

International Conference on Space Optics—ICSO 2018

Chania, Greece

9–12 October 2018

Edited by Zoran Sodnik, Nikos Karafolas, and Bruno Cugny



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Jérémy Oudin

Ajmal Mohamed

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International Conference on Space Optics — ICSO 2018, edited by Zoran Sodnik, Nikos Karafolas, Bruno Cugny, Proc. of SPIE Vol. 11180, 111802N · © 2018 ESA and CNES · CCC code: 0277-786X/18/\$18 · doi: 10.1117/12.2536014

Integrated Path Differential Absorption LIDAR measurements on atmospheric CO₂ and H₂O using dual comb spectroscopy

Jérémy Oudin¹, Ajmal Khan Mohamed¹, Philippe-Jean Hébert²

¹ ONERA Chemin de la Hunière BP 80100 FR-91123 Palaiseau cedex, France

² CNES 18 Avenue Edouard Belin, 31400 Toulouse, France

ABSTRACT

We are developing an Integrated Path Differential Absorption (IPDA) LIDAR based on dual comb spectroscopy and recently achieved measurements of CO₂ and H₂O concentrations over 280 meters in the atmosphere. High repetition rate (100 MHz) femtosecond fiber laser frequency combs with spectral bandwidth over 100 nm around 1550 nm are used.

Keywords: Dual comb spectroscopy, Lidar, spectroscopy, frequency comb

1. INTRODUCTION

Knowledge of greenhouse gases at worldwide level is a major issue to assess and eventually correct climate changes. We can quote two spatial missions coming up for 2021 having this purpose. On the one hand the mission MICROCARB will use a grating spectrometer in order to map sources and sinks of carbon dioxide, the most important greenhouse gas, on a global scale¹. On the other hand MERLIN will go to measure concentrations of atmospheric methane using an IPDA LIDAR². LIDARs are foreseen for such global measurements, however most of the actual LIDAR are mono-specie and with a long acquisition times. The use of frequency combs as a laser source appears as a good workaround. Intrinsically these lasers have a wide spectral coverage with a high repetition rate making possible a multi-species and fast acquisitions detection. Moreover, dual comb spectroscopy gets around the difficulty to record a wide spectral bandwidth with high resolution compared to classical Fourier spectrometers using a Michelson device.

2. DUAL COMB SPECTROMETER

The frequency comb results from the sum of phase locked optical frequencies³ delivered by a femtosecond laser as it is shown in fig. 1. These lasers are fully described by two parameters, the pulse repetition rate f_{rate} which fixes the gap between the teeth of the comb. The second parameter is the so called carrier envelope offset frequency f_0 , and because of it, the teeth of the comb are not the overtones of the pulse repetition rate. The carrier envelop offset comes from the temporal phase difference $\Delta\Phi$ between pulses. This phase difference results from the difference between the group velocity and the phase velocity inside the active laser medium.

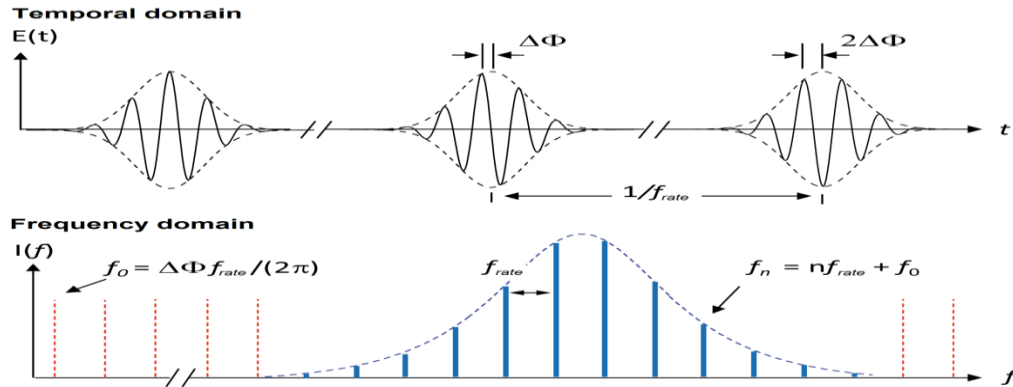


Fig. 1 : The frequency comb in the time domain: two subsequent pulses are separated by the round trip time $1/f_{rate}$ and show a phase shift $\Delta\Phi$. In the frequency domain two subsequent teeth are separated by the repetition rate f_{rate} . The temporal phase shift causes an offset f_0 in the frequency domain

In 2002 Schiller and al proposed a new spectroscopy method using two frequency combs⁴. Like a traditional Fourier transform spectrometer, this approach is based on the principle of down-converting optical frequencies. The principle of the experiment involves two sources of frequency combs with slightly different repetition frequencies. The two sources are combined and sent to probe the gas sample under study. Figure 2 shows that down conversion between the combs produces a comb in the radio frequency (RF) domain which can be detected with a a standard 1 Ghz bandwidth photodiode. The signal acquired on the photodiode shown in fig.3 is called an interferogram, its Fourier transform gives a correspondence of the optical spectrum in the RF domain. To retrieve the optical spectrum, the $\frac{\Delta f_{rate}}{f_{rate}}$ and f_0 values must be known. Measuring f_0 requires a spectrum spanning over one octave in order to use the well-known “f-2f” method⁵. It is also possible to use a reference absorption peak and only the $\frac{\Delta f_{rate}}{f_{rate}}$ ratio to retrieve the optical spectrum, method which we have used in the present work.

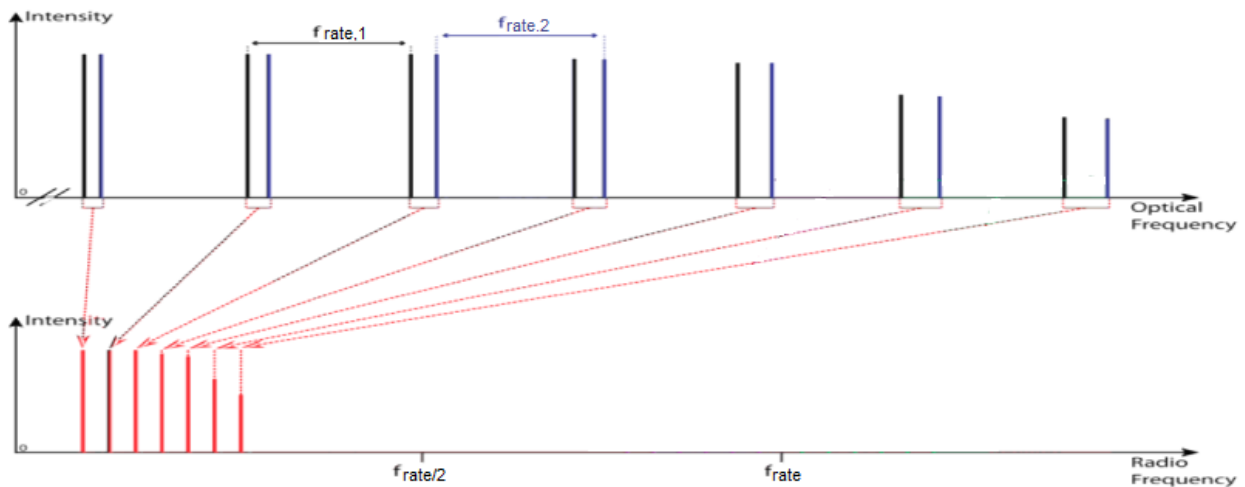


Fig. 2 : Two frequency combs with different repetition rates $f_{rate,1}$ and $f_{rate,2}$ are overlapped and form a beating signal that is a comb in the radio frequency domain. Hence, the optical frequencies are down-converted to radio frequencies by the scaling factor $\frac{\Delta f_{rate}}{f_{rate}}$ that is dependent of the detuning of the combs' repetition rates.

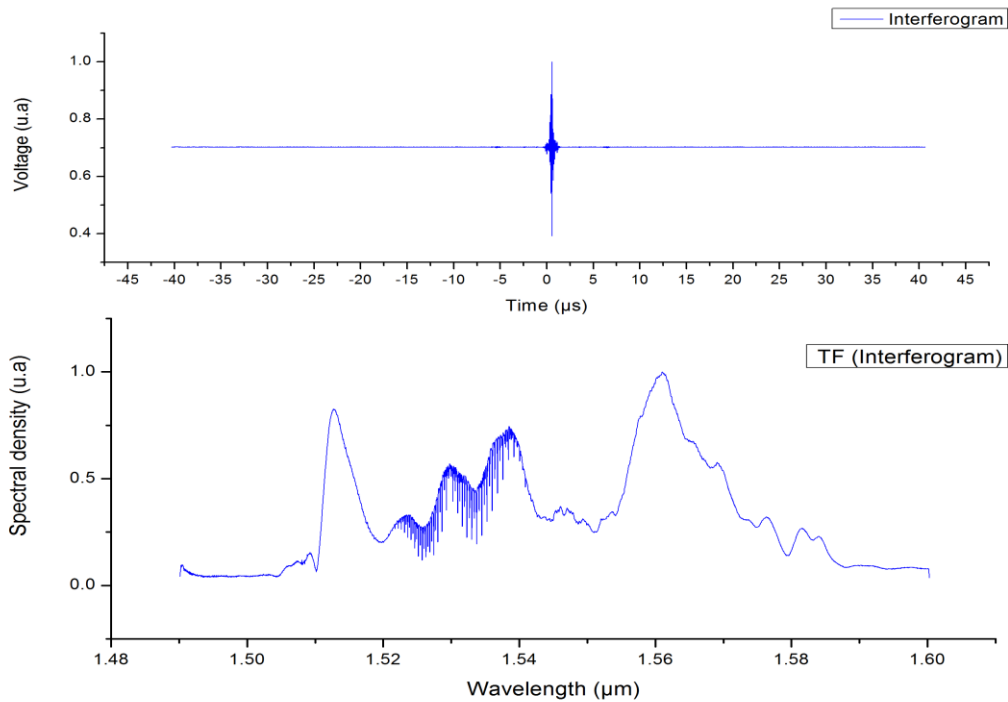


Fig. 3 : Upper graph shows an inferogram obtained probing a 10 cm cell filled with Acetylene. The acquisition time of the interferogram is 80 μ s and its Fourier transform shown in the lower graph gives the spectrum with many acetylene absorption peaks.

3. EXPERIMENTAL SETUP AND RESULT

We have performed some ground tests of our IPDA LIDAR similarly to those performed recently by Rieker and al⁶. The sketch of our experimental setup is illustrated in Fig.4. Two Er-doped fiber optical frequency combs working at 100 MHz are arranged to have an offset repetition rate of 220 Hz. Each comb provides a pulse train with an average power of ~ 60 mW and pulse duration of ~ 70 fs. The laser beams from the two frequency combs are combined and sent through 140 meters of atmosphere then reflected by a retroreflector before collection by a 200 mm diameter telescope focusing on a photodiode. The interferometric signal is then digitized by a high speed acquisition card.

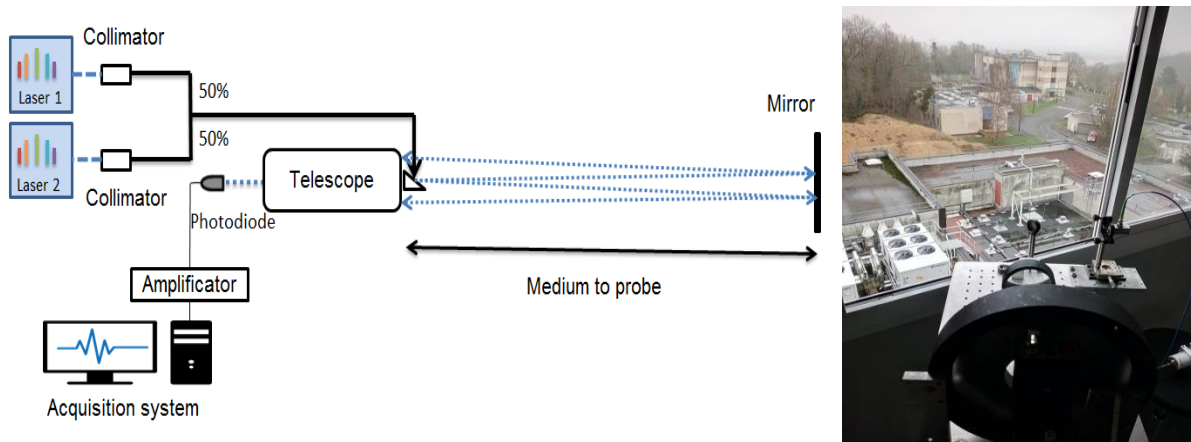


Fig. 4 : a) Sketch of the IPDA Lidar setup. Two Er frequency combs are sent in the atmosphere, passing through 140 meters targeting a retro reflector. The reflected beam is collected by a 200 mm diameter telescope focusing on a photodiode. The interferometric signal is then digitized by a high speed acquisition card. b) Picture of the beam's path. In the foreground the telescope and in the background the building having on the retro reflector.

A single shot interferogram is recorded in $70 \mu\text{s}$ which can be repeated every 5 ms ($1/\Delta f$). Its Fourier transform gives the absorption spectrum of the probed 280 meters path length. The recorded spectral domain equals the spectral bandwidth of the sources (100 nm around 1550 nm) where H_2O and CO_2 lines are present with a spectral resolution of about 6.2 GHz. We recorded 500 interferograms in approximately 5 seconds ($500 \times 5 \text{ ms} = 2.5 \text{ s}$), the mean enables to reach a satisfactory signal to noise ratio allowing to observe easily CO_2 and H_2O absorption peaks as shown in fig.5.

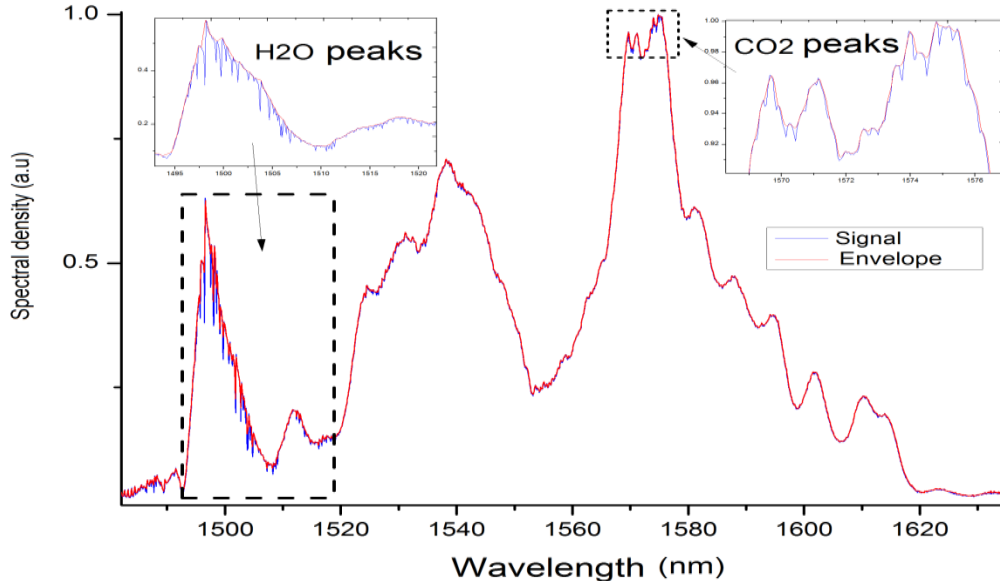


Fig. 5 : In blue line the absorption spectrum with H_2O absorption peaks around 1500 nm and CO_2 absorption peaks around 1580 nm. In red line the envelope which is obtained by removing the points belonging to the absorption peaks. Linear interpolation is used to substitute the deleted points.

The spectrum is calibrated with respect to a reference absorption peak of water at 1498 nanometers and the $\frac{\Delta f_{rate}}{f_{rate}}$ value. Then the envelope of the spectrum is suppressed to get the atmospheric transmission through the 280 meters. Afterwards we fit the transmission from the HITRAN 2012 data base to extract the concentration in CO_2 and H_2O along the laser beam path (Fig.6). The standard ambient temperature and pressure are used to configure the fit. The analysis of the absorption spectra provides concentrations of 5000 ppm for H_2O and 300 ppm for CO_2 with our present signal to noise ratio. We are looking for a method to estimate the uncertainties of these results. Different sources of uncertainty have to be considered: the stability of the parameters f_0 and f_{rate} , the error induced by the difference between the real envelope and the obtained envelope, the fit error and the signal to noise ratio.

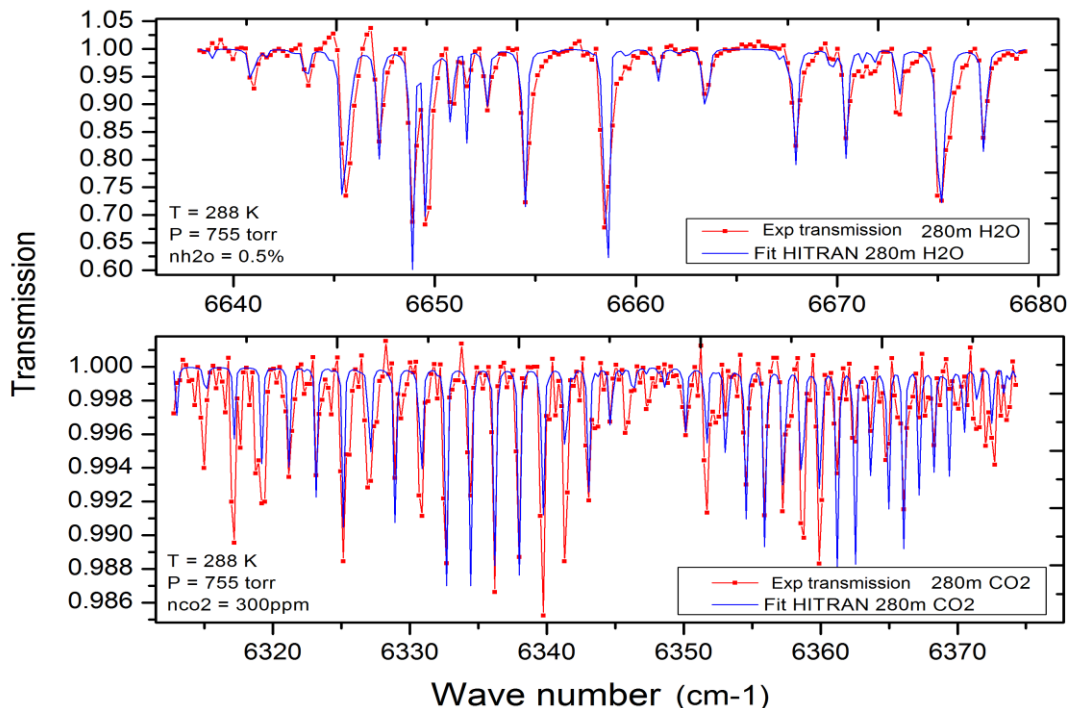


Fig. 6 : Upper graph shows the atmospheric transmission centered on the H_2O absorption peaks and lower graph the atmospheric transmission centered on CO_2 absorption peaks. Experimental data is shown in red and the fit from the HITRAN 2012 data base is drawn in blue. The fits from HITRAN return concentrations of 5000 ppm for H_2O and 300 ppm for CO_2 under standard ambient temperature and pressure.

4. CONCLUSION:

A dual comb spectroscopy set up was achieved allowing to detect and measure the concentration of some greenhouse gas species in the atmosphere. The first results give the CO_2 and H_2O concentrations measured over 200 meters laser path in the atmosphere. Currently the lasers temporal stability is the limited parameter. We are presently working on this issue through improvement of the control or compensation⁷ of the parameters f_{rate} and f_0 in order to increase the sensitivity of the spectrometer. We are also working on a method to estimate the uncertainties on the measured concentrations from different identified error sources.

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