fluctuations.

Spectroscopic results were obtained in the UV range using our cavity.

Reaching the UV range was done through a second harmonic generation process with a BBO crystal. 80 mW output power was reached, which is theoretically enough for trace gas concentration retrieval in real atmosphere over a km long path. Proof-of-concept UV-DCS measurements was performed on cesium atomic lines, spanning over 3 THz with a resolution of the order of 50 GHz. Finally, trace gas measurements were performed on NO₂, spanning over 5 THz with a resolution of the order of 50 GHz, retrieving the expected concentration in a gas cell with an accuracy under 20%. These novel results are encouraging.

The implementation of real-time correction and/or post-processing algorithms are a prerequisite to improve the SNR of the DCS measurements in the IR and UV ranges. Adaptative clock sampling, as described in annex B, would allow to minimized the influence of the fluctuations of Δf_{rep} and Δf_0 during the acquisition process. Post-correction protocols are still to be adapted to our temporal baseline that is distorted by the gain depletion effect. These perspectives are currently experimentally and numerically investigated.

Reducing the uncertainty to the expected level is essential to make our UV-DCS spectrometer suitable for environmental monitoring.

Appendix A

Useful formulas for DCS modelisation

A.1 Formulas for dielectric electromagnetism

A.1.1 Definitions

Fourier transform

In this work, the Fourier transform $\widehat{E}(\omega)$ of a function of time $E(t)$ is defined by:

$$\widehat{E}(\omega) = \text{TF}(E) = \int_{-\infty}^{+\infty} E(t)e^{+i\omega t} dt \quad (\text{A.1})$$

and its inverse Fourier transform is defined by:

$$E(t) = \text{TF}^{-1}(\widehat{E}) = \frac{1}{2\pi} \int_{-\infty}^{+\infty} \widehat{E}(\omega)e^{-i\omega t} d\omega. \quad (\text{A.2})$$

Thus, the convolution product $*$ transforms under Fourier transform as:

$$(f * g)(t) = \text{TF}^{-1}(\widehat{f}(\omega)\widehat{g}(\omega)) \text{ and } (\widehat{f * g})(\omega) = 2\pi \text{TF}(f(t)g(t)). \quad (\text{A.3})$$

Linear, homogeneous, isotropic dielectric medium

In a linear, homogeneous, isotropic dielectric medium, the polarisation $\widehat{P}(\omega)$, dielectric susceptibility $\chi(\omega)$ and dielectric permittivity $\varepsilon(\omega)$ are linked by [219]:

$$\widehat{P}(\omega) = \varepsilon_0 \chi(\omega) \widehat{E}(\omega) = \varepsilon_0 \widehat{E}(\omega) (\varepsilon(\omega) - 1). \quad (\text{A.4})$$

The dielectric susceptibility can be decomposed into real and imaginary part:

$$\widehat{\chi}(\omega) = \widehat{\chi}'(\omega) + i\widehat{\chi}''(\omega). \quad (\text{A.5})$$

Its real part is linked with the optical index $n(\omega)$ by:

$$n(\omega) = \varepsilon(\omega)^{1/2} = \sqrt{1 + \widehat{\chi}'(\omega)}. \quad (\text{A.6})$$

Intensity

The intensity is defined up to a constant, by:

$$I(t) = |E(t)|^2 \quad (\text{A.7})$$

A.1.2 From Maxwell equations to the dispersion relation

Maxwell equations, in general, are given by (the bold terms are vectors) [219]:

$$\begin{aligned} \operatorname{div}(\mathbf{E}) &= \rho/\varepsilon_0 & \mathbf{rot}(\mathbf{E}) &= -\partial_t \mathbf{B} \\ \operatorname{div}(\mathbf{B}) &= 0 & \mathbf{rot}(\mathbf{B}) &= \mu_0 (\mathbf{j} + \varepsilon_0 \partial_t \mathbf{E}) \end{aligned} \quad (\text{A.8})$$

with $\mathbf{E}$ and $\mathbf{B}$ the electric and magnetic fields, ρ the electric charge density, $\mathbf{j}$ the electric current surface density, μ_0 the vacuum permeability and $\varepsilon_0 = 1/\mu_0 c^2$ the vacuum dielectric permittivity.

In a linear, homogeneous, isotropic medium without any magnetism, free charge or free current, they simplify into:

$$\begin{aligned} \varepsilon_0 \operatorname{div}(\mathbf{E}) &= \operatorname{div}(\mathbf{P}) & \mathbf{rot}(\mathbf{E}) &= -\partial_t \mathbf{B} \\ \operatorname{div}(\mathbf{B}) &= 0 & \mathbf{rot}(\mathbf{B}) &= \frac{1}{c^2} \partial_t \mathbf{E} \end{aligned} \quad (\text{A.9})$$

with $\mathbf{P}$ the polarisability of the medium, defined by:

$$\widehat{\mathbf{P}} = \widehat{\chi} \widehat{\mathbf{E}}. \quad (\text{A.10})$$

A.2 Semi-classical model for spectral lineshape

Using a classical damped spring-mass system to modelise the electrons' radial displacement around the nucleus, one obtains the following expression of $\frac{\omega}{c} \widehat{\chi}(\omega)$:

$$\frac{\omega}{c} \widehat{\chi}(\omega) = \widehat{\chi}_e(\omega) = \frac{NS}{\pi} \frac{\omega_0 - \omega + i\gamma_{\perp}}{(\omega - \omega_0)^2 + \gamma_{\perp}^2}. \quad (\text{A.11})$$

A.2.1 Temporal susceptibility

The inverse Fourier transform of the latter expression gives:

$$\chi_e(t) = i \frac{NS}{\pi} \theta(t) e^{-i\omega_0 t} e^{-\gamma_{\perp} t}. \quad (\text{A.12})$$

To use it in our model, the Fourier transform of the imaginary part of $\widehat{\chi}_e(\omega)$ is needed. To extract it, Fourier transform properties will be used.

A.2.2 Parenthesis on FT's mathematical properties

An arbitrary function $\widehat{f}(\omega)$ can be decomposed into $\widehat{f}(\omega) = \widehat{f}'(\omega) + \widehat{f}''(\omega)$, where $\widehat{f}'(\omega)$ is purely real and $\widehat{f}''(\omega)$ purely imaginary. Then (with $*$ the conjugate operation):

$$\begin{cases} f'(t) = f'^*(-t) \\ f''(t) = -f''^*(-t) \end{cases} \quad \text{so} \quad \begin{cases} f'(t) = \frac{1}{2} (f(t) + f^*(-t)) \\ f''(t) = \frac{1}{2} (f(t) - f^*(-t)). \end{cases} \quad (\text{A.13})$$

The introduction of the notation $\widehat{f}'' = ig$ allow to write:

$$g(t) = -\frac{i}{2} (f(t) - f^*(-t)). \quad (\text{A.14})$$

from which this useful result can be extracted:

$$\operatorname{TF}^{-1} \left(\Im(\widehat{f}(\omega)) \right) (t) = -\frac{i}{2} (f(t) - f^*(-t)) \quad (\text{A.15})$$

A.2.3 Final expression of temporal susceptibility

The latter property is applied to $\widehat{\chi}_e(\omega)$ (with the notations from 2.9), giving:

$$NSg(t) = -\frac{i}{2} (\chi_e(t) - \chi_e^*(-t)) = \frac{NS}{2\pi} e^{-i\omega_0 t} (\theta(t)e^{-\gamma_\perp t} + \theta(-t)e^{\gamma_\perp t}) \quad (\text{A.16})$$

and finally:

$$NSg(t) = \frac{NS}{2\pi} e^{-i\omega_0 t} e^{-\gamma_\perp |t|} \quad (\text{A.17})$$

From this, the quantity of interest can be identified, the envelope term of the free induction decay $\mathcal{A}(t)$ that is defined in equation 2.20 and that appears in the expression of the measured intensity (equation 2.43):

$$\mathcal{A}(t) = \frac{1}{2\pi} e^{-\gamma_\perp |t|} \quad (\text{A.18})$$

Finally, to be more realistic, one can take into account the Doppler effect, by adding a gaussian broadening term.

Appendix B

Adaptive clock

To improve the Signal/noise ratio (SNR) of the retrieved spectra, an adaptive clock can be set up. The idea is to synchronize the acquisition rate with the fluctuations of Δf_{rep} . This synchronisation would prevent the effects of Δf_{rep} fluctuations, which are important with our cavity, as seen in subsection 4.3.2.

B.1 Experimental setup

The experimental setup is schematised in figure B.1. It is similar to the one used in [151].

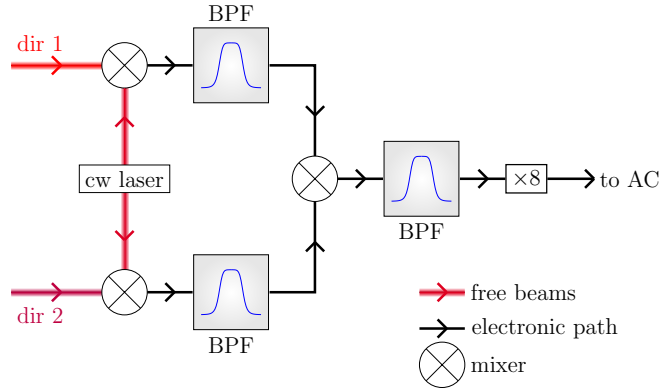


Figure B.1: Schema of the adaptive clock. Both beams beat with the same continuous wave (cw) laser. The beatings are filtered and made to beat with one another on a third mixer. The output signal is multiplied and sent to the acquisition card (AC) to serve as a clock signal.

The phase noise of each comb is sampled on a single tooth by making the combs beat with a stable, continuous-wave (cw) laser and bandpass-filtering the beating signal. The two beating signal then beat, which allows to sample the phase noise difference, namely the Δf_{rep} fluctuations. This beating signal is multiplied and used as a clock signal: hence the acquisition is synchronised with Δf_{rep} , including its fluctuations.

B.2 Modelisation of the adaptive clock protocol

The temporal field of a noisy comb ($j = 1, 2$) is given by:

$$E_j(t) = \sum_{n \in \mathbb{N}} E_{j,n} e^{2i\pi(f_{0,j} + n f_{\text{rep},j})t} e^{2i\pi(\delta f_{0,j}(t) + n \delta f_{\text{rep},j}(t))t} \quad (\text{B.1})$$

with a complex envelope $E_{j,n}$.

These combs are to interfere with a stable, continuous wave laser, whose temporal field is given by:

$$E_{\text{cw}}(t) = E_{\text{cw}} e^{2i\pi f_{\text{cw}} t}. \quad (\text{B.2})$$

The beating between the comb and the cw laser is given by:

$$I_{\text{b}} = E_j(t) E_{\text{cw}}^*(t) + \text{c.c.} \quad (\text{B.3})$$

Only one term of this beating is within the bandwidth of the filters. Let us note n_j the closest tooth of the comb j from the frequency of the stabilised cw laser. Then, the filtered beating is given by:

$$I_j = E_{j,n_j} E_{\text{cw}}^* e^{2i\pi(f_{0,j} + n_j f_{\text{rep},j} - f_{\text{cw}})t} e^{2i\pi(\delta f_{0,j}(t) + n_j \delta f_{\text{rep},j}(t))t} + \text{c.c.} \quad (\text{B.4})$$

This beating allows to sample the phase noise of one comb on a down-converted comb tooth.

Both combs beat with the continuous wave laser, and the beating of these two beatings reads:

$$I_{\text{mix}} \propto I_1^* I_2 = |E_{\text{cw}}|^2 E_{1,n_1}^* E_{2,n_2} e^{2i\pi(\Delta f_0 + n_1 \Delta f_{\text{rep}} + \Delta n f_{\text{rep},2})t} e^{2i\pi(\delta \Delta f_0(t) + n_1 \delta \Delta f_{\text{rep},j}(t) + \Delta n \delta f_{\text{rep},2})t} + \text{c.c.} \quad (\text{B.5})$$

Hence, the fluctuations of Δf_{rep} and Δf_0 are sampled, and using this signal as a clock corrects for these fluctuations. The frequency of this signal lies in the radiofrequency range. The frequency multiplication step illustrated in figure B.1 allows to increase this frequency to the order of 100 MHz, depending on the wanted acquisition rate.

This development relies on one implicit hypothesis: the absence of delay due to the travel time in the optic fibers and electronic wires. These delays raise experimental difficulty. To take them into account in this model, the t variable must be corrected into a $t + \delta t(t)$ in each term.

The main experimental challenge is to achieve a high enough SNR on the beating signals. This SNR is limited by the average power of the UV beams and the relative intensity noise of the cavity, whose effect is multiplied on the UV signal because of the power-dependant SHG efficiency.

To improve the SNR, optic fibers could be used as supports, because they allow a better coupling with the beating device. This experimental setup is being implemented.

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