Spectroscopy techniques using frequency-comb : the special case of Dual comb spectroscopy

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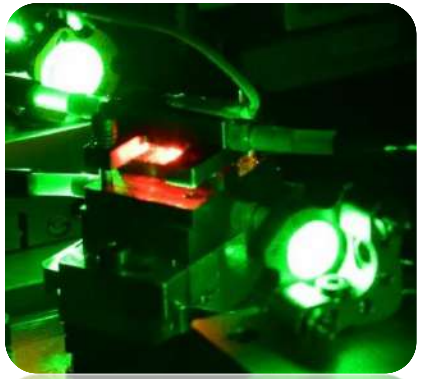
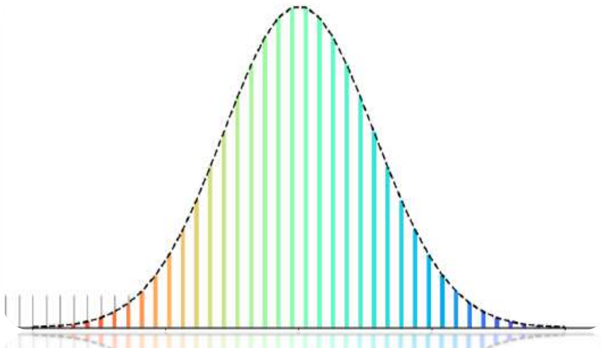




JSM – March, 10 , 2025

Spectroscopy techniques using frequency-comb

Partie I: Spectroscopy with combs

- Introduction
- Different techniques of Frequency comb spectroscopy

Partie II: Dual Comb spectroscopy

- Dual Comb Principle
- Dual Comb in the international community
- Extension towards the UV range

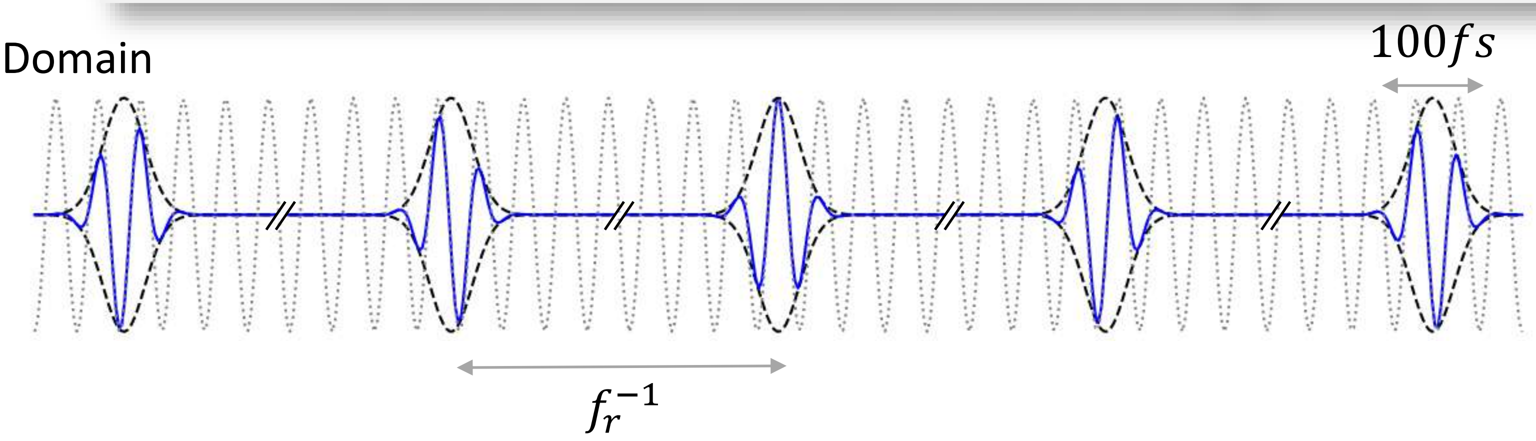
Partie III: The Dual Comb experiment at ILM

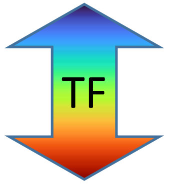
- Experimental setup
- Results

A Keer-Lens mode-locked frequency comb

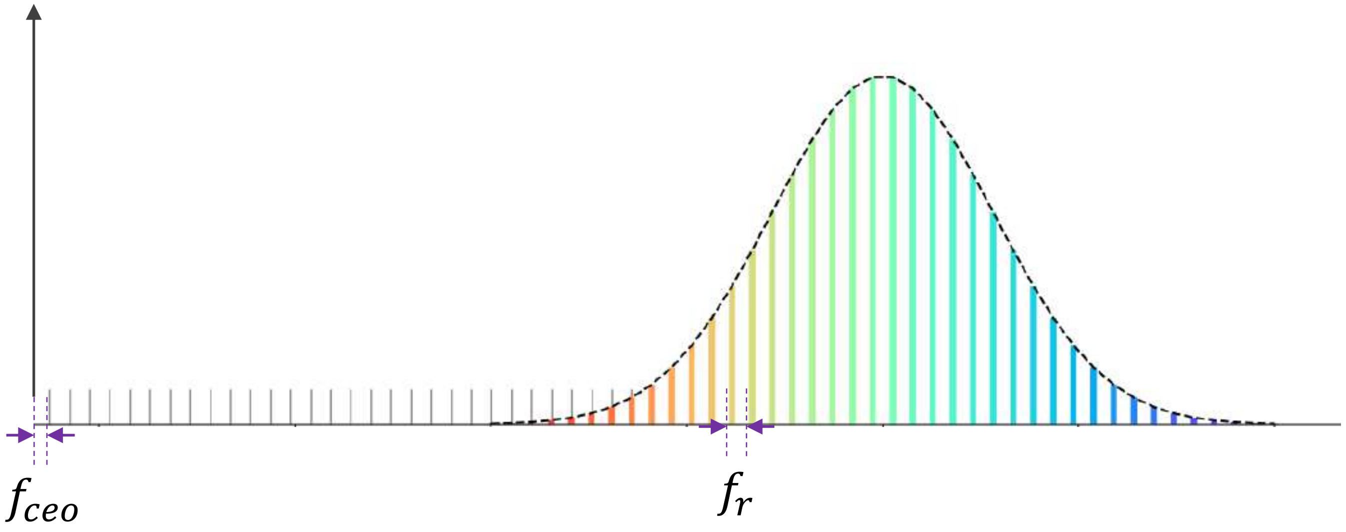
Time Domain

Passively Modelocked laser





Frequency domain

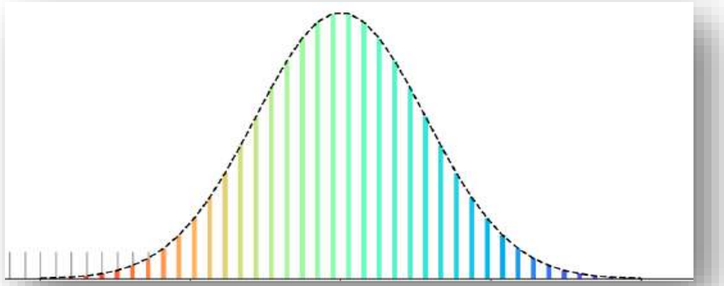


$\nu_n = f_{ceo} + n\, f_r$

Nobel Prize 2005
Theodor Hänsch
John Hall

Frequency comb spectrometer(S)

Broadband spectroscopy + comb mode resolution + frequency precision



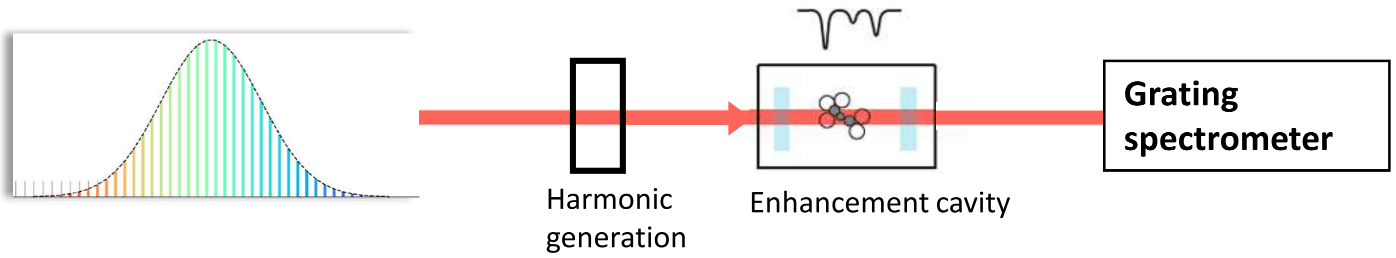
Fluorescence on atomic system

Dispersive element (+Fabry Perot)
- Using a diffraction grating
- Virtually-imaged phasearray (VIPA)
- Vernier spectroscopy

Time domain techniques
- FTS using a Michelson interferometer
- FTS with Dual Comb
- Ramsey-comb spectroscopy
- Other heterodyne measurements (THz)

First Cavity-enhanced Frequency-comb spectrocsopy

Frequency comb = bright source + broadband + harmonic generation possible

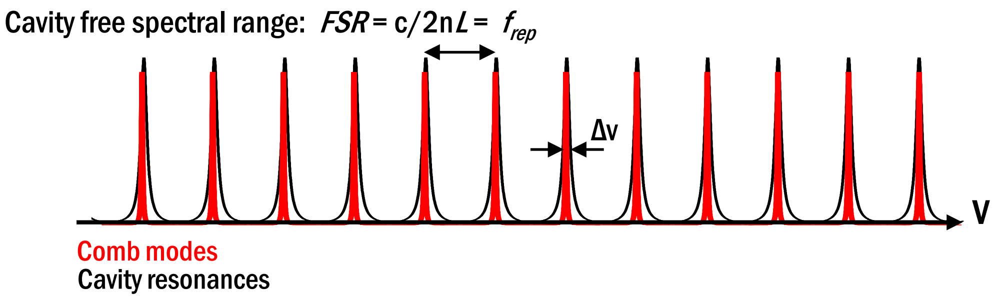


Resolution 0.03 to 0.07 nm in the UV range

[T. Gherman, D. Romanini, Opt. Express 10, 1033 (2002)]
[G. Méjean, S. Kassi, and D. Romanini – Optics Letters Vol 33 n°11 (2008)]
[Grilli *et al*, Environmental Science & Technology, Vol 26 (2012)]

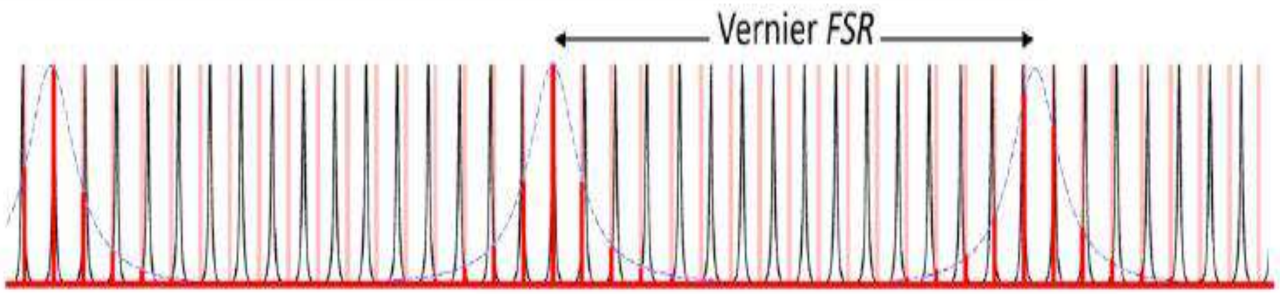
- **Mode-locked Cavity enhanced Dual Comb Spectroscopy (ML-CEAS)**

In-situ and Real Time Measurements of IO, BrO, NO2, and H2CO at pptv and ppqv



Bandwidth between 50 et 100 cm^{-1} depending on the cavity finesse
Need of low-dispersion cavity mirrors.

Comb resolution with a Grating-based spectrometer?



[Review: Lu et al, Photonics 2022, 9, 222]

- **Vernier spectroscopy**

GHz resolution over 2000 cm^{-1}

[L. Rutkowski and J. Morville, JQSRT , vol 187 (2017)]

see talk L1_Talk4 Jérôme Morville “Cavity Enhanced Frequency Comb Vernier Spectroscopy d'hydrures métalliques produit par pulvérisation cathodique »

- **virtually-imaged phasearray (VIPA) spectrometer**

1.2 GHz sampling frequency, at 633nm over 10nm bandwidth.

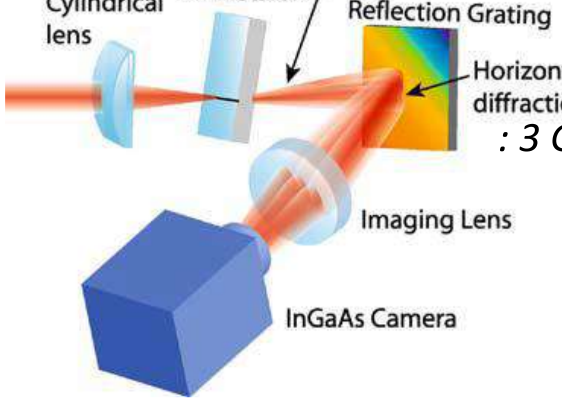
[S.A. Diddams, L. Hollberg, V. Mbele, Nature 445, 627 (2007)]

- First observation of the OD + CO → DOCO kinetics

[B. J. Bjork *et al* Ye, *Science* **354**, 444-448 (2016)]

- Recently 94 MHz resolution, observation of N2, H2, CH4 in a plasma

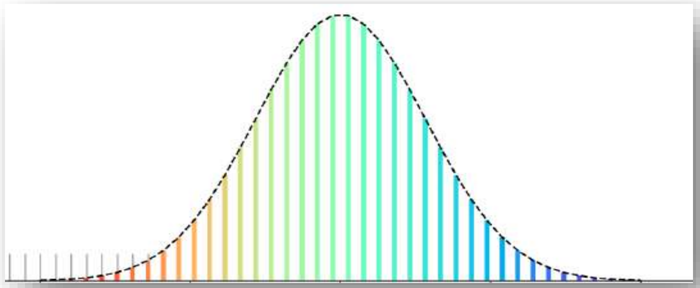
[Sadiek *et al*, Optics Express 32, 26 (2024)]



[Adapted from Review of:
Thorpe, M.J and J. Ye Appl. Phys. B 91, 397–414 (2008)]

Frequency comb spectrometer(S)

Broadband spectroscopy + comb mode resolution + frequency precision



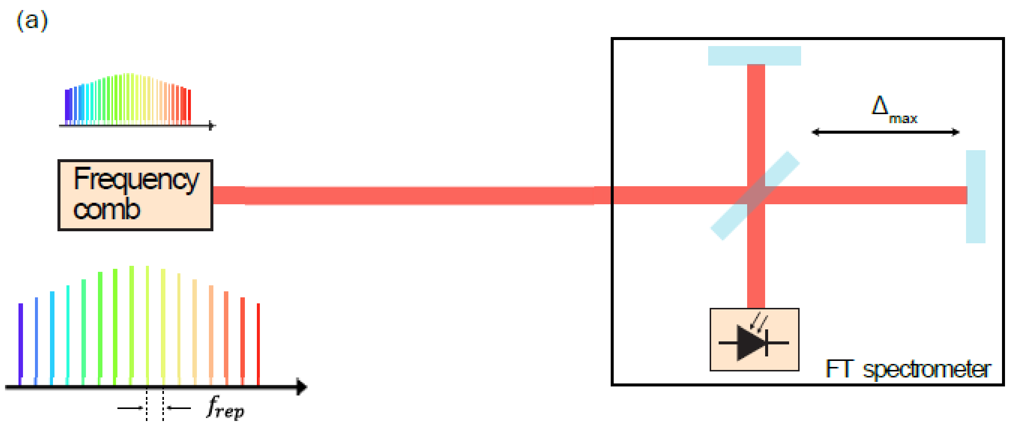
Dispersive element (+Fabry Perot)

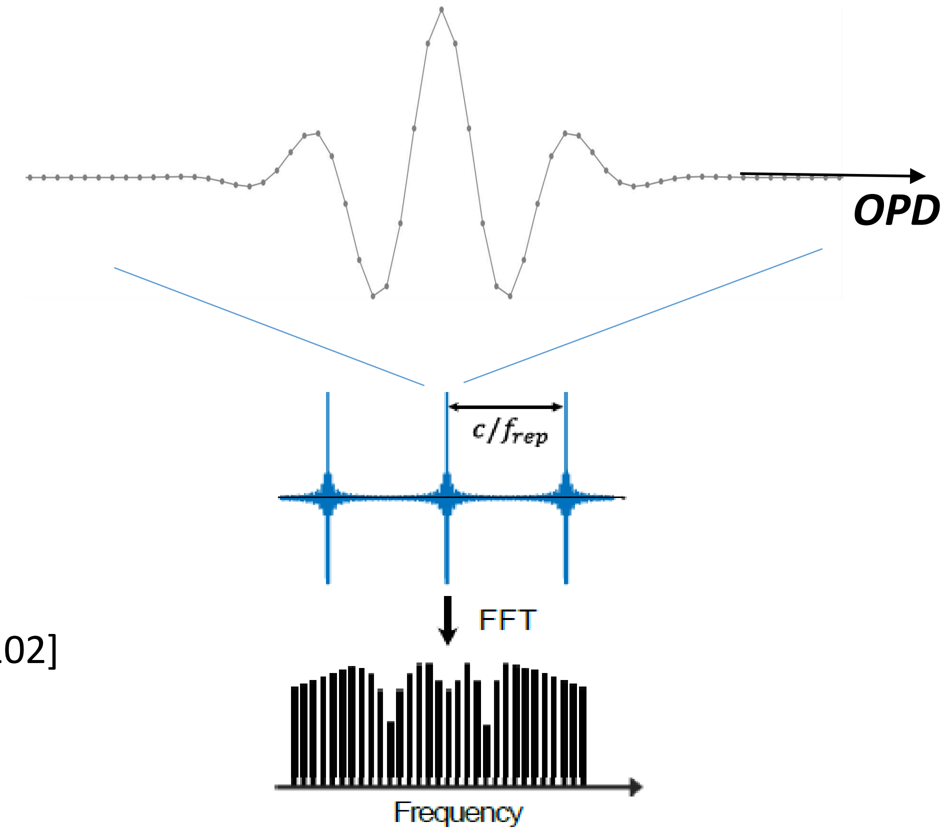
- Using a diffraction grating
- VIPA
- Vernier spectroscopy

Time domain techniques

- FTS using a Michelson interferometer
- FTS with Dual Comb
- Ramsey-comb spectroscopy
- Other heterodyne measurements

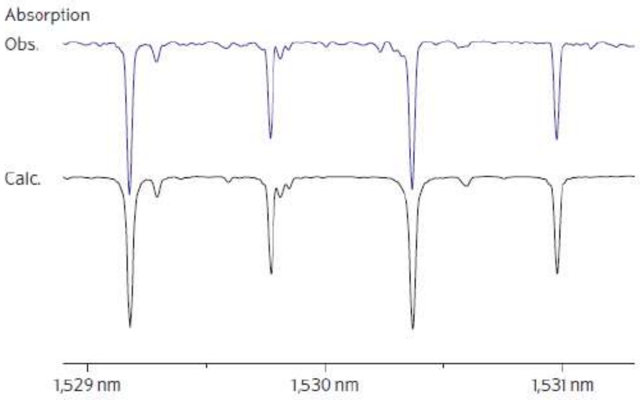
Fourier Transform spectroscopy with Combs



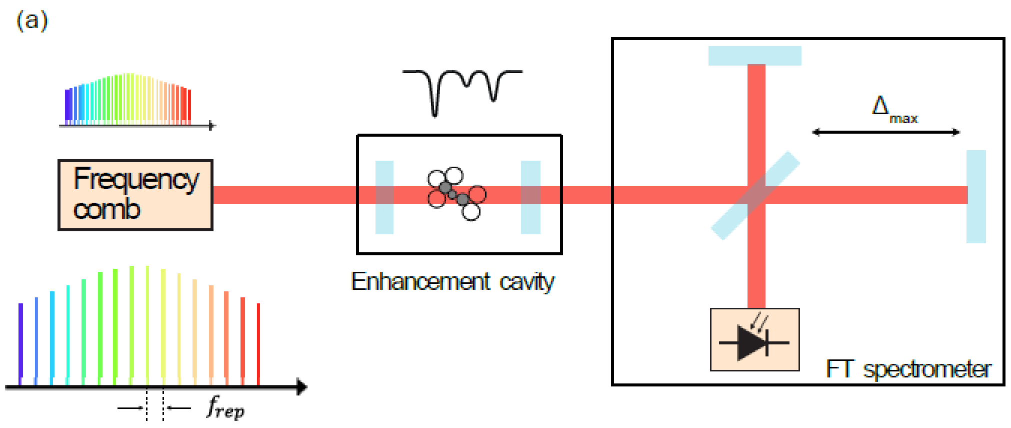


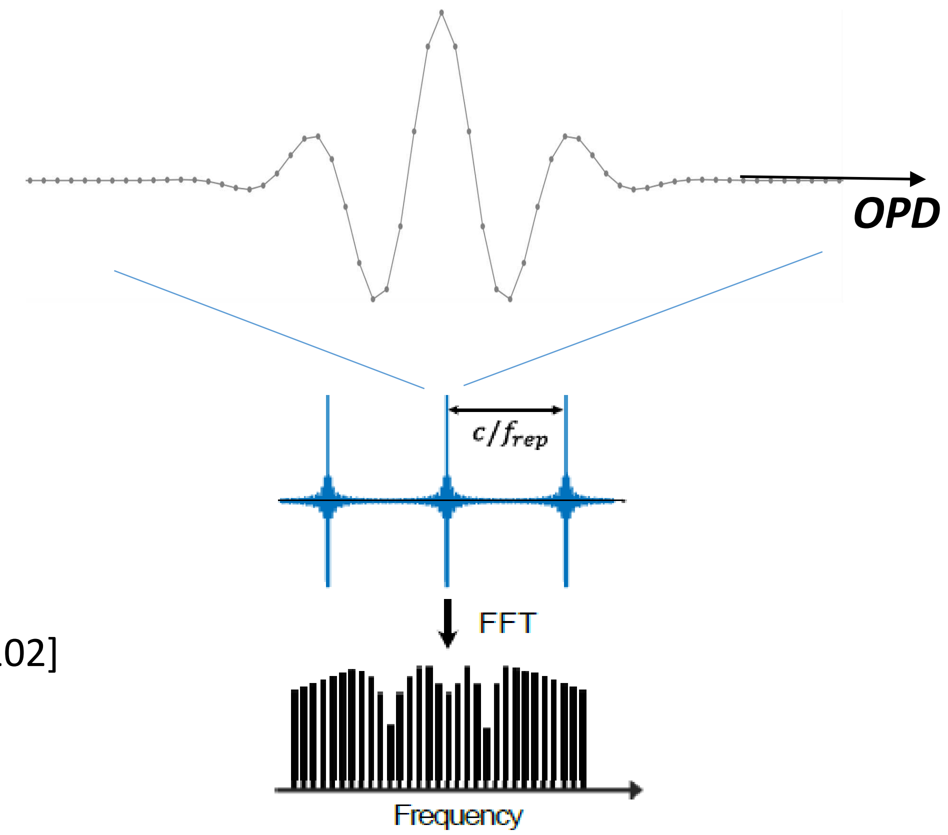
[Mandon, J.; Guelachvili, G.; Picqué, N. Nat. Photonics 2009, 3, 99–102]

resolution of 1.5 GHz, spanning 80 nm @1.5μm (C_2H_2):



Fourier Transform spectroscopy with Combs





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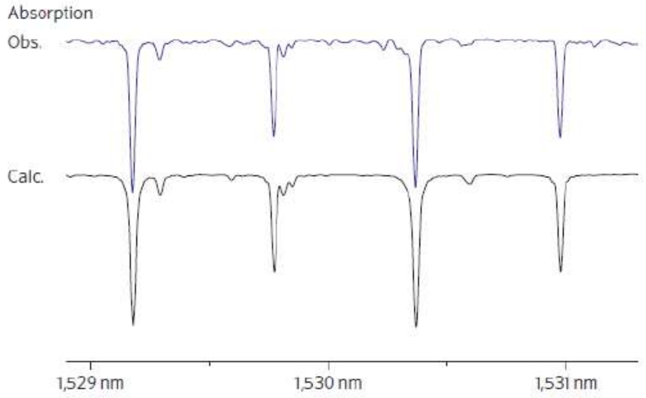
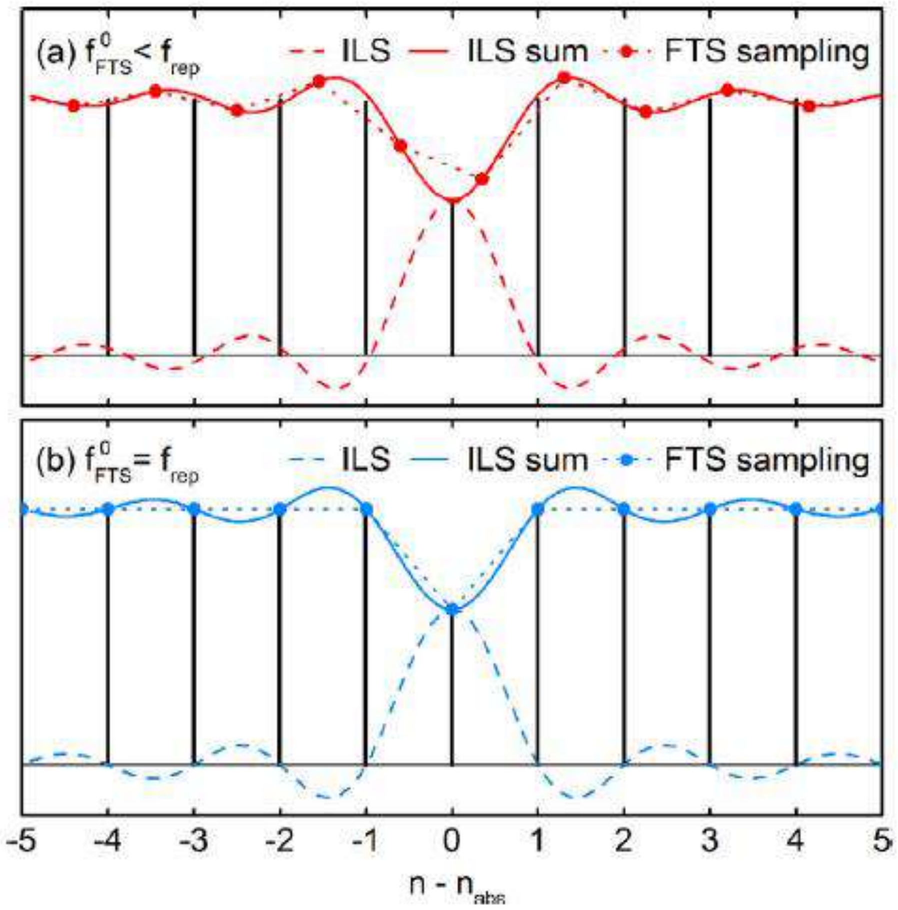


+ Enhancement cavity : [Kassi et al, Spectrochimica, Spectrochimica Acta Part A 75 (2010) 142–145]
weak b–X transition of 16O_2 in the atmosphere, at 0.1 cm^{-1} resolution

Comb resolution with a Fourier-Transform Spectrometer?



[Rutkowski, L *et al*, J. Quant. Spectrosc. Radiat. Trans 2018, 204, 63–73 (2018)]

The instrumental function is not impacting the adjacent comb modes

subnominal FTS

Resolution of 111 MHz, over 100 cm^{-1} (CO_2 at 1.5μm)
[Maslowski, P. *et al.* Phys. Rev. A, 93, 021802. (2016)]

+ Interleaving
-> precision beyond the Voigt profile

- **Cavity Ring Down - Fourier transform - Comb spectroscopy** -> see talk L1_Talk2 Romain Dubroeucq "Spectroscopie par temps de déclin cavité Fabry Perot par peignes de fréquences »

[Dubroeucq *et al*, Optics Express, vol 3 (2022) + arXiv:2409.09531 (2024)]
[Liang et al, Nature, vol 638 (2025)]

Comb resolution with a Fourier-Transform Spectrometer?
Dual Comb Spectroscopy

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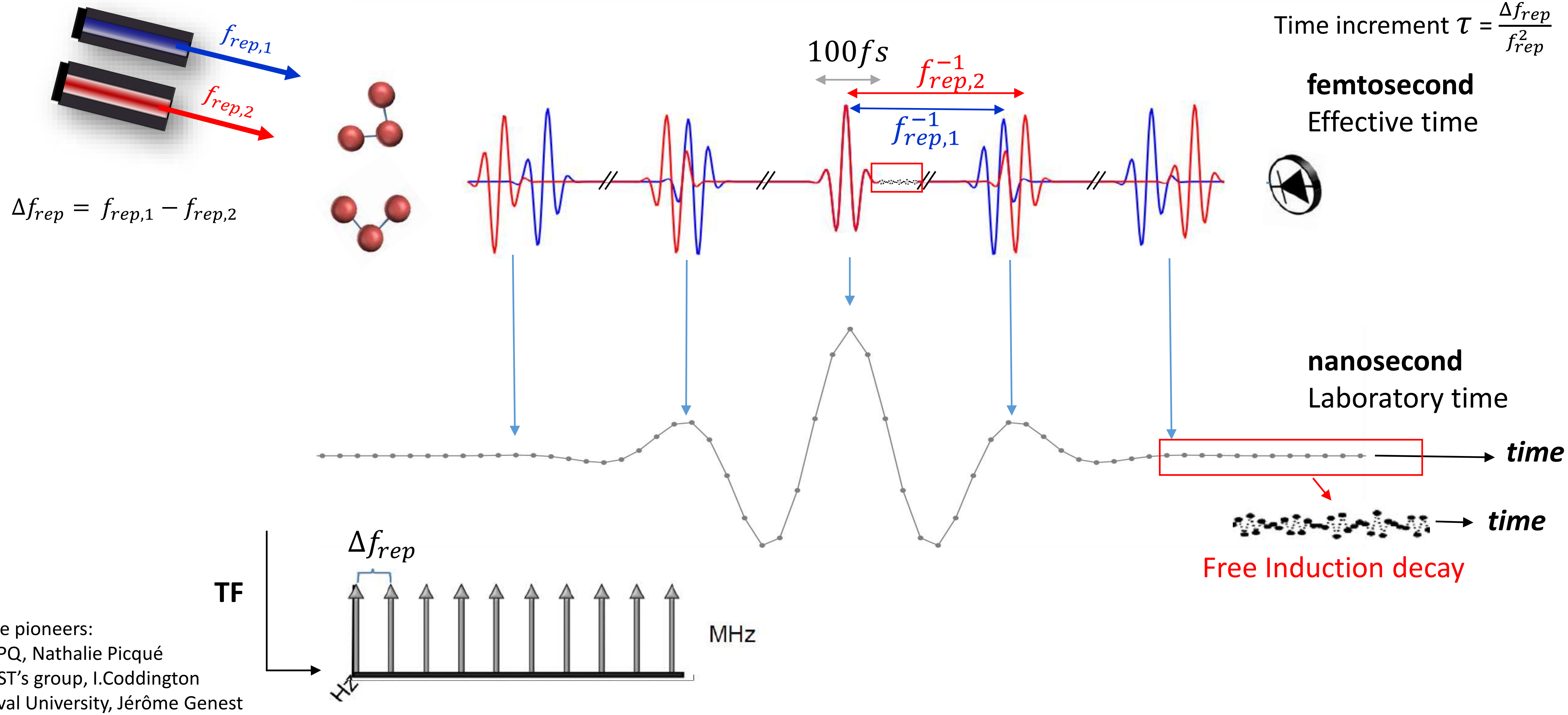
Dual-Comb Spectroscopy – temporal representation

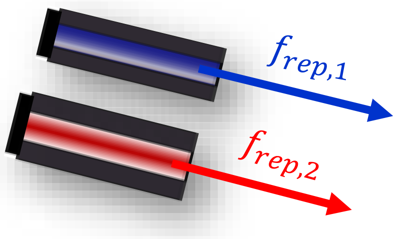
Time increment $\tau = \frac{\Delta f_{rep}}{f_{rep}^2}$



The pioneers:
MPQ, Nathalie Picqué
NIST's group, I.Coddington
Laval University, Jérôme Genest

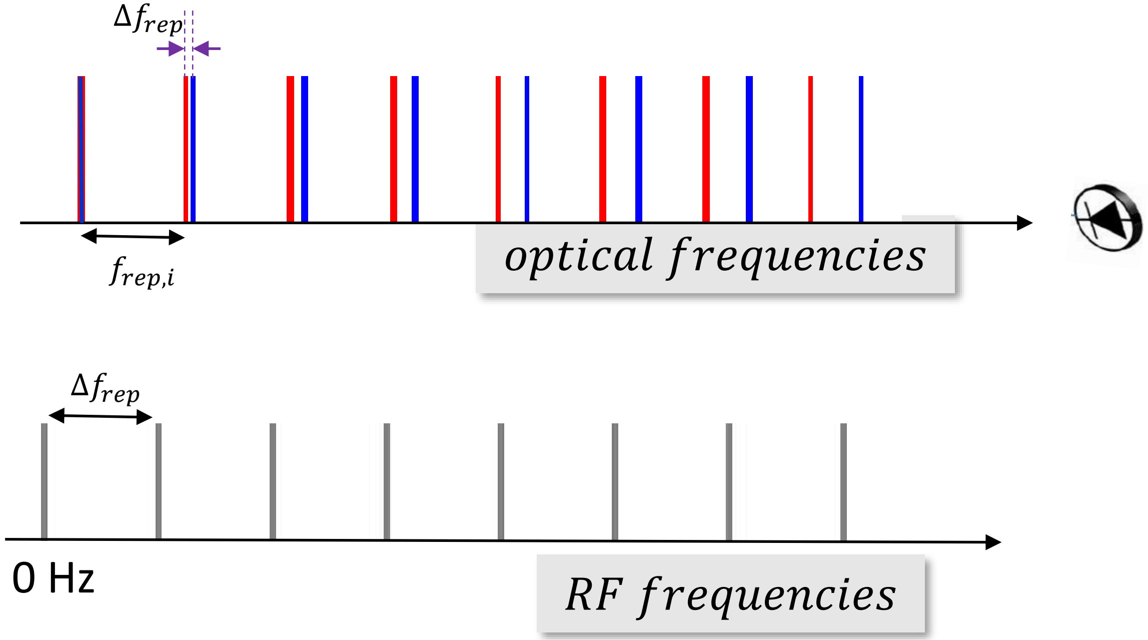
Relative coherence
<1 rad during the acquisition time

$\Delta f_{rep} = f_{rep,1} - f_{rep,2}$



The dual-comb methodology spectral domain

« Or how to map optical frequencies into RF frequencies »



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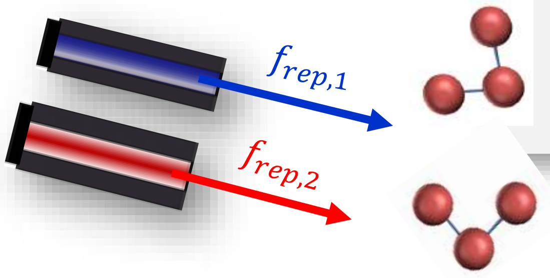
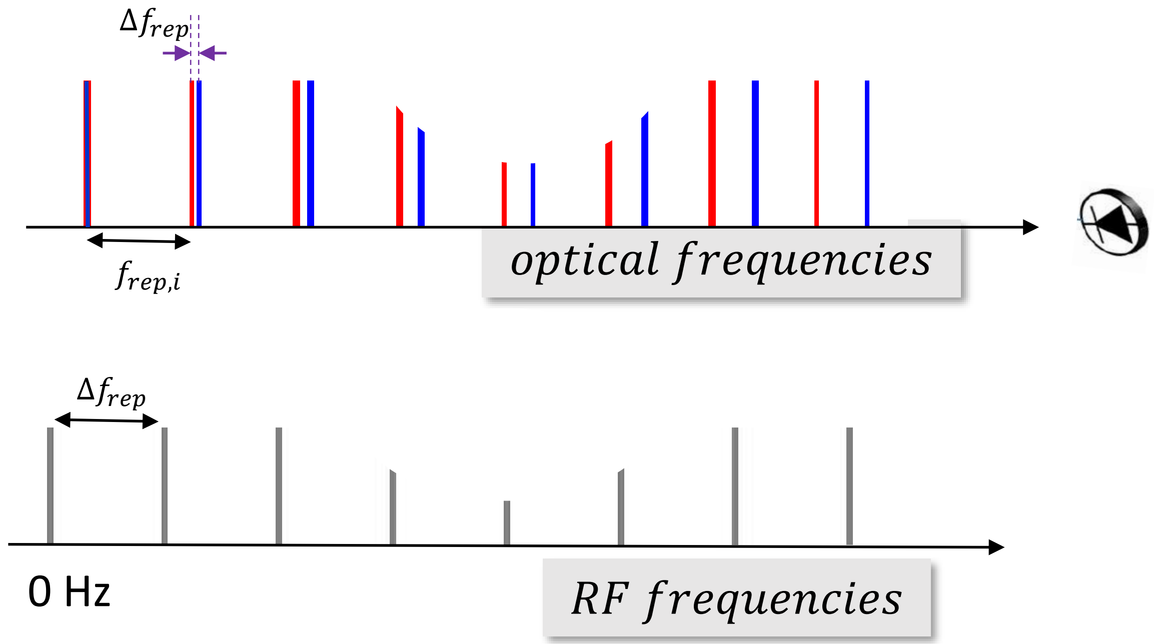
Université Claude Bernard Lyon 1

The dual-comb methodology spectral domain



$\Delta f_{rep} = f_{rep,1} - f_{rep,2}$

« Or how to map optical frequencies into RF frequencies »



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Dual Comb Spectroscopy

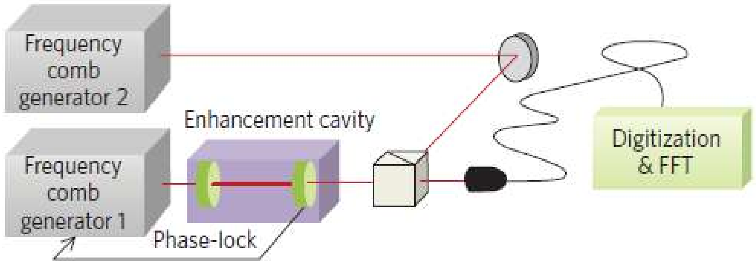
History

2005: Ammoniac detection around $10\mu m$ (2 KLM free-running TiSa lasers + DFG)
[Schliesser and Keilmann, Optics Express Vol 13, 22 (2005)]

2005: 82.6 MHz resolution in the THz domain [Yasui, E. Saneyoshi, and T. Araki, Appl. Phys. Lett. 87, 061101 (2005)]

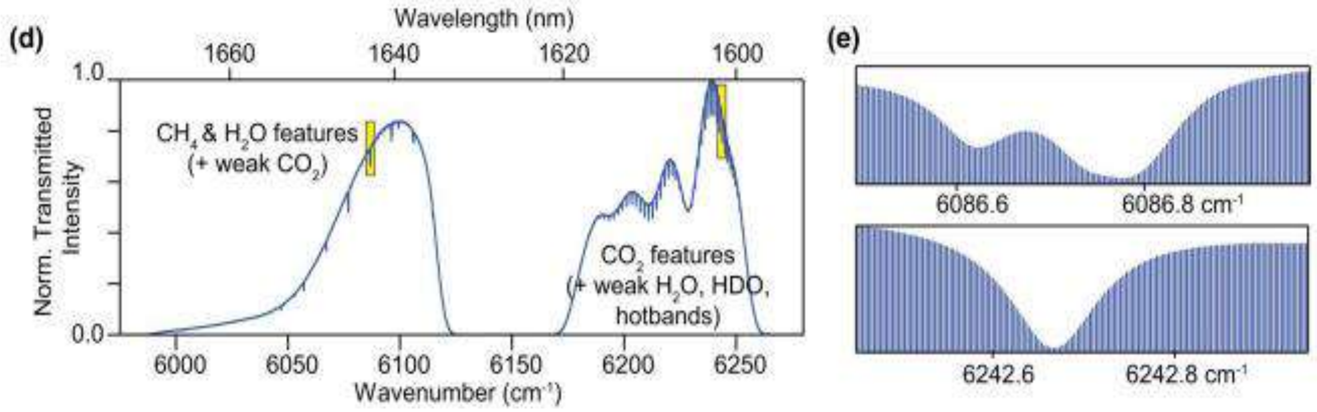
2006: Vater vapor detection in the THz range (2 KLM TiSa lasers + antenna) [Brown et al, Applied Spectroscopy, Vol 60, n°6 (2006)]
Comb resolution in the THz domain: [Yasui *et al*, Appl. Phys. Lett. 88, 241104 (2006)]

2010's: **DCS emerging among the IR spectroscopist community**
[Coddington, Opt. Lett. **34**(14), 2153–2155 (2009)]
[J. Roy, *et al* Opt.Express Vol20, n°20 (2012)] Comb resolved.

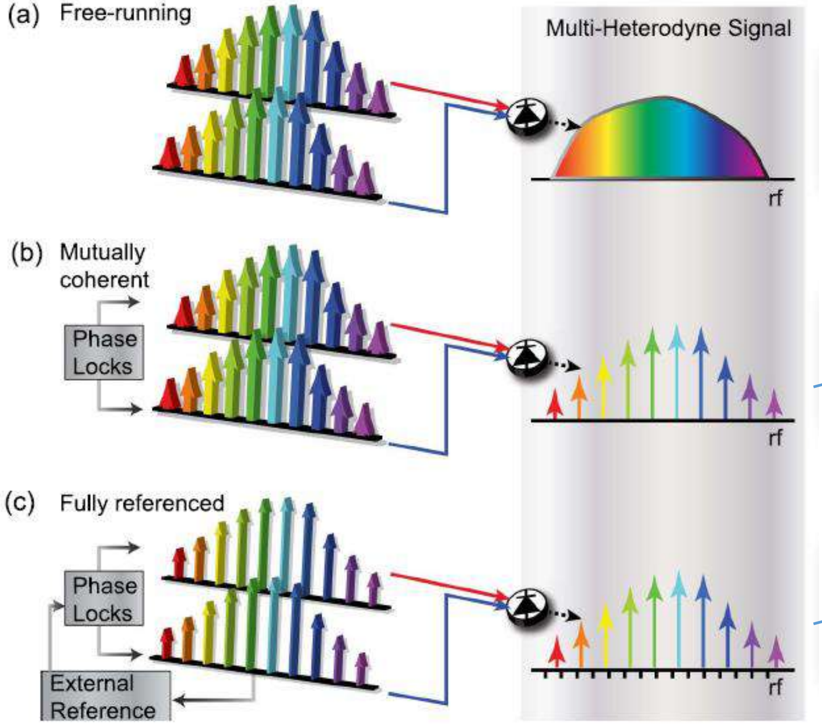


[Bernhardt, B *et al*.. *Nature Photon* **4**, 55–57 (2010).]
4.5GHz resolution, 20nm bandwidth, Yb laser
Acétylène C2H2 and Ammoniac NH3

2014 -> outside the laboratory
[Rieker et al, Optica, Vol 1, issue 5 (2014)]:



Dual Comb Spectroscopy
Architectures and Applications



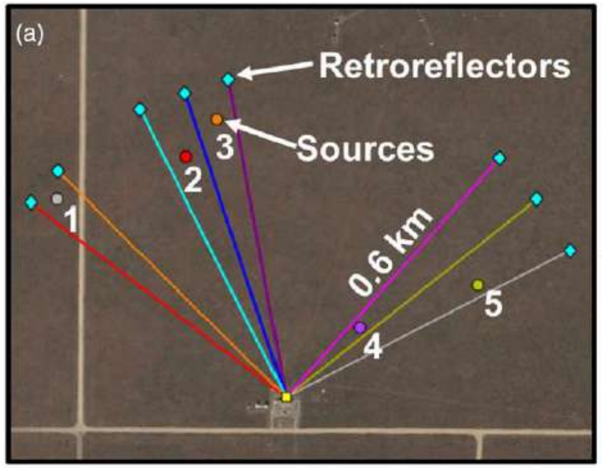
[from review : Coddington et al. Optica Vol. 3, No. 4 (2016)]

Typically GHz resolution
+ phase correction algorithms to retrieve the comb structure
[Walsh *et al* J. Phys. B: At. Mol. Opt. Phys. **58** (2025)]

Typically GHz to MHz resolution
+ phase correction
Phase-locked or mutually coherent by the laser architecture *(ex. Part III)*

Sub-MHz resolution -> **Spectral interleaving possible**
Systematic error of **200kHz to 300kHz, at 3.4 μm and 1.5μm** (Méthane)
[Zolot et al, JQSRT, 118, 26-39 (2013)]
-> HITRAN data base improvement
Systematic error of **600kHz at 7.8 µm** (38.4 THz) over 1.2THz (N2O)
[Komagata et al, Physical Review Research **5**, 013047 (2023)]
-> HITRAN data base improvement

Long-path absorption LIDAR

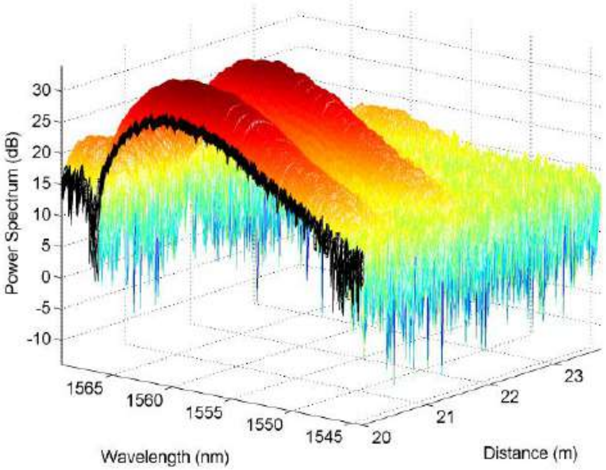


Methan leak detection [Coburn *et al,* Optica Vol. 5, No. 4 (2018)]
Simulatenous **CH_4, NH_3, CO_2, and H_2O**
NH_3 : subppm.m and CH_4: ppm.m [Herman2021]

Retroreflection on a natural target (forest):
[W. Patiño Rosas and N. Cézard, Opt. Express **32**, 13614-13627 (2024)]

+ Isotopic ratios

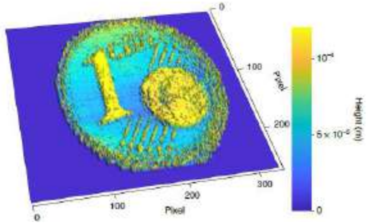
Hyperspectral - LIDAR



HCN absorption line
From the backscattered light of water-vapor dropled

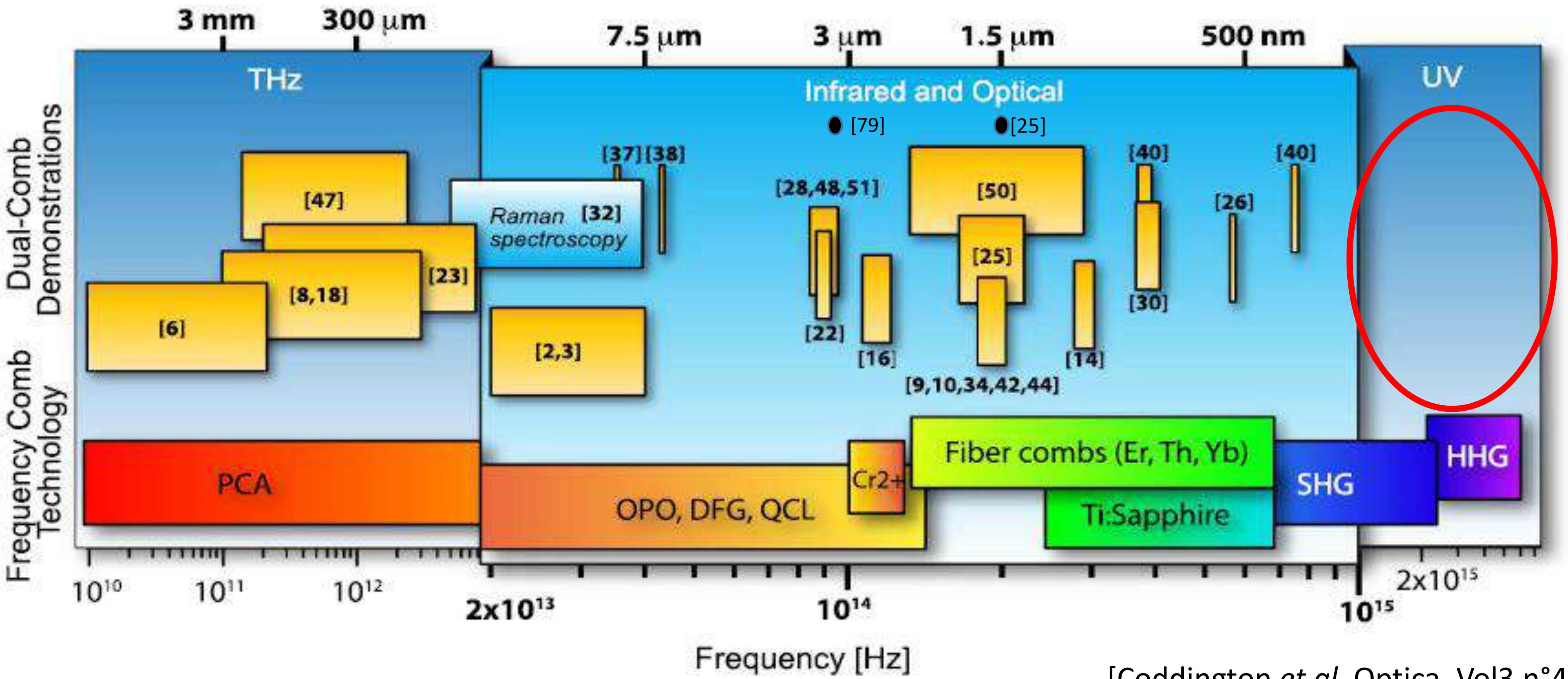
[Boudreau, Optics Express, Vol. 21, No. 6 (2013)]

Hyperspectral - IMAGING



[Vicentini, Nature Photonics, vol15, 890 (2021)]

Dual Comb Spectroscopy – Spectral ranges



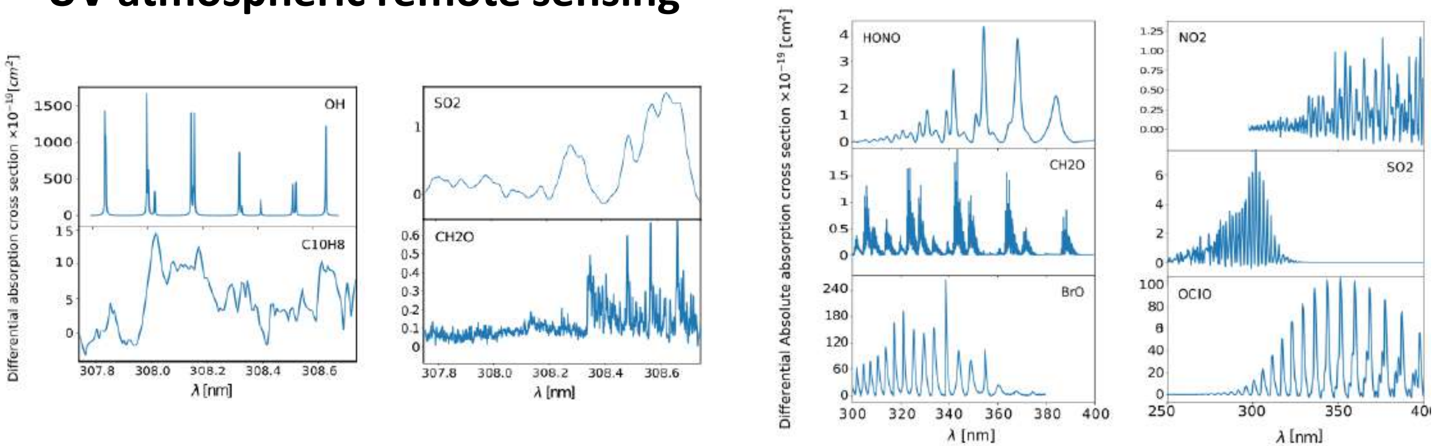
[Coddington *et al,* Optica, Vol3 n°4 (2016)]

⬬ EOM

+ Microresonnators (chip-based): 1.5 μm

DCS in the UV and D-UV range – Motivations?

UV atmospheric remote sensing



High spectral resolution to explore the full dynamic of fundamental transient effects
(combustion, chemical reaction dynamics at the microsecond time scale)

Absorption line metrology in the UV
broadband and high resolution spectroscopy -> UV congestionned lines

"table-top" synchrotron beamline?

SOLEIL SYNCHROTRON

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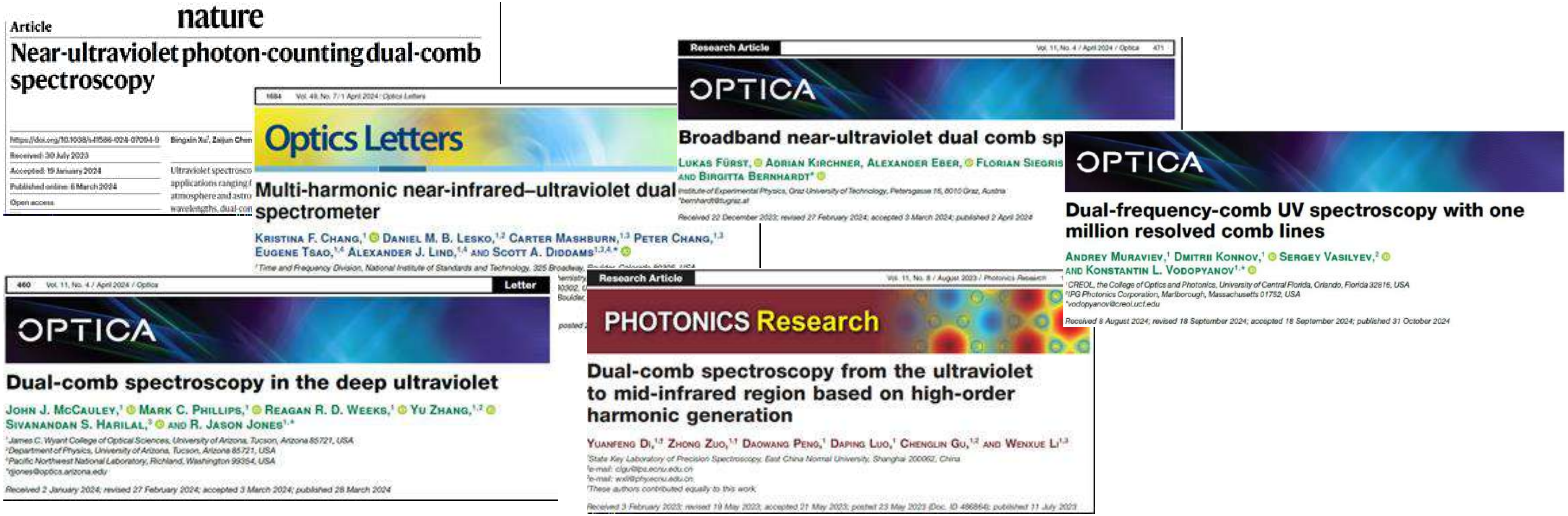
UV – Dual Comb: State of the art

R. Jason Jones’ group in Arizona [McCauley *et al*, Optica, Vol. 11, No. 4, April 2024] (2 Yb fiber comb **+ 3rd harmonic** - FeI and FeII absorption lines – 2THz wide)
N. Picqué’s group at MPQ-Garching [Xu *et al*, Nature, vol°627, 2024] (Erb fiber + EOM combs +**2X SHG** - Cesium – 50GHz wide)
S. Diddams’ group in Colorado [Chang *et al*, Optics Letters, vol°49, 7, 2024] (Erb fiber comb **+ PPNL for 5th harmonic**)
L. Vodopyanov's group in Florida [Muraviev et al, Optica, vol 11, 11 2024] (2 KLM laser + **HHG, 7 harmonic**)
B. Bernhardt’s group in Graz [Fürst *et al,* Optica vol n°11, 2024] (2 Yb sources in one, **THG + broadening**, 50GHz resolution, 37.5 THz wide)


Article nature
Near-ultraviolet photon-counting dual-comb spectroscopy
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Open access

1684 Vol. 49, No. 7 / 1 April 2024 / Optics Letters
Optics Letters
Multi-harmonic near-infrared–ultraviolet dual spectrometer
Kristina F. Chang,[1] Daniel M. B. Lesko,[1,2] Carter Mashburn,[1,3] Peter Chang,[1,3] Eugene Tsao,[1,4] Alexander J. Lind,[1,4] and Scott A. Diddams[1,3,4,*]

Research Article Vol. 11, No. 4 / April 2024 / Optica 471
OPTICA
Broadband near-ultraviolet dual comb sp
Lukas Fürst, Adrian Kirchner, Alexander Eber, Florian Siegris and Birgitta Bernhardt*
Institute of Experimental Physics, Graz University of Technology, Petersgasse 16, 8010 Graz, Austria
Received 22 December 2023; revised 27 February 2024; accepted 3 March 2024; published 2 April 2024

OPTICA
Dual-frequency-comb UV spectroscopy with one million resolved comb lines
Andrey Muraviev,[1] Dmitrii Konnov,[1] Sergey Vasilyev,[2] and Konstantin L. Vodopyanov[1,*]
[1]CREOL, the College of Optics and Photonics, University of Central Florida, Orlando, Florida 32816, USA
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*vodopyanov@creol.ucf.edu
Received 8 August 2024; revised 18 September 2024; accepted 18 September 2024; published 31 October 2024

460 Vol. 11, No. 4 / April 2024 / Optica Letter
OPTICA
Dual-comb spectroscopy in the deep ultraviolet
John J. McCauley,[1] Mark C. Phillips,[1] Reagan R. D. Weeks,[1] Yu Zhang,[1,2] Sivanandan S. Harilal,[3] and R. Jason Jones[1,*]
[1]James C. Wyant College of Optical Sciences, University of Arizona, Tucson, Arizona 85721, USA
[2]Department of Physics, University of Arizona, Tucson, Arizona 85721, USA
[3]Pacific Northwest National Laboratory, Richland, Washington 99354, USA
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Research Article Vol. 11, No. 8 / August 2023 / Photonics Research
PHOTONICS Research
Dual-comb spectroscopy from the ultraviolet to mid-infrared region based on high-order harmonic generation
Yuanfeng Di,[1,†] Zhong Zuo,[1,†] Daowang Peng,[1] Daping Luo,[1] Chenglin Gu,[1,2] and Wenxue Li[1,3]

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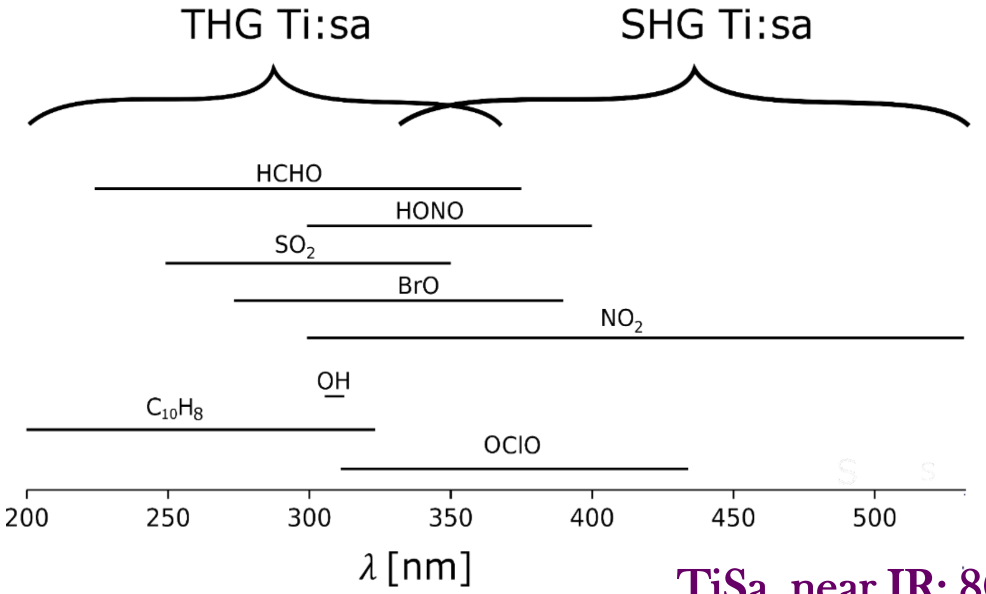
UV-DCS, with a … TiSa laser !

- Sufficiently low optical phase noise 0.11 rad (1 kHz : 50 MHz) [Sutyrin et al, Optical Engineering, vol n°12, 2014]
  + Low harmonics generation (SHG - THG) to reach the UV range
    -> **coherent UV-comb pulses**

- Sufficient power in the UV range (**>10 mW**) -> **OK remote sensing**

- Broad and versatile spectral range (**>10nm**)
  -> multispecies monitoring

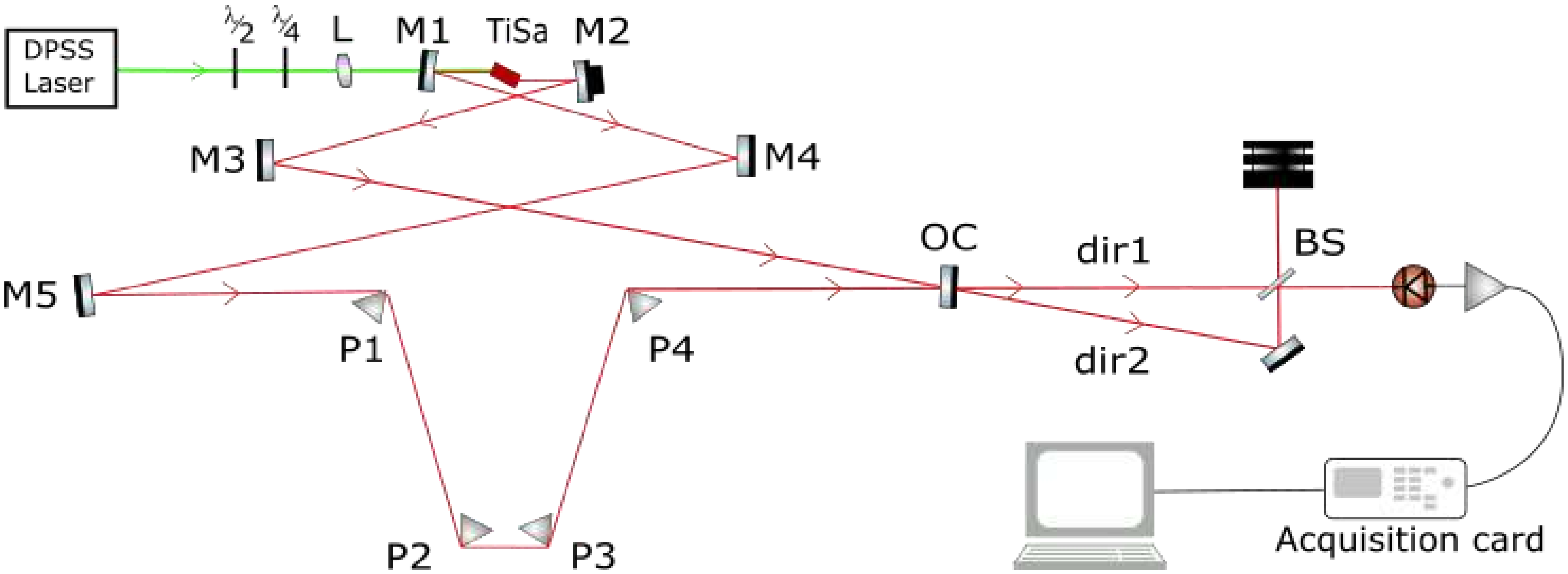


TiSa, near IR: 800nm

[S. Galtier, C. Pivard, P. Rairoux. Remote Sensing, 12(20), 2020]

Home-made bidirectional ring laser cavity

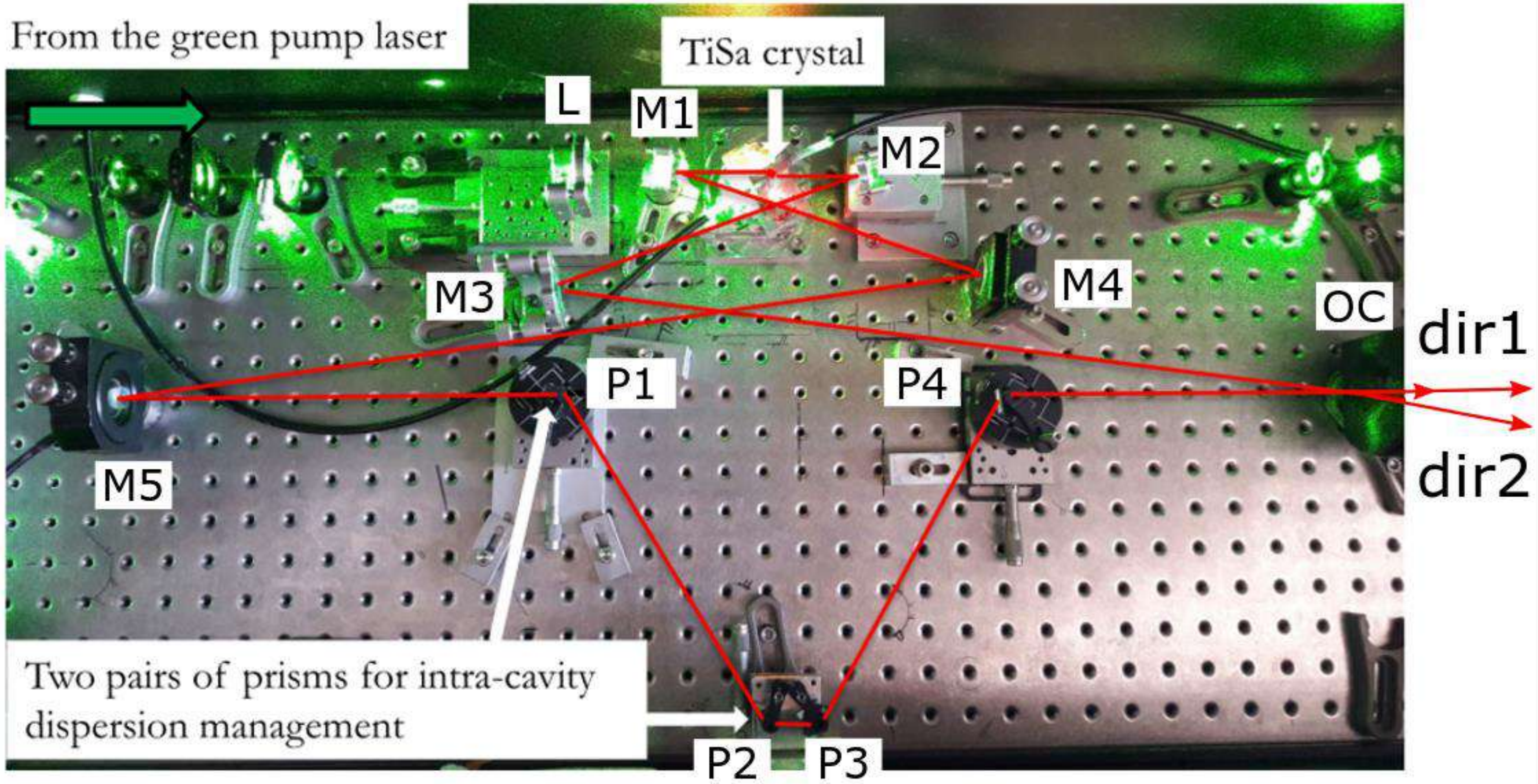
-> intrinsic mutual coherence



OC: output coupler
BS: beam splitter
P1-4: prisms for dispersion compensation

[Ideguchi *et al*, Optica, vol 3 n°7 ,2016] , [Garduno-Mejia *et al*, Optic Comm, vol n°171, 1999], [Bartels *et al*, Opt. Lett., vol n°24, 1999], [Fortier *et al*, Opt. Lett., vol n°31, 2006], [Pelouch *et al*, Opt. Lett., vol n°17, 1992],

Home-made bidirectional ring laser cavity



Clément Pivard, Dr

[Clément Pivard. *Development of a bidirectional Dual Comb laser source towards atmospheric trace gases monitoring*. PhD thesis, Université Lyon 1 Claude Bernard - Institut Lumière Matière (ILM), defense date: december, 15, 2022.]

Δf_{rep} ?? Asynchronous operation?

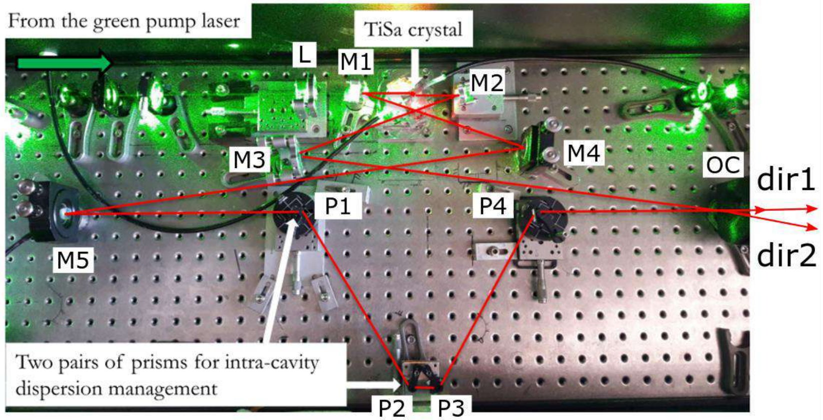
How to variate Δf_{rep} ?

Crystal translation (µm) / Lens translation (µm) / Prism translation (µm) / Mirror adjustment (µm)

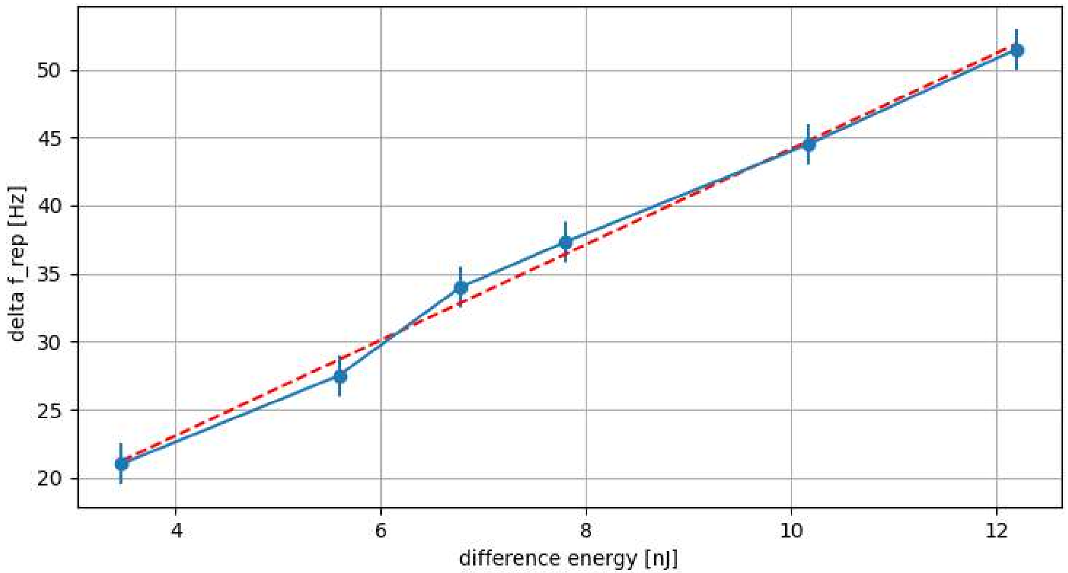
Source of Δf_{rep} ?

Correction of the Non-linear Schrodinger equation :

Self-steepening effect: group velocity dependance as function of pulse duration and energy



Predicted group velocity change: $\approx 0.1(2) fs/nJ$ [Sander *et al*, Optics Express Vol 18 n°5 (2010)]



Measurement Δf_{rep} versus differential intracavity energy on the ILM-BDTiSa

Coef_dir = 0.25(11) fs/nJ

✓

Behaviour as function of Δf_{rep}

How to variate Δf_{rep} ?

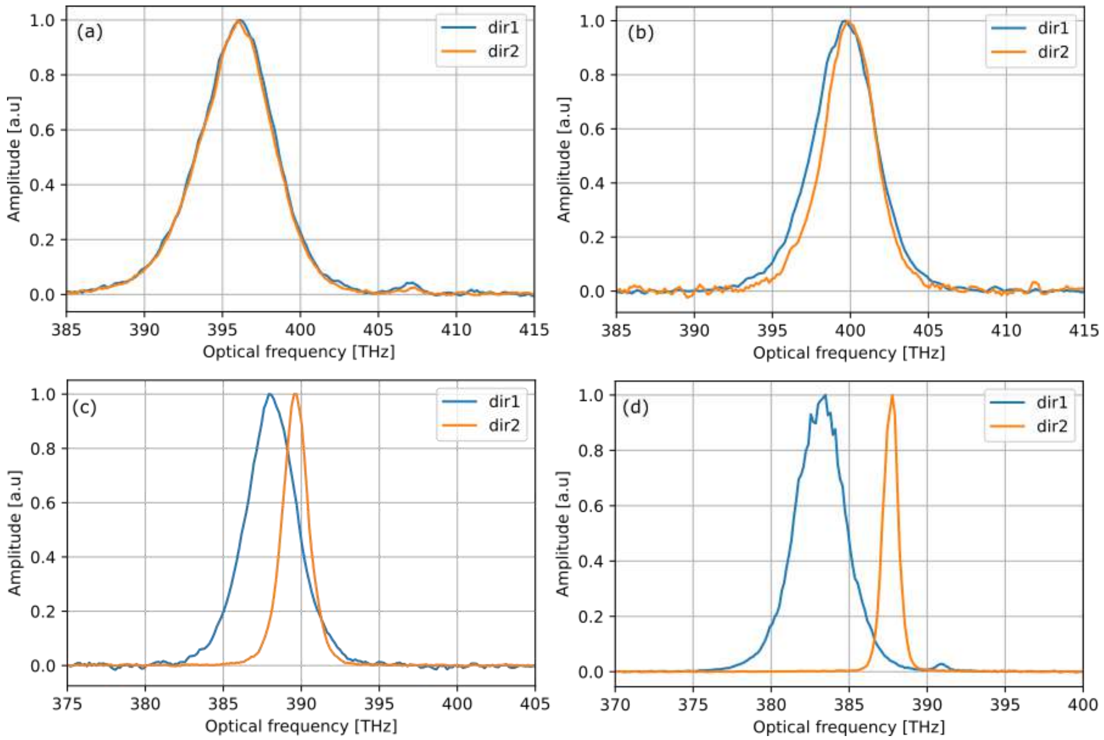
Crystal translation (µm) / Lens translation (µm) / Prism translation (µm) / Mirror adjustment (µm)

$\Delta f_{rep} = 1.2\ Hz$

$\Delta f_{rep} = 10.4\ Hz$

$\Delta f_{rep} = 107\ Hz$

$\Delta f_{rep} = 425\ Hz$

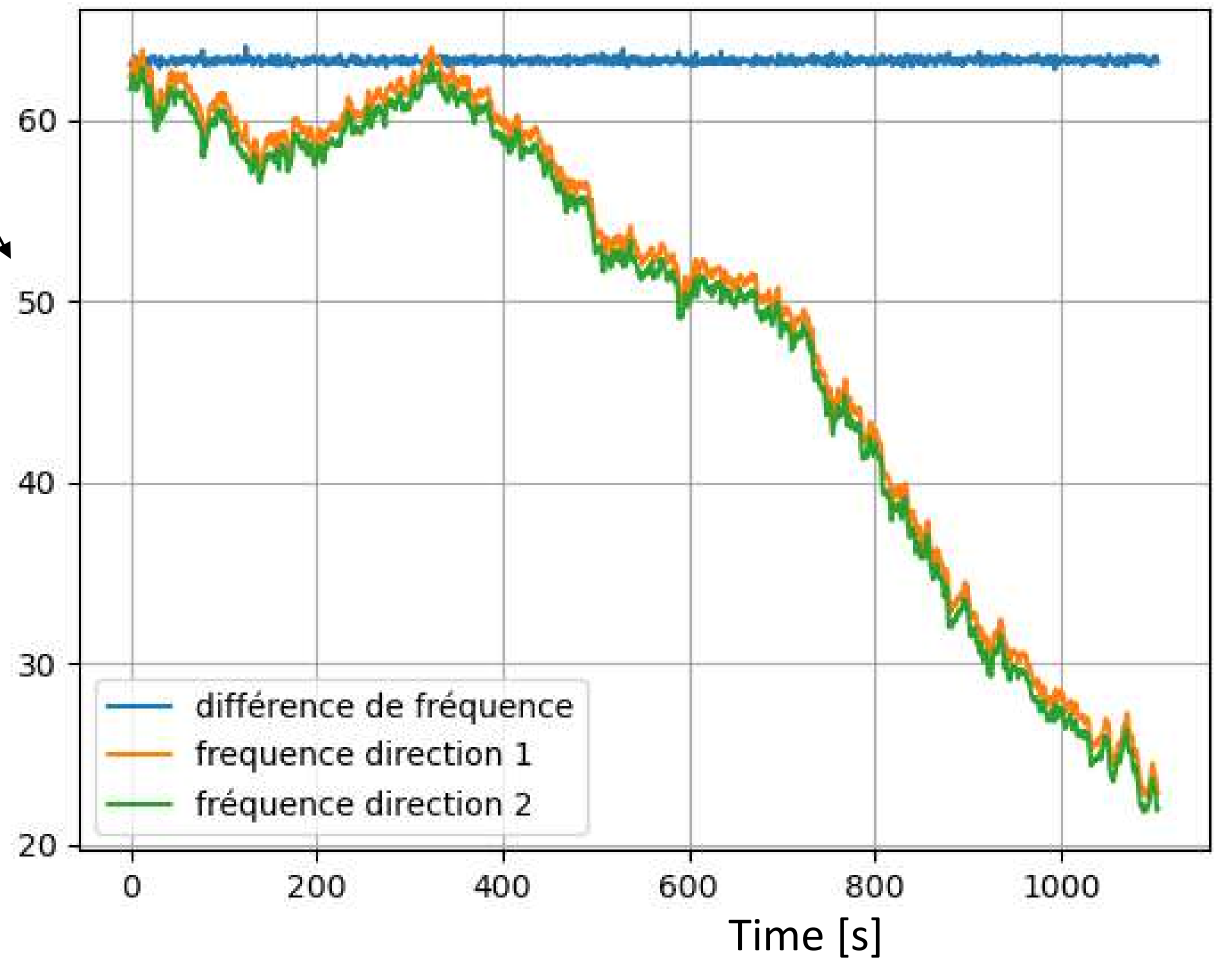


[S. Galtier, C. Pivard, J. Morville and P. Rairoux '*High-resolution dual comb spectroscopy using a free-running, bidirectional ring titanium sapphire laser.*' Opt. Express, **30** n°12, 21148-21158 (2022)]

Mutual coherence of f_{rep1} and f_{rep2}

f_{rep1}, f_{rep2} and Δf_{rep} measurements using a frequency counter FCA3000 Tektronix™

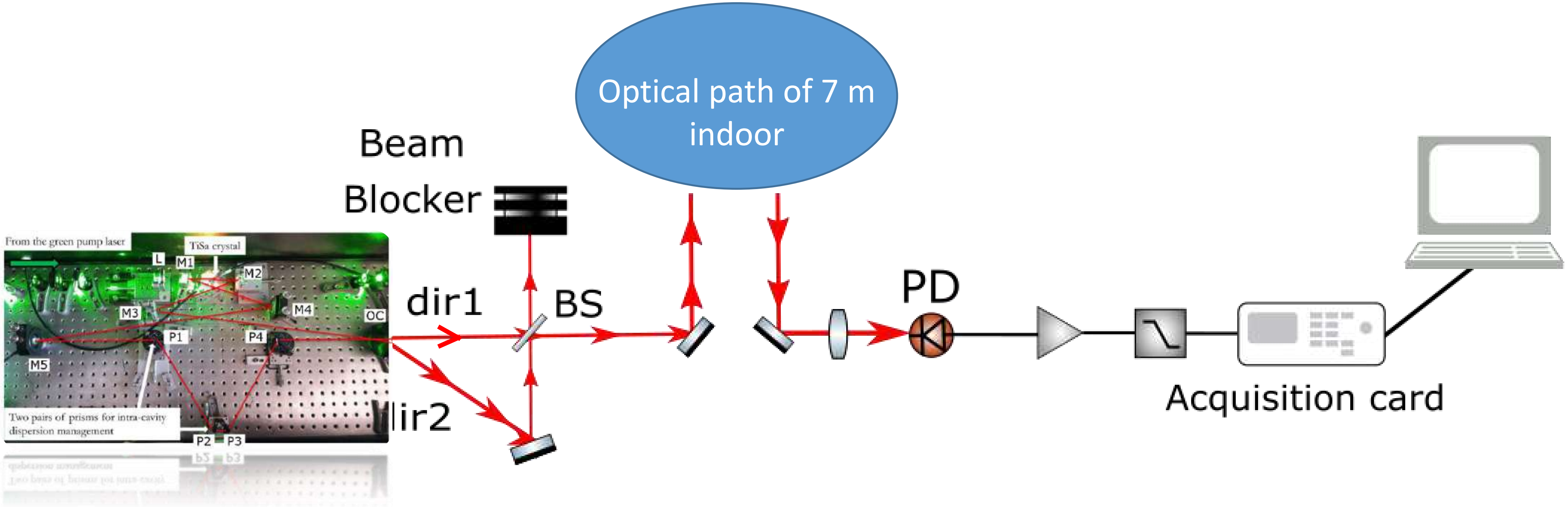
Frequency [Hz] -118.504 300 MHz



Δf_{rep}= 0.89 +/- 0.14 Hz

[Armand Veau M1 internship 2022]

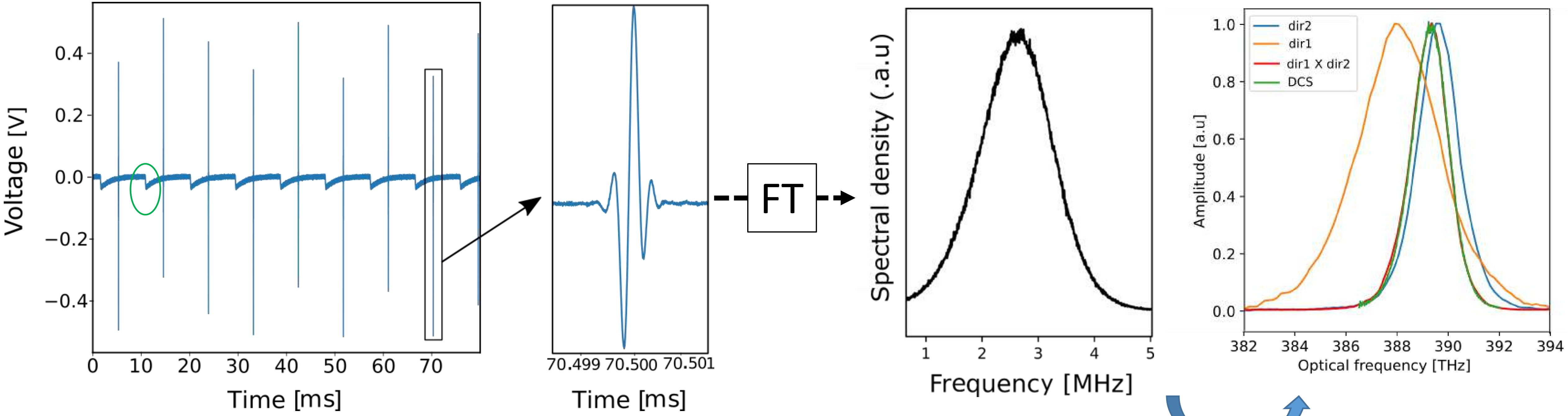
Di-oxygene DCS measurement



Schematic setup for O_2 DCS spectroscopy

DCS signal: temporal trace and retrieved spectrum

DCS temporal trace | Centerburst | RF spectrum | IR spectra

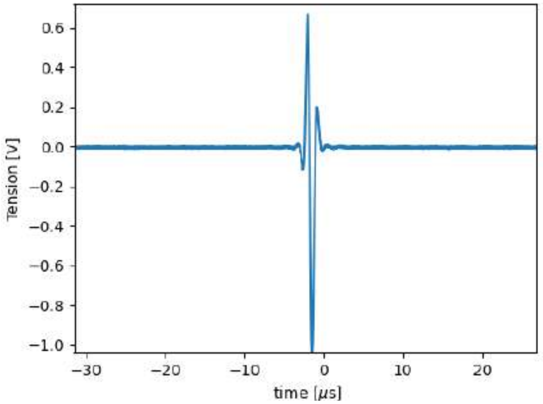


Gain depletion

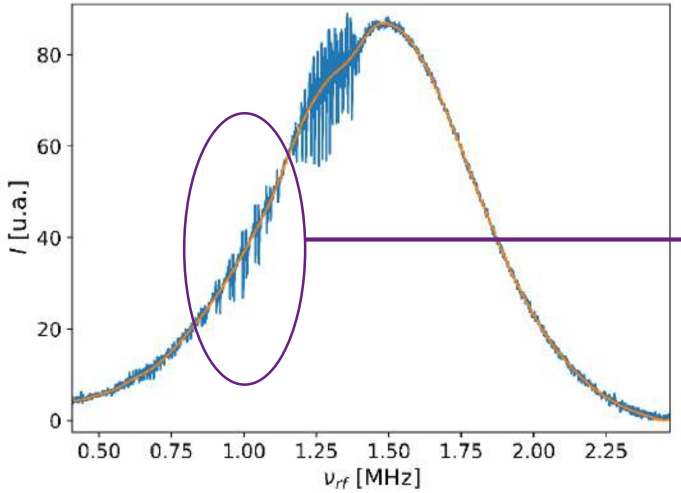
RF frequencies towards optical frequencies

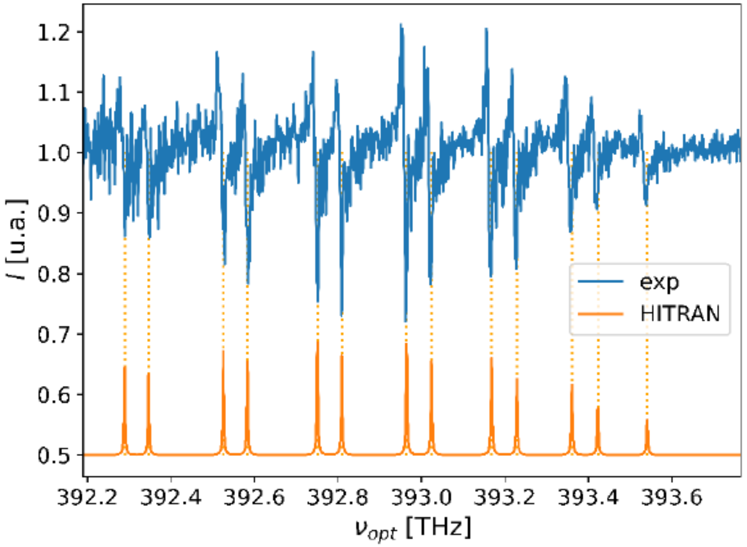
$$\nu_{opt} \approx \frac{\boldsymbol{f_{rep}}}{\boldsymbol{\Delta f_{rep}}} \times \left(k f_{rep} \pm (\nu_{RF} - \boldsymbol{\Delta f_{CEO}}) \right)$$

Di-oxygene DCS measurement

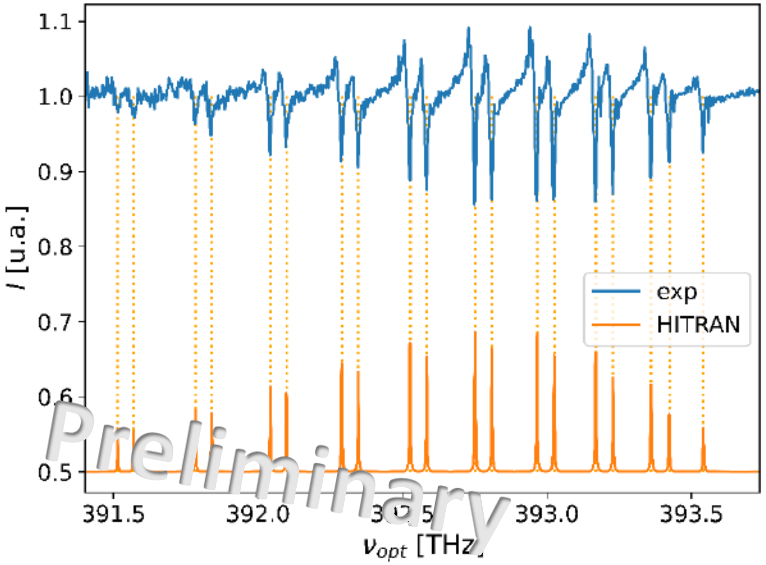


TF





One-shot measurement - **3 ms acquisition time**
Instrumental line-width: 0.7 GHz in the optical domain
sensitivity per spectral element at 1 second ≈ $10^7\ /\sqrt{Hz}$



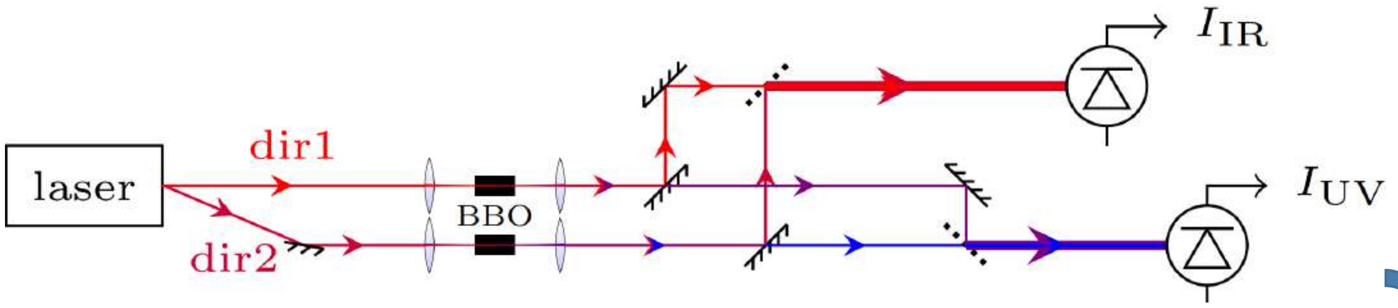
$b^1\Sigma_g^+ - X_3\Sigma_g^-$ electronic transitions of oxygen with ambient linewidth of 1.5GHz

→ GHz resolution level

→ Asymmetric lineshape under investigation

[S. Galtier, C. Pivard, J. Morville and P. Rairoux '*High-resolution dual comb spectroscopy using a free-running, bidirectional ring titanium sapphire laser*.' Opt. Express, **30** n°12, 21148-21158 (2022)]

Firsts results on UV -DCS

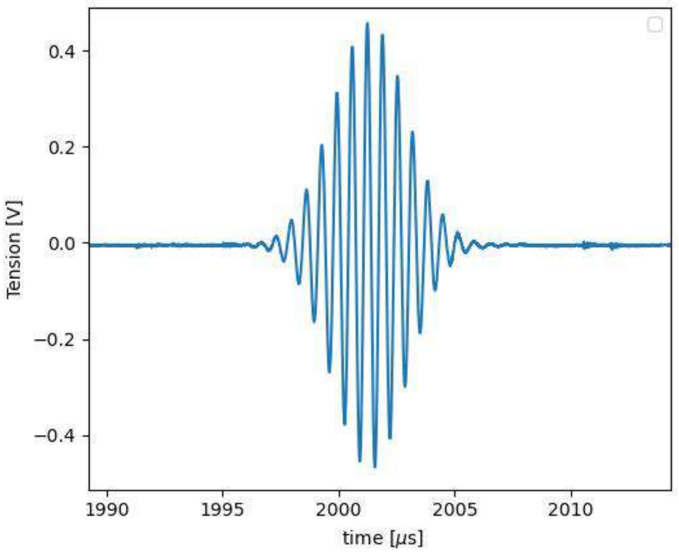


The principle

Abel Feuvrier (PhD)

BBO crystal
 – type I phase matching - 1mm thick
8% to 20% SHG efficiency
Power: **10 to 80 mW at 400nm**

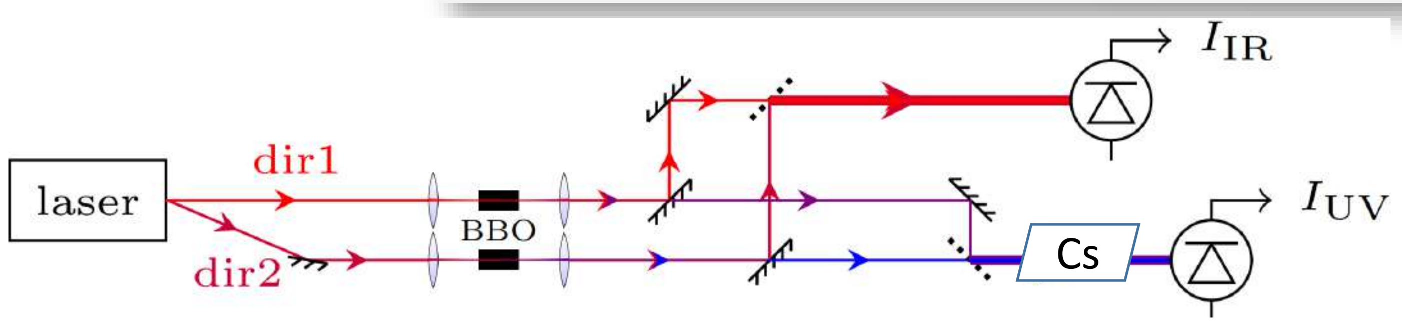
-> **UV-IGM - Contraste 66%**



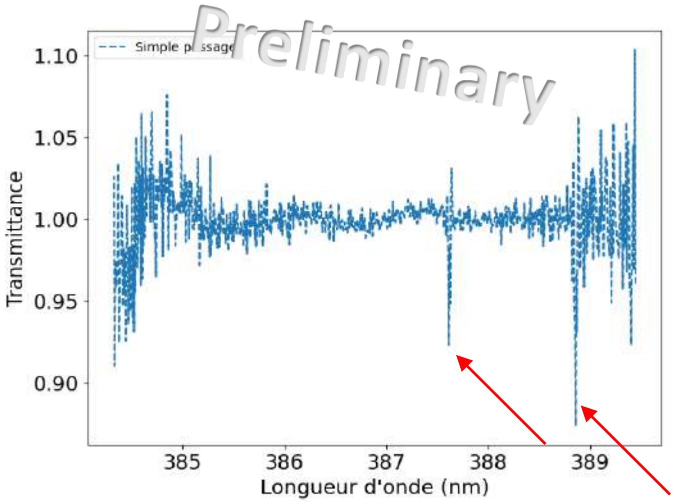
IGM @390nm

[Abel Feuvrier *et al* –EOSAM October 2023 - EPJ Web of Conferences **287**, 07019 (2023)]
ANR UV-ATMOCOS

Cs measurement



Schematic setup using a Cs vapor cell



Retrieved UV spectra from FFT of a 2.5ms window around the IGM centerburst.
Singe shot interferogram.

2 resonances of ^{133}Cs - atomic linewidth ≈ 1GHz.
$6S_{1/2}$–$8P_{3/2}$ @ 387.615 nm
$6S_{1/2}$–$8P_{1/2}$ @ 388.861 nm
-> Cs lines to assess the resolution of the spectrometer ?

-> Towards quantitative transmission measurement using a NO2 spectral lines in a vacuum gas chamber.

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Conclusion and perspectives
Frequency comb spectroscopy

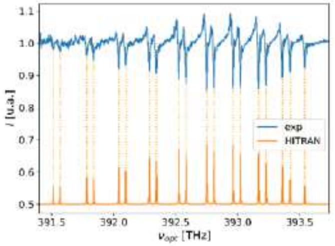
- **Frequency comb spectroscopy**

  Variety of spectrometers: Speed - accuracy/precision - Spectral coverage

- **Dual Comb in the UV range:**

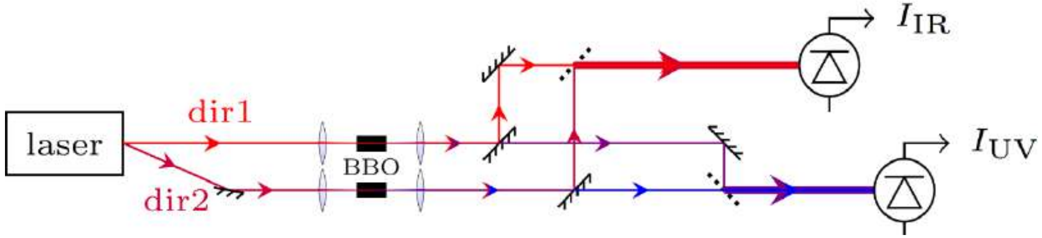
The free-running bidirectional KLM TiSa laser at ILM

- Δf_{rep} observed from 0 to 450 Hz – measurement of **resolved molecular lines.**
- UV Spectroscopic measurements with ≈ 10mW total power



- **How to improve the sensitivity:**

Averaging protocols: a posteriori (computing) or real-time correction protocols.
Address the asymmetrical line shapes.



- **Field-deployable instrument ?**

- Frequency comb spectroscopy at other spectral ranges ?

  The case of Ramsey-Comb spectroscopy (XUV)

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Remerciements

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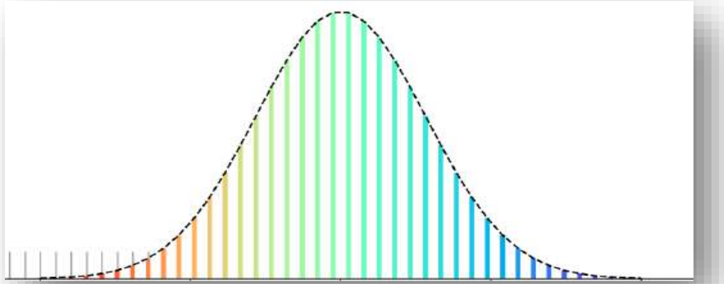
Abel Feuvrier (PhD), Dat Nguyen(PostDoc),
Sandrine Galtier and Patrick Rairoux.

Abel Feuvrier (PhD) **Dat Nguyen (PostDoc)**



Merci à vous pour votre attention!

Collaborators:
Jérôme Morville (ILM),
Jesus GardunoMejia and Catalina Karolina (Institute of New Mexico)

Thanks to the ATMOS team of ILM