EPJ Web of Conferences **287**, 07019 (2023) https://doi.org/10.1051/epjconf/202328707019
EOSAM 2023

Feasibility of dual comb spectroscopy in the UV range using a free-running, bidirectional ring titanium sapphire laser

Abel Feuvrier[1*], *Clément* Pivard[1], *Jérôme* Morville[1], *Patrick* Rairoux[1] and *Sandrine* Galtier[1]

[1]Université de Lyon, Université Claude Bernard Lyon 1, CNRS, Institut Lumière Matière, 69622 Villeurbanne, France

Abstract. We show that our developed free-running, bidirectional ring Ti:Sa laser cavity meets the requirements for Dual Comb Spectroscopy in the UV range (UV-DCS). Two counter-propagative frequency combs with slightly different repetition rate are generated in such a cavity and we show quantitatively that this repetition rate difference can be explained by the self-steepening effect. Molecular absorption lines of the O_2 *A*-band centered around 760~nm are measured with a 1,5 GHz spectral resolution, demonstrating that the mutual coherence of the two combs allows GHz-resolution DCS measurements. Moreover, we demonstrate that the generated output peak power allows for efficient second harmonic generation (SHG), in the scope of developing laboratory and open-path UV-DCS experiments.

1 Introduction

Dual-Comb Spectroscopy (DCS) is an emerging spectroscopy technique that combines large application bandwidths (from THz to visible), fast acquisition time (down to ms) and spectral resolution (down to kHz). It has successfully been used to probe the molecular composition of the atmosphere accurately, on large bandwidth, and without suffering from air turbulence [1-3]. Its use in the UV range (UV-DCS) would be of great interest for atmospheric physics and chemistry, allowing to remotely observe oxidant short living molecular species such as OH, HONO, BrO, etc [4].

DCS uses the beating of two optical combs with slightly different repetition rate ($\Delta f_{rep} \neq 0$) in order to produce a detectable, radio-frequency signal, in which the spectroscopic information of the medium is encoded. Thus, to achieve UV-DCS, one has to produce two optical combs with $\Delta f_{rep} \neq 0$ and a high degree of mutual coherence; moreover, these combs must have sufficient power to overcome light scattering and absorption of the atmosphere in this spectral range. In this proceeding, we show that the developed laser cavity meets these requirements without the need for active stabilization of combs.

2 Experimental setup

The production of two frequency combs with $\Delta f_{rep} \neq 0$ with an unique cavity can be achieved though several multiplexing techniques [5,6]; we use here a bidirectional Ti:Sa cavity, whose design is shown in figure 1 [7]. In order to quantify the source of $\Delta f_{rep} \neq 0$, we have investigated the differential intracavity pulse energy as function of Δf_{rep}. Note that this differential pulse energy is linked to the asymmetry of the cavity in which the Ti:Sa crystal is not strictly centered at the focus point between M1 and M2 mirrors. A clear correlation between the

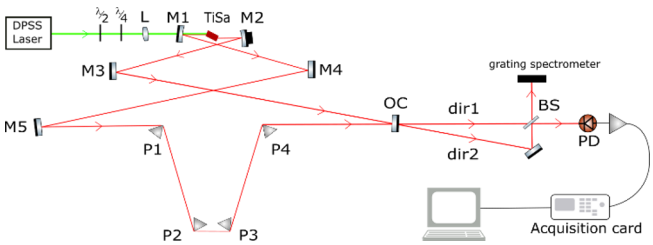

Figure 1. Home-made bidirectional Ti:Sa cavity, pumped by a 532 nm commercial cw laser (Coherent® Verdi V5). L: lens; M: mirrors; P: prisms; OC: output coupler; BS: beamsplitter; PD: photodiode. $f_{rep} \approx 118$ MHz.

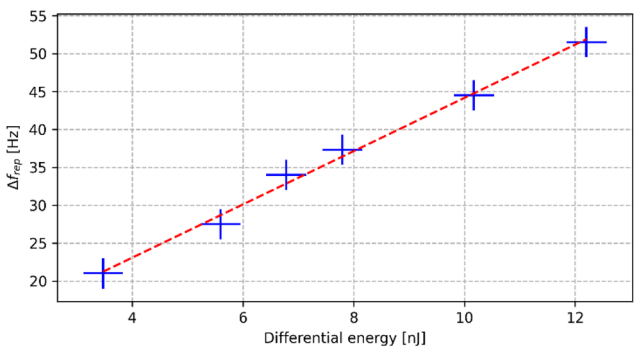

Figure 2. Repetition rate difference between the two directions as function of intra-cavity energy difference. Blue: experimental data; red: linear fit.

differential intra-cavity pulse energies and Δf_{rep} emerges (see figure 2).
The slope of the linear fit (3,51±0,14 Hz/nJ) applied to the data of figure 2 translates into a change of group velocity of 0.25(3) fs/nJ. This value is consistent with the magnitude of the self-steepening effect given in the literature [8]. This result validates quantitatively, and for the first time, the self-steepening effect as the source of differential repetition rate in the bidirectional ring cavity.

* Corresponding author: abel.feuvrier@univ-lyon1.fr

Moreover, by recording low resolution spectra of both combs as a function of Δf_{rep} (figure 3), we observe that the higher the value of the differential repetition rate the more the two spectra differ in width and spectral position. This shows the existence of an upper limit for Δf_{rep}, as the DCS spectrum spanning is given by the overlap of the two spectra.

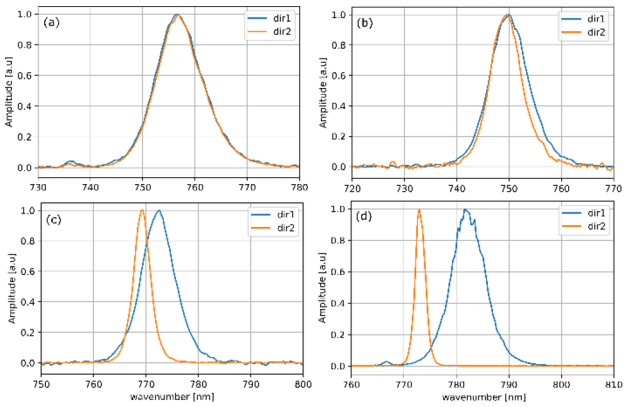

Figure 3. Normalized spectra of the two laser beams (dir1 and dir2), measured with a grating-based spectrometer for different Δf_{rep} values: 1.2 Hz (a), 10.4 Hz (b), 107 Hz (c), 425 Hz (d).

3 Demonstration of the mutual coherence of the combs: O_2 detection

In the bidirectional ring Ti:Sa laser cavity, the two combs are created with a single set of optics, share the same gain medium (Ti:Sa crystal), and the same pump laser. Therefore, the counter-propagating combs present an intrinsic mutual coherence, without a need for any feedback loop. To obtain an (inferior) estimation of this mutual coherence, a O_2 detection DCS measurement has been carried out. We use the setup depicted on figure 1 to obtain interferograms. One single-shot interferogram is then truncated within a 5,3-ms time window. The Fourier transform of this window gives the RF spectrum, whose baseline is removed using a polynomial fit to retrieve the transmittance graph illustrated in figure 4. The optical frequencies are calibrated using the observed O_2 $b^1\Sigma^+_g - X^3\Sigma^-_g$ absorption lines. The scaling factor from the RF to optical frequencies $(\Delta f_{rep}/f_{rep})$ was verified independently thanks to a Fabry-Perot interferometer [9].

The comparison with the HITRAN database shows that O_2 lines are resolved with a 1,5-GHz resolution, which is a two order of magnitude resolution improvement compared to previous results obtained with a bidirectional-ring Ti:Sa cavity [12]. Moreover, the quality factor is 3.5×10^6 $Hz^{1/2}$ (5.5×10^6 $Hz^{1/2}$ at maximum SNR [9]). This value is of the same order of magnitude than the state-of-the-art value using single-cavity dual-comb spectroscopy in the near-infrared [13].

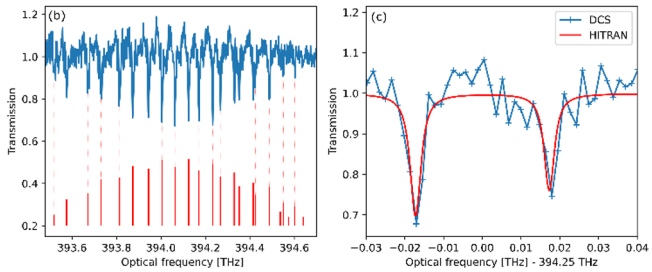

Figure 4. Absorption lines obtained with the setup from fig.2. (b) is a close-up of (a). Blue: experimental data; red: model from the HITRAN database [10]. The lines correspond to O_2 $b^1\Sigma^+_g - X^3\Sigma^-_g$ electronic transitions.

4 Extension towards the UV range

Ti:Sa wavelengths (760-810 nm) give access to near UV (NUV, 380-405 nm) by second harmonic generation (SHG), and to UV (250-270 nm) by third harmonic generation (THG). The relative coherence and power of the SH and TH frequency combs are critical to achieve UV-DCS measurements.
The reported resolution in the infrared range meets the requirements for atmospheric spectroscopy in the UV range, as reported in our feasibility study [4].
Output power ranges from 200 mW to 400 mW in each direction, depending on the position of the prisms and intra-cavity mirror alignment. Fourier-limited pulse duration ranges from 100 to 200 fs (intensity FWHM) and have been measured using a commercial autocorrelator (APE pulseCheck NX). The pulse temporal trace corresponds to a $sech^2$ shape. With such values, we generate 80 mW averaged output power in the UV range through SHG with a BBO crystal (type I SHG). In a previous paper [4], we have shown that these values meet the prerequisites for open-path UV-DCS measurements.

Fundings. Univ. Lyon 1 (FRAMA), Agence Nationale de la Recherche : project n°ANR-22-CE42-0002-01.

References

1. I. Coddington et. al., Optica **3**, 4 (2016)
2. N. Picqué et. al., Nature Photonics **13**, 146 (2019)
3. D. Herman et. al., Science Advances **7**, 14 (2021)
4. S. Galtier et. al., Remote Sensing **12**, 20 (2020)
5. K. Fritsch et. al., Nature Comm. **13, 2584** (2022)
6. N. Modsching et. al., Optics Express **29**, 10 (2021)
7. C. Pivard, PhD thesis, UCBL Lyon 1 (2021)
8. M. Sander et. al., Optics Express **18**, 5 (2010)
9. S. Galtier et. al., Optics Express, **30**, 12 (2022)
10. I. Gordon et. al., JQSRT, **277**, 107949 (2022)
11. N. Hébert et. al., Optics Express **25**, 7 (2017)
12. T. Ideguchi et. al., Optica **3**, 7 (2016)
13. L.A. Sterczewski et al, Opt. Express **27**, 17 (2019)