JSM2025

anr© METEORIT

# Spectroscopie moléculaire THz haute précision par photo-mélange à 1550 nm

**Loïc LECHEVALLIER**, Samir KASSI, LIPhy, Grenoble, France

UGA Université Grenoble Alpes

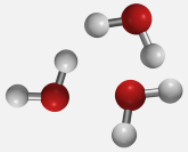
LIPhy

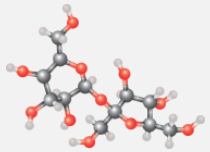
cnrs

# THz domain

## *Gas phase spectroscopy*

**Rotational transition of light molecules**





**Ro-vibration of heavy molecules**

**Hyperfine structures**

**Line profile**

**Doppler broadening very weak compare to IR**

= good spectral signature

## *Astrophysics [1][2]*

**Probing molecular clouds**

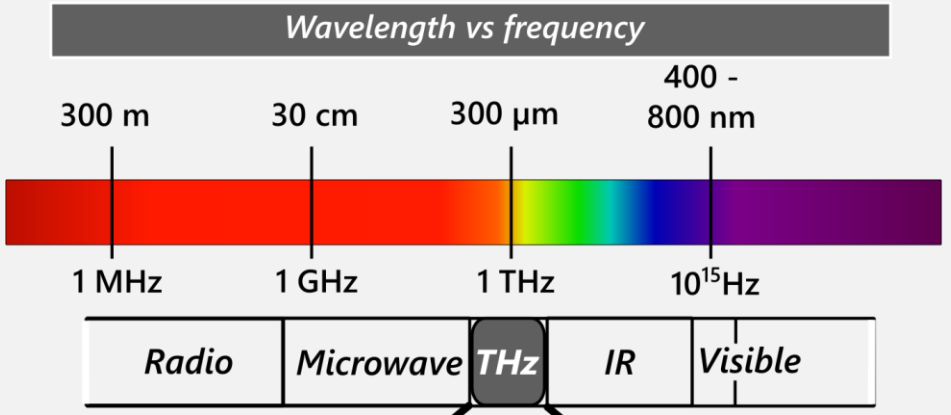
**Composition planetary atmosphere**

## *Atmospheric measurement [3]*

**Traces detection**


[1] V. V. Ilyushin *et al.*, « Rotational and rovibrational spectroscopy of CD $_3$ OH with an account of CD $_3$ OH toward IRAS 16293−2422 », *Astron. Astrophys.*, vol. 658, p. A127, févr. 2022, doi: 10.1051/0004-6361/202142326.
[2] A. D'Arco *et al.*, « Terahertz continuous wave spectroscopy: a portable advanced method for atmospheric gas sensing », *Opt. Express*, vol. 30, nº 11, p. 19005, mai 2022, doi: 10.1364/OE.456022.
[3] A. Cuisset *et al.*, « Terahertz Rotational Spectroscopy of Greenhouse Gases Using Long Interaction Path-Lengths », *Appl. Sci.*, vol. 11, nº 3, p. 1229, janv. 2021, doi: 10.3390/app11031229.

# THz generation




**Multiplication chain**

**Difference frequency**

*0.1 - 10 THz*

**3 – 330 cm$^{-1}$**

Laser 1

Laser 2

**Photomixer**

**source development**

**Optic**

**Telecom wavelength**

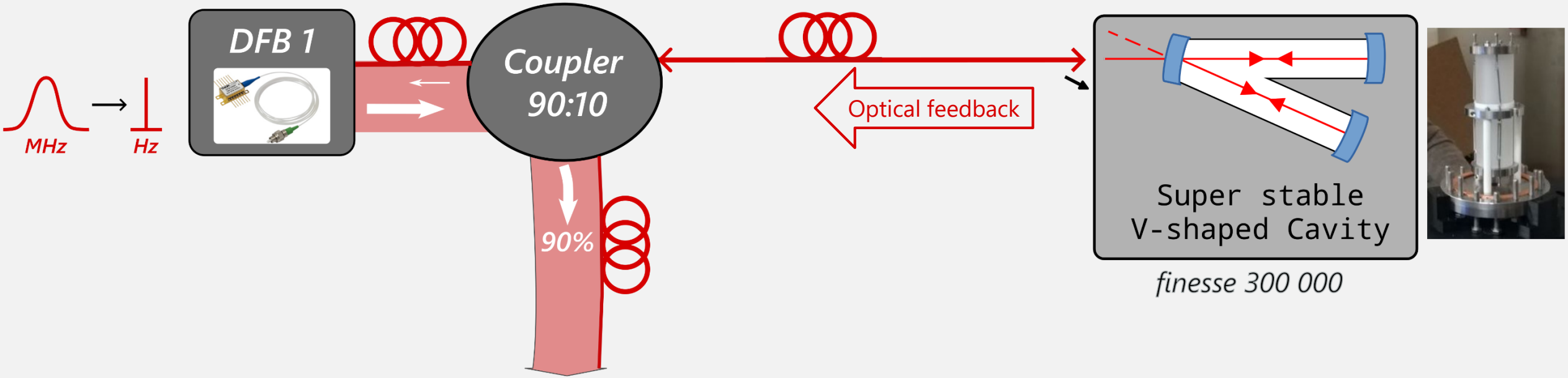
**190-195 THz**

*Easily tunable by changing temperature or current*

# Optical feedback technique with DFB laser

[1] P. Laurent, A. Clairon and C. Breant, "Frequency noise analysis of optically self-locked diode lasers," in *IEEE Journal of Quantum Electronics*, vol. 25, no. 6, pp. 1131-1142, June 1989

Reduction in the emission linewidth of
**D**istributed **F**eed **B**ack laser from MHz to **Hz** level



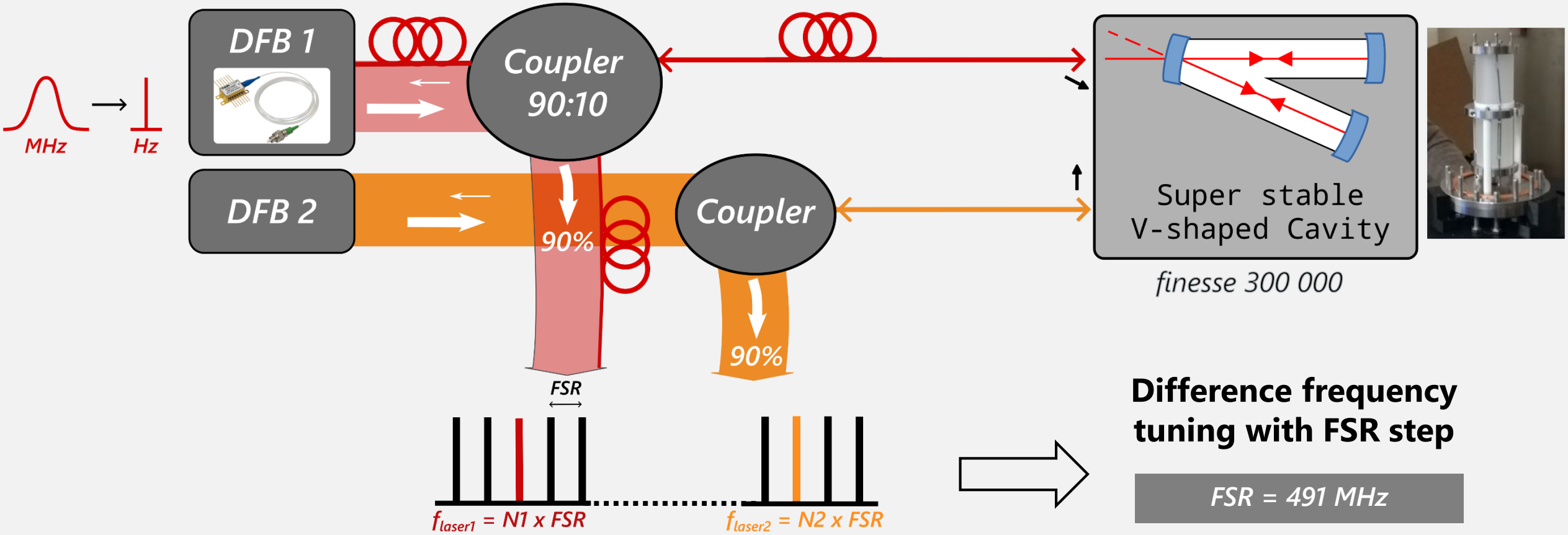

**Optical fiber**
@telecom wavelength

Powerful
output

Copy the **cavity stability**
**& Hz level** linewidth

Drift
**~10 Hz/s**
**@192 THz**

# Optical feedback technique with DFB laser : two lasers



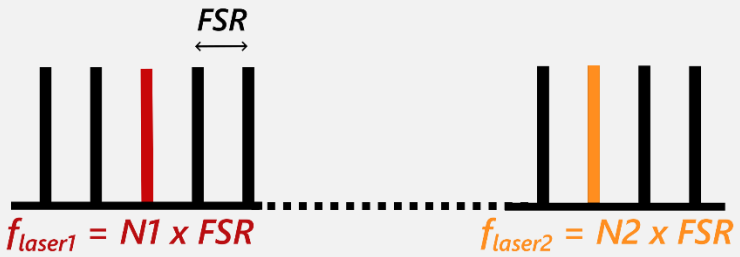

**Difference frequency tuning with FSR step**

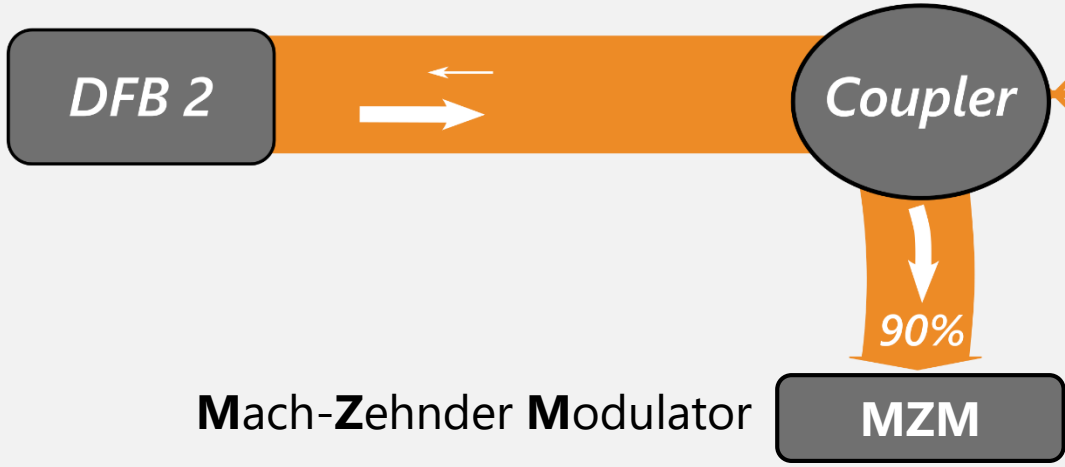
*FSR = 491 MHz*

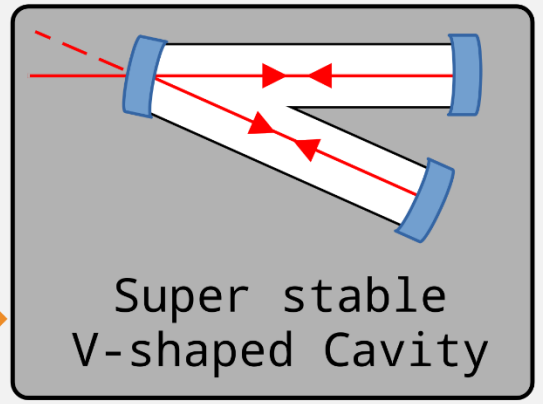
# Fine tuning for spectroscopy

**Frequency step**

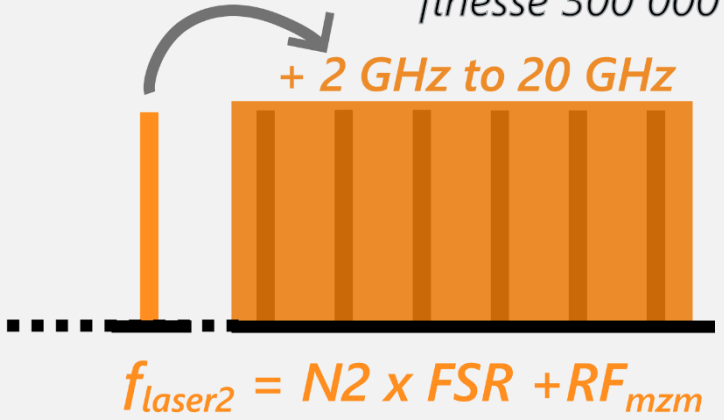
*FSR = 491 MHz*



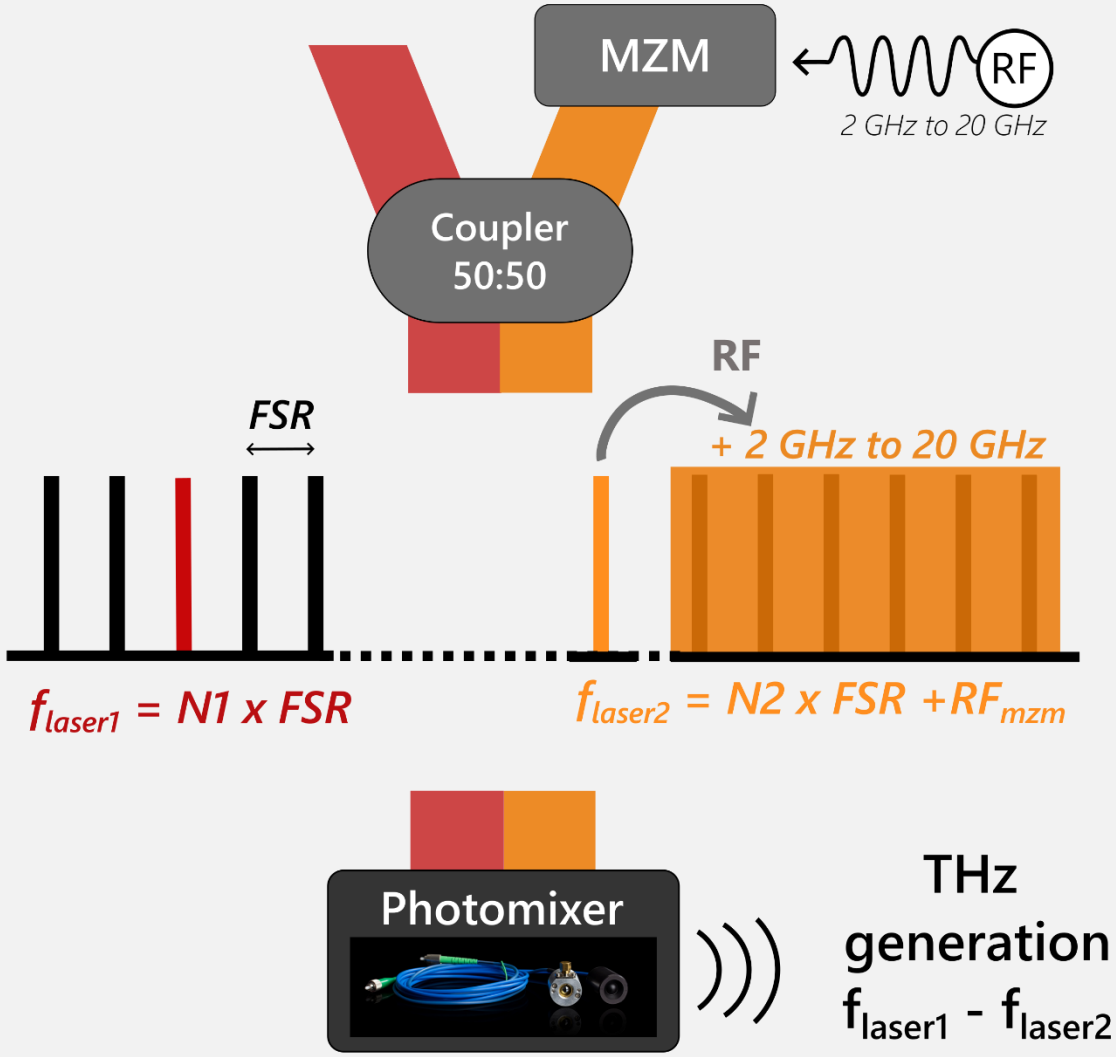



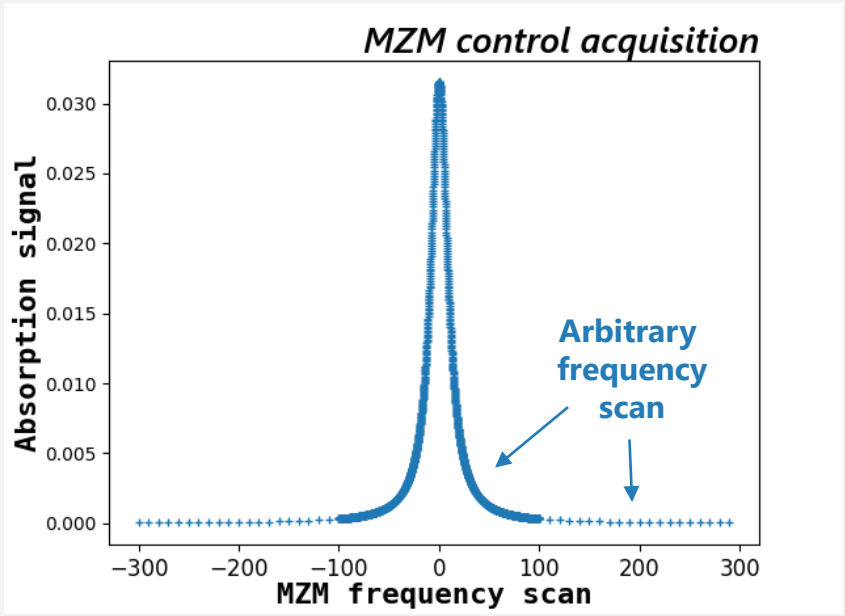

*finesse 300 000*




**Continuous frequency tuning**

# THz source in practice




## *Absolute frequency measurement*

GPS Beatnote vs OFC

Refimeve+ Réseau fibré métrologique à vocation européenne

## *Performances*

**Purity & Stability < Hz**

[5]L Djevahirdjian et al Nature Communications nov. 2023

# Study of absorption profile : $H_2O$

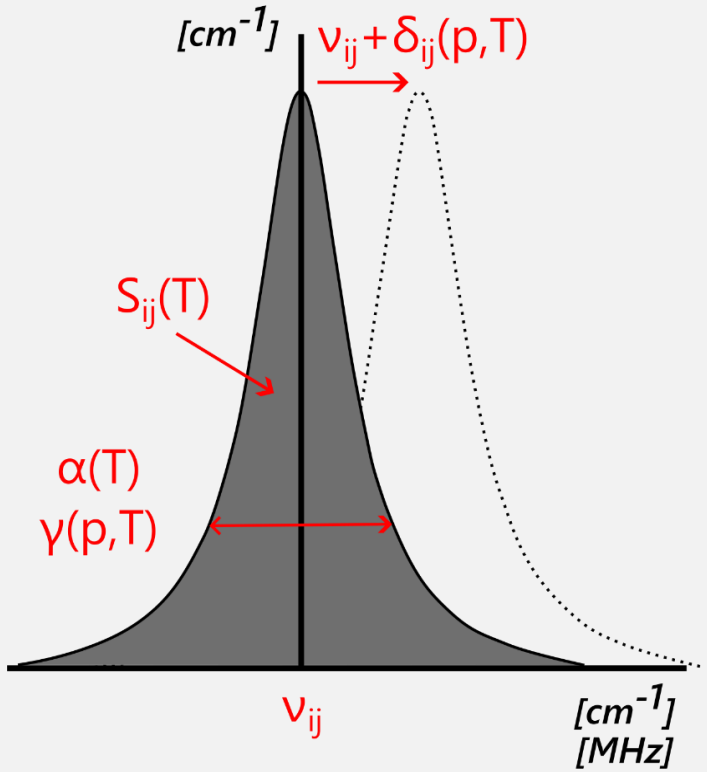
***Now***

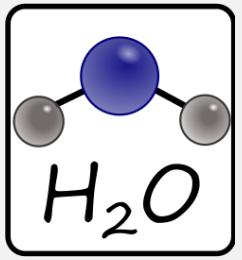
**Metrologic & Quantitative measurement**

**$H_2O$**

**Already studied**[6][7][8][9]
- **Comparison**
- **Improvement of calculated parameters**[10]
- **Not given (delta self)**


**Pressure broadening (gamma) and shift (delta)**

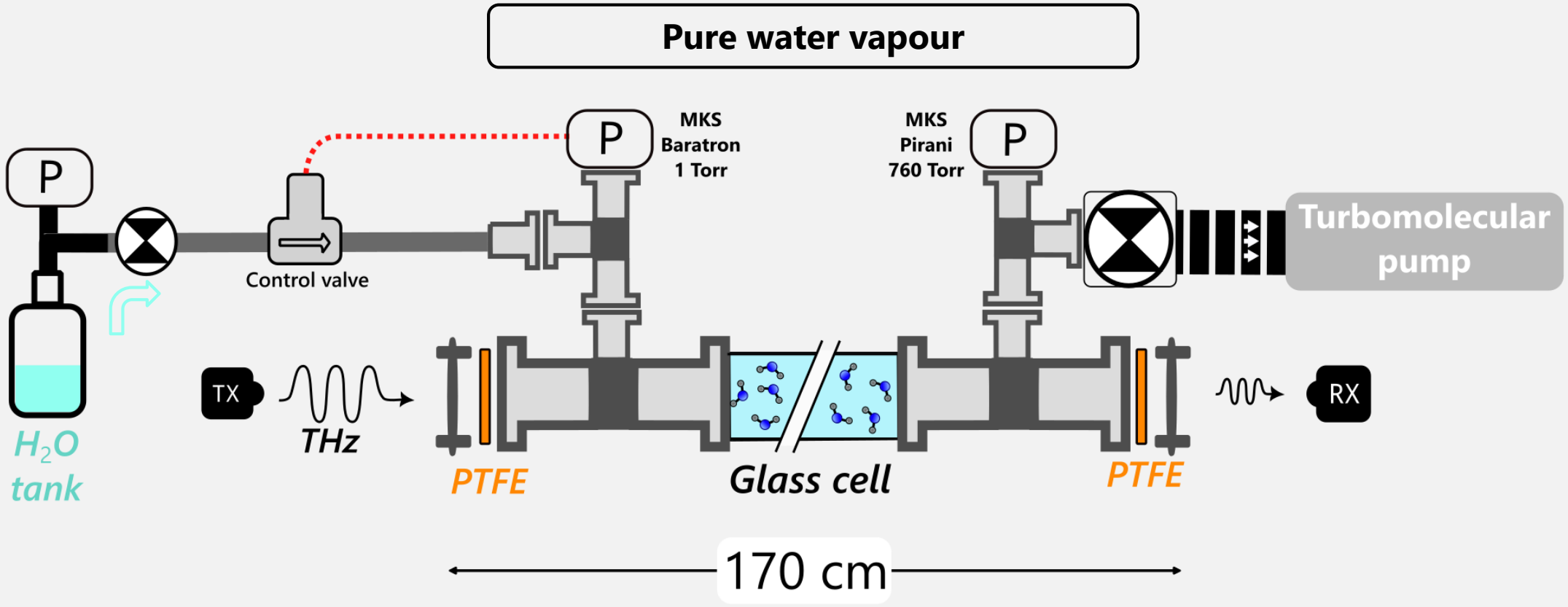




*[6] G. Yu. Golubiatnikov, Journal of Quantitative Spectroscopy and Radiative Transfer, juill. 2008*
*[7] V.B. Podobedov, Journal of Quantitative Spectroscopy and Radiative Transfer*
*[8] G. Cazzoli, Journal of Quantitative Spectroscopy and Radiative Transfer, juin 2008*
*[9] G. Cazzoli,, Journal of Quantitative Spectroscopy and Radiative Transfer, nov. 2008,*
*[10] R.R. Gamache, (2020) private communication. CRB calculation for different isotopologues of water vapor*

# $H_2O$ gas setup




**Pressure control with solenoid valve**

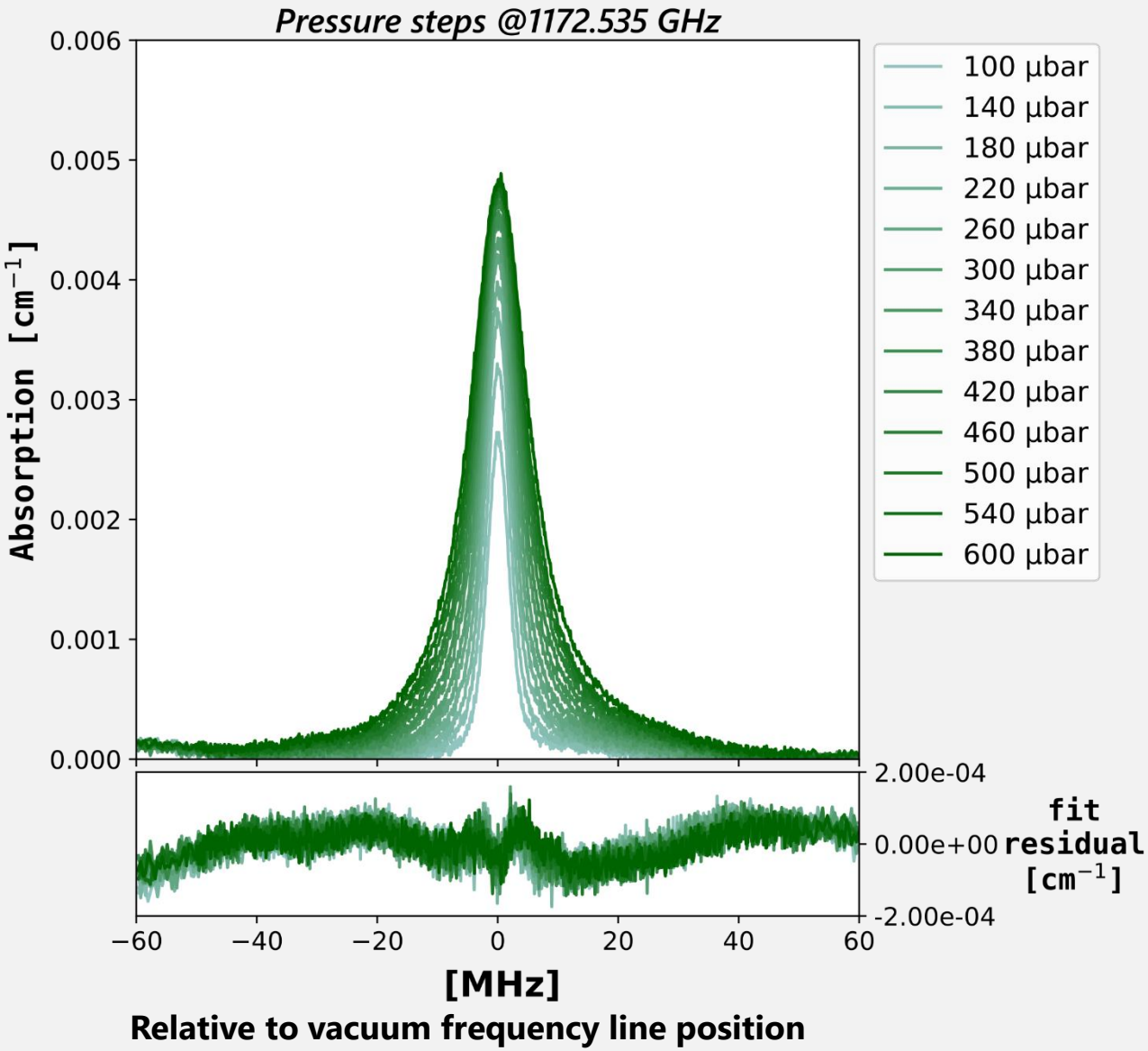
from 10 µbar to 1 mbar

**Direct absorption**

Three pressure sensors

- **$H_2O$ tank**
- **Cell precise : 1 Torr**
- **Cell coarse : pirani**

# Pressure dependence




**Record x30 - 40 spectra each pressure step**

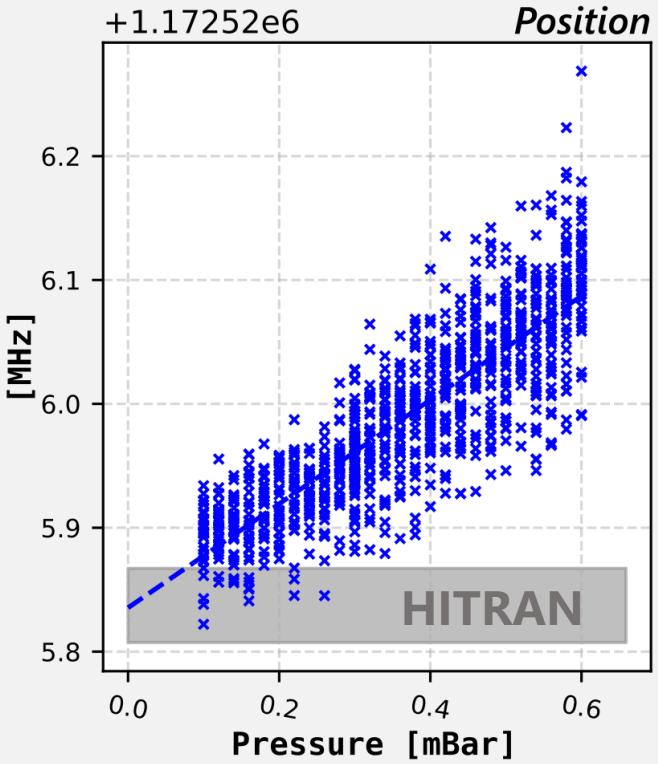
**Voigt profile : Doppler fixe**

- Area
- Lorentzian width
- Line position

Work in progress reduce fit residual to get a better accuracy

# $H_2O$ $7_{44}$ – $6_{51}$ @1172 GHz

*preliminary results*



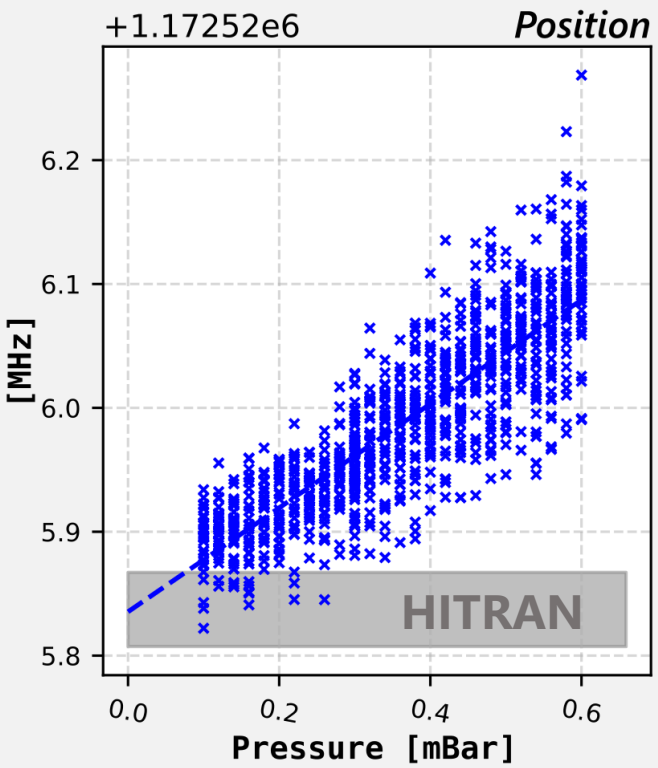

**~1000 spectra**

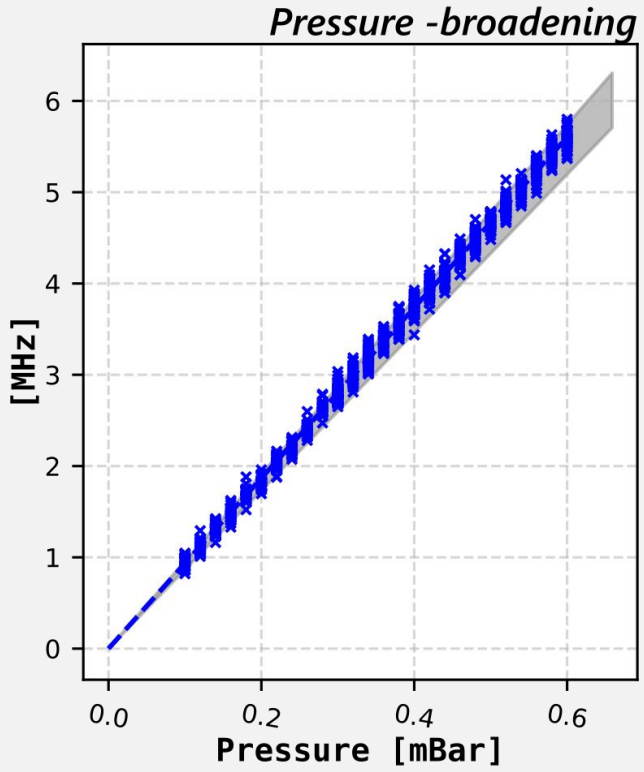
**Pressure shift**
0.420(9) MHz/mbar

**Position** 1172525. 835(4) MHz

# $H_2O$ $7_{44}$ – $6_{51}$ @1172 GHz

*preliminary results*



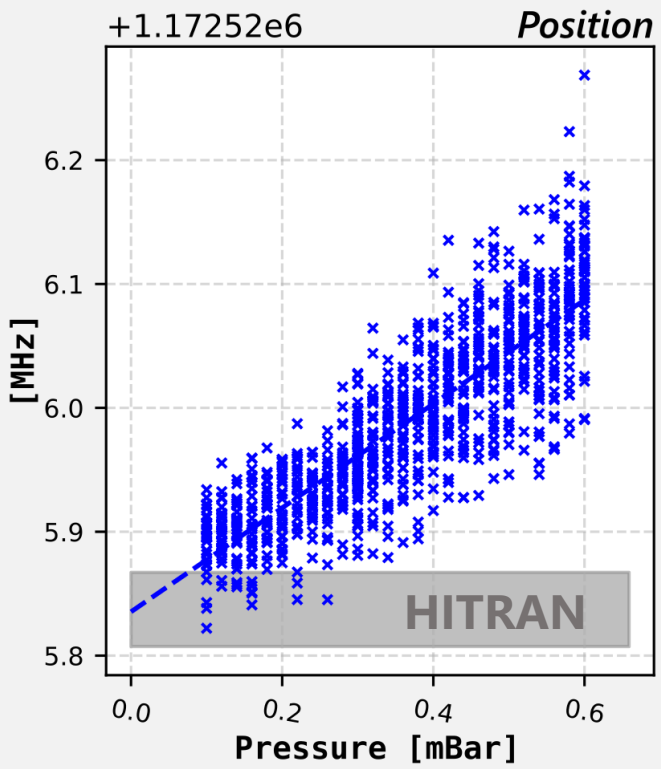

**Pressure shift**
0.420(9) MHz/mbar

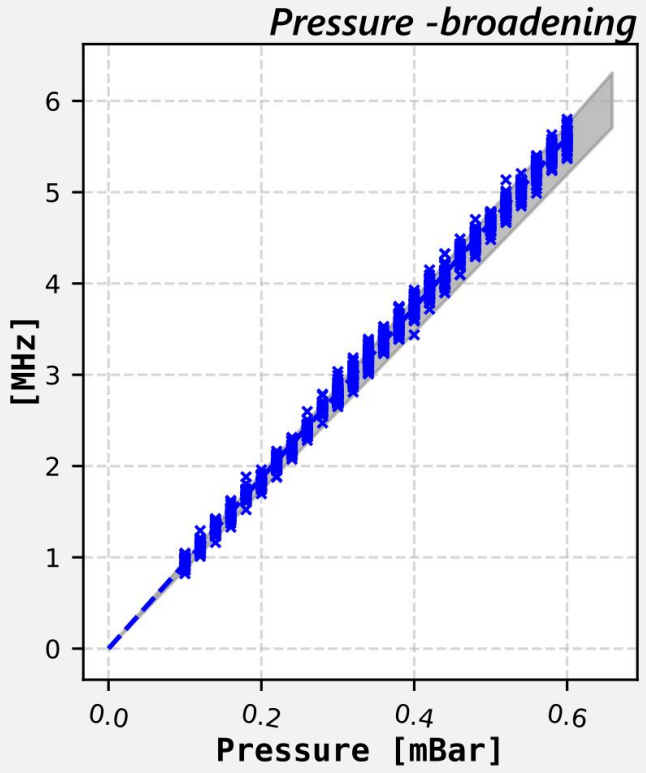
**Position** 1172525. 835(4) MHz

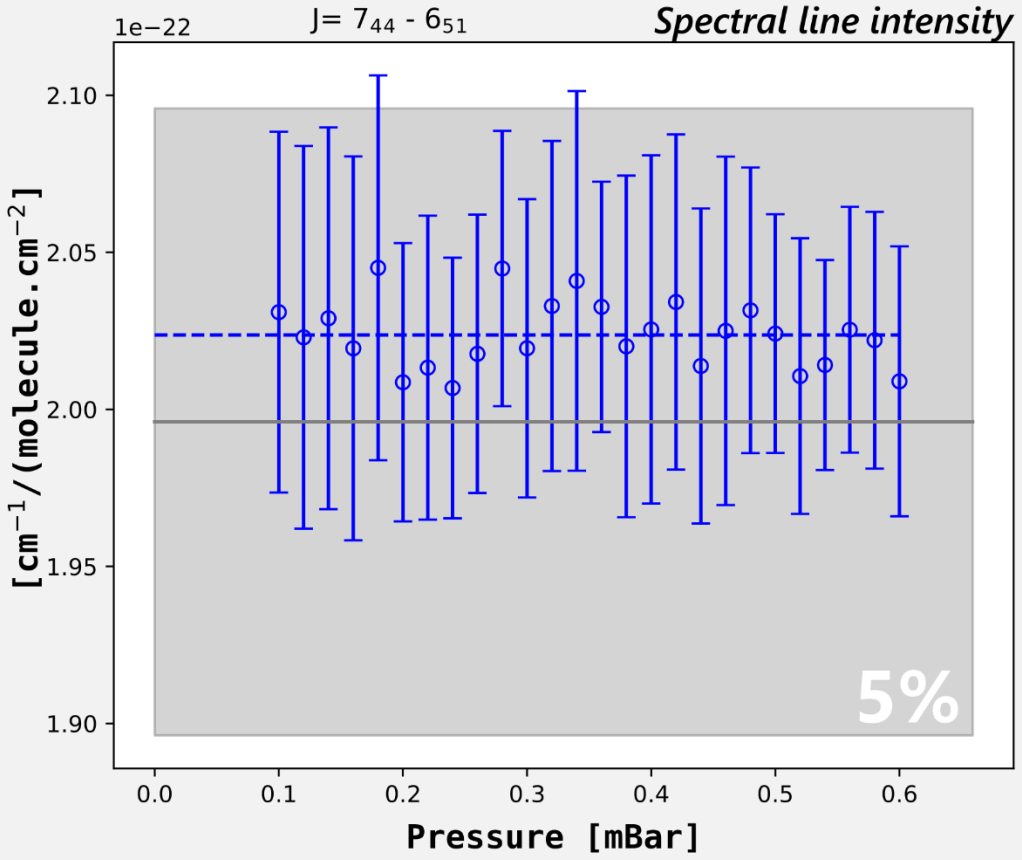
**Self-broadening**
9.350(7) MHz/mbar

# $H_2O$ $7_{44}$ – $6_{51}$ @1172 GHz

*preliminary results*




**Pressure shift**
0.420(9) MHz/mbar

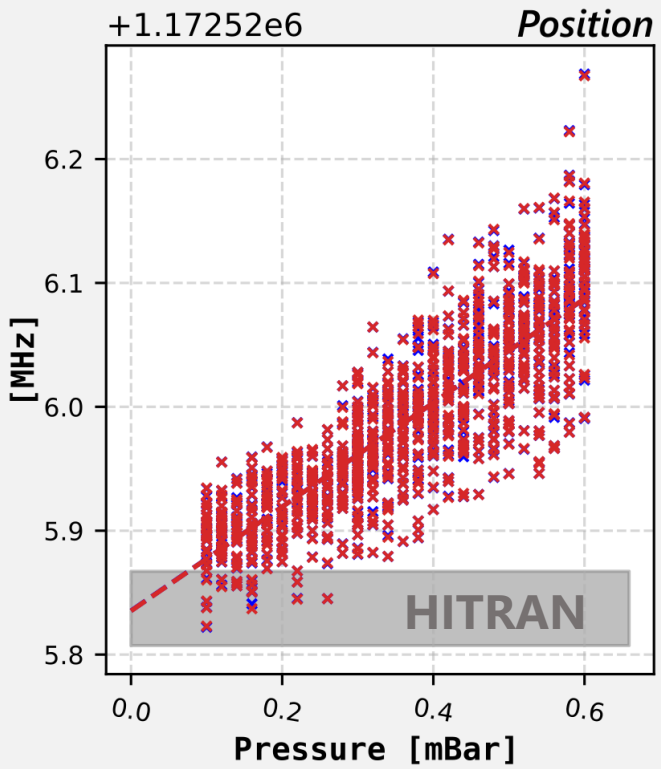
**Position** 1172525. 835(4) MHz

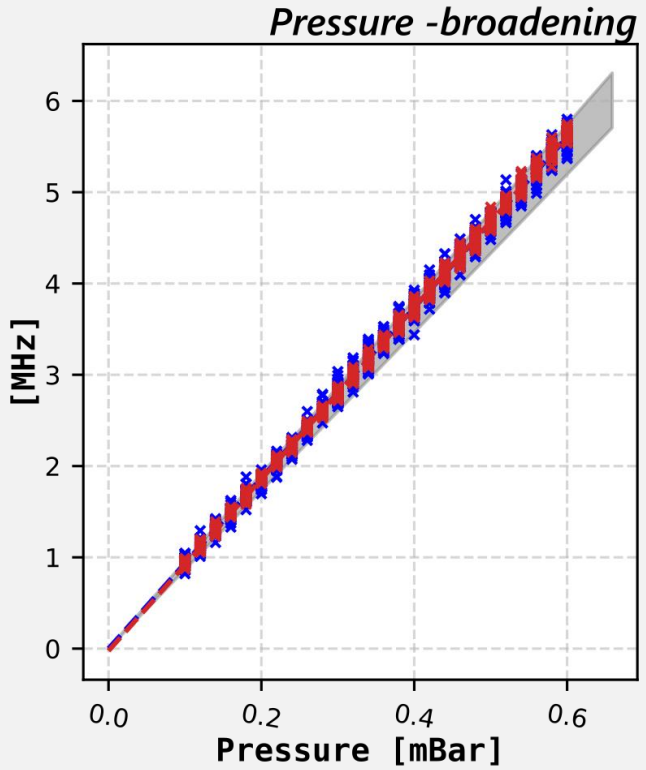
**Self-broadening**
9.350(7) MHz/mbar

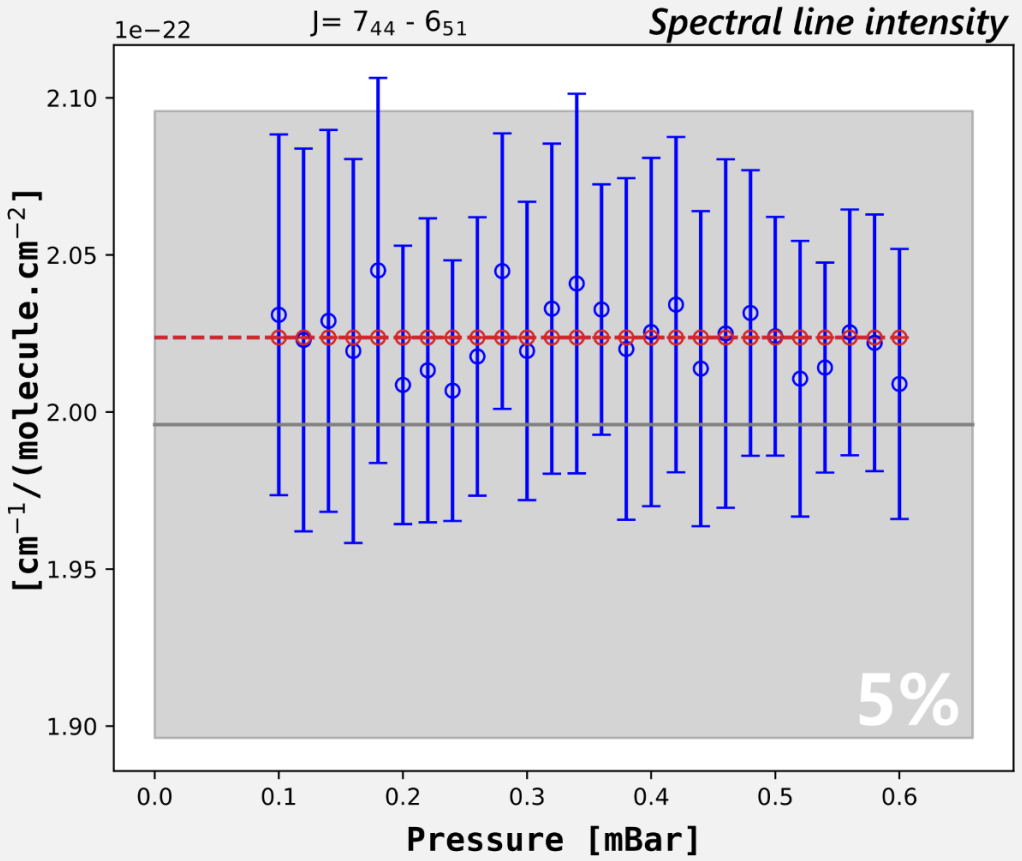
**Line intensity fixed to $2.016\times10^{-22}$**

# $H_2O$ $7_{44} - 6_{51}$ @1172 GHz

*preliminary results*




**Pressure shift**
0.420(9) MHz/mbar

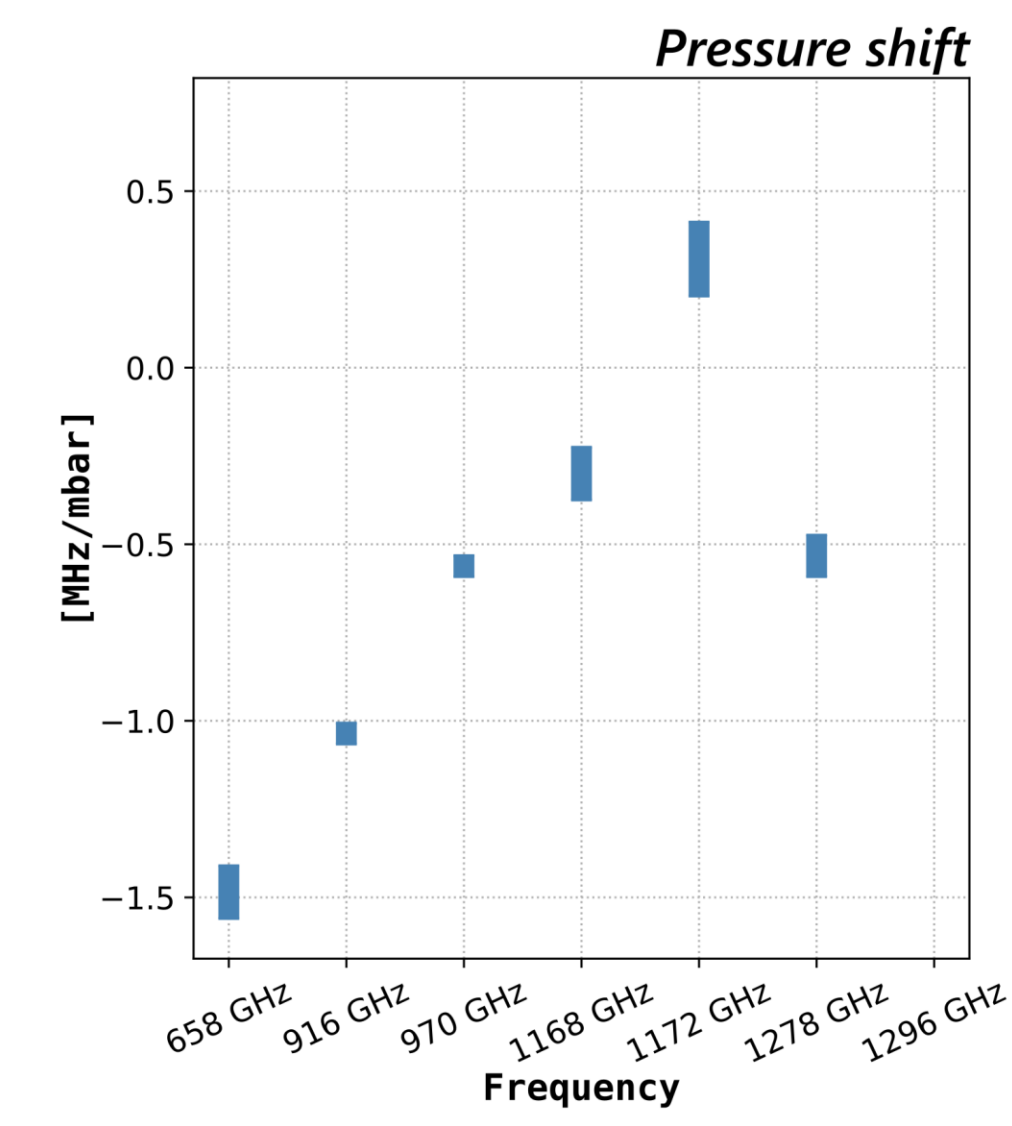
**Position** 1172525. 835(4) MHz

**Self-broadening**
9.350(4) MHz/mbar
(7)

**Line intensity fixed to $2.016 \times 10^{-22}$**

# Results overview : pressure shift

*preliminary results*





*[6] G. Yu. Golubiatnikov, Journal of Quantitative Spectroscopy and Radiative Transfer, juill. 2008*

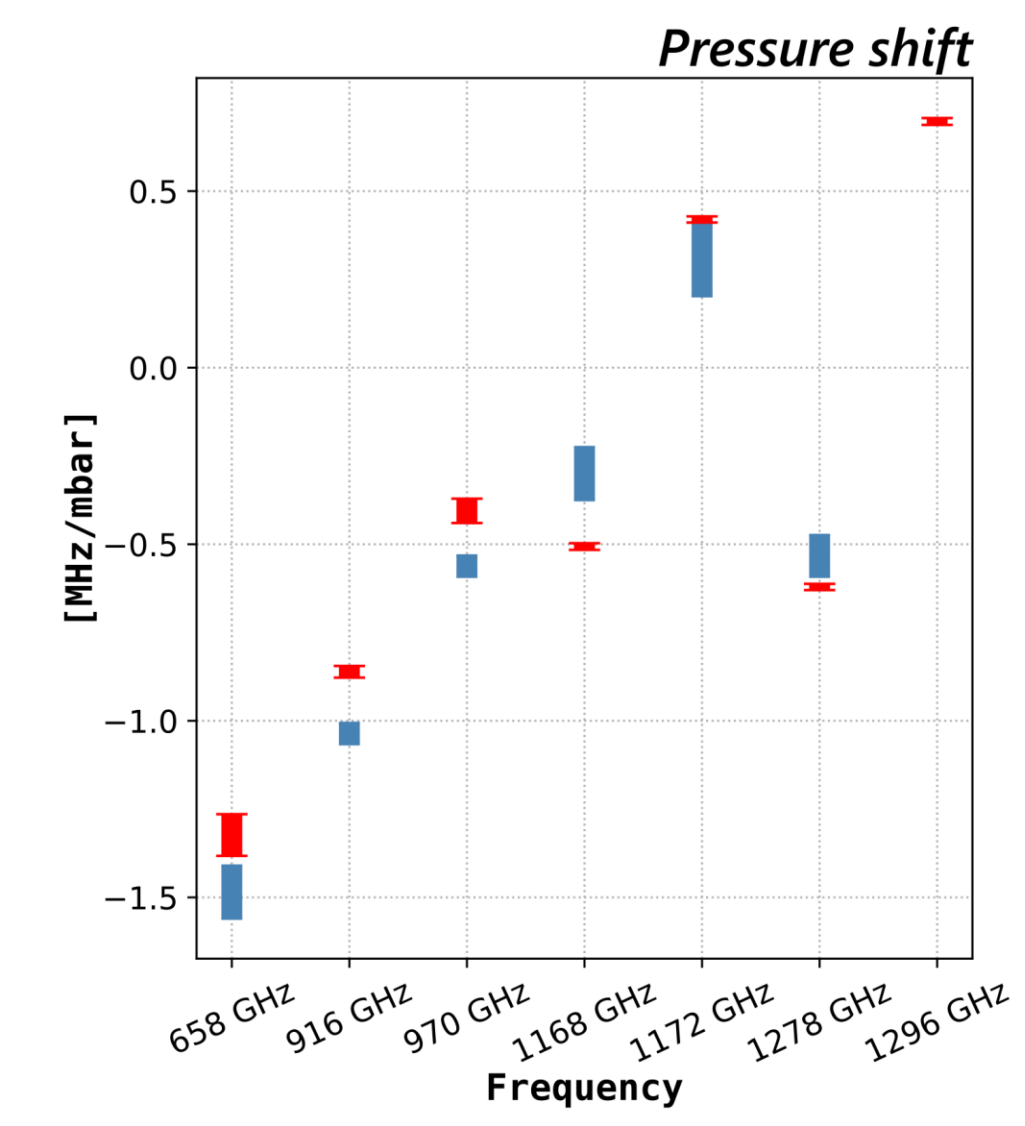
*[7] V.B. Podobedov, Journal of Quantitative Spectroscopy and Radiative Transfer, 2004*

*[8] G. Cazzoli, Journal of Quantitative Spectroscopy and Radiative Transfer, juin 2008*

*[9] G. Cazzoli,, Journal of Quantitative Spectroscopy and Radiative Transfer, nov. 2008,*

# Results overview : pressure shift

*preliminary results*





*[6] G. Yu. Golubiatnikov, Journal of Quantitative Spectroscopy and Radiative Transfer, juill. 2008*

*[7] V.B. Podobedov, Journal of Quantitative Spectroscopy and Radiative Transfer, 2004*

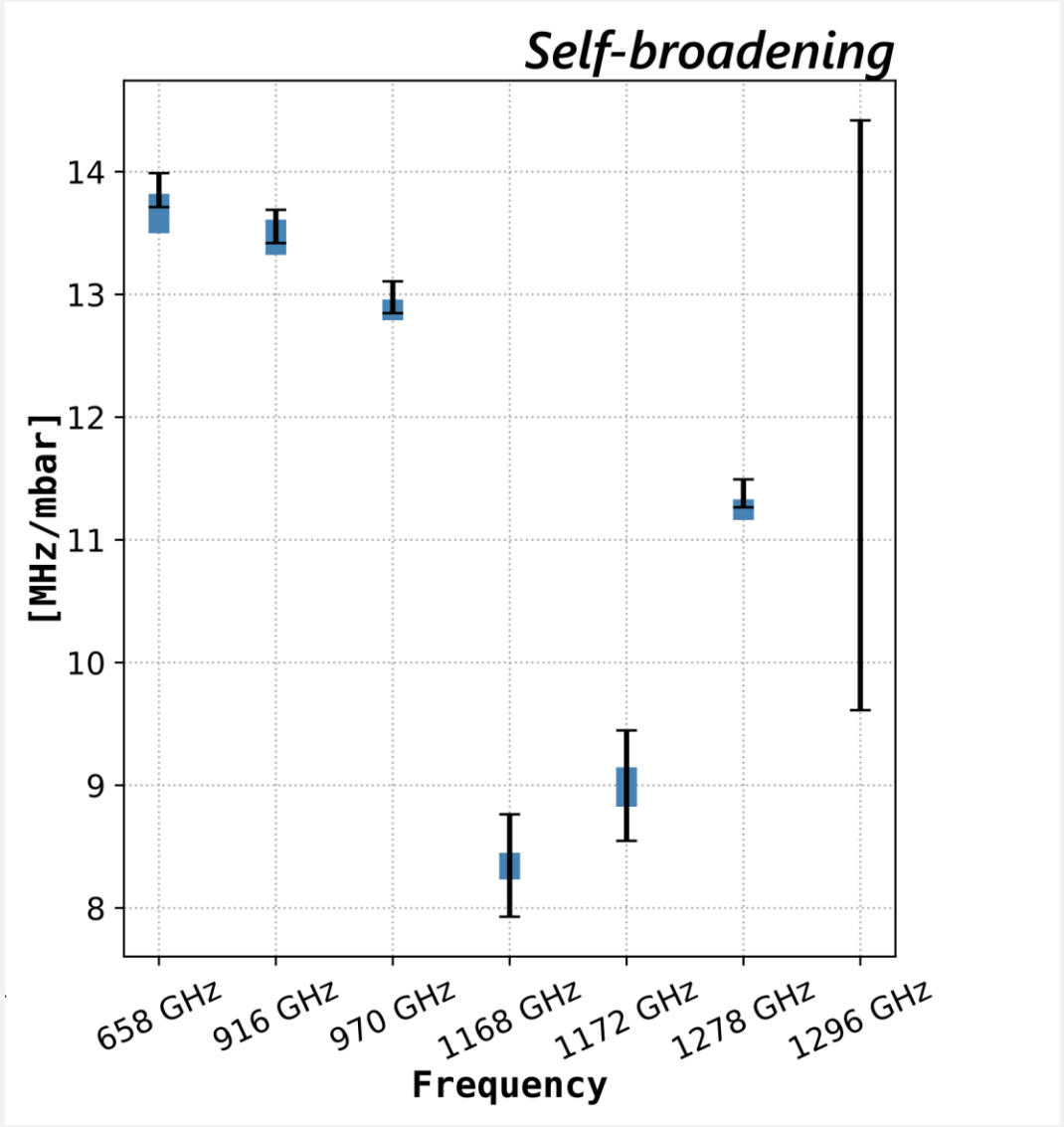
*[8] G. Cazzoli, Journal of Quantitative Spectroscopy and Radiative Transfer, juin 2008*

*[9] G. Cazzoli,, Journal of Quantitative Spectroscopy and Radiative Transfer, nov. 2008,*


**No agreement with literature**

# Results overview : self broadening

*preliminary results*


*[6] G. Yu. Golubiatnikov, Journal of Quantitative Spectroscopy and Radiative Transfer, juill. 2008*

*[7] V.B. Podobedov, Journal of Quantitative Spectroscopy and Radiative Transfer, 2004*

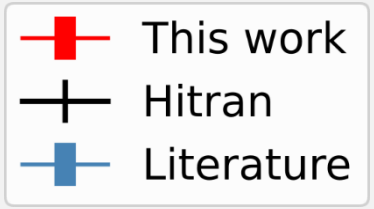
*[8] G. Cazzoli, Journal of Quantitative Spectroscopy and Radiative Transfer, juin 2008*

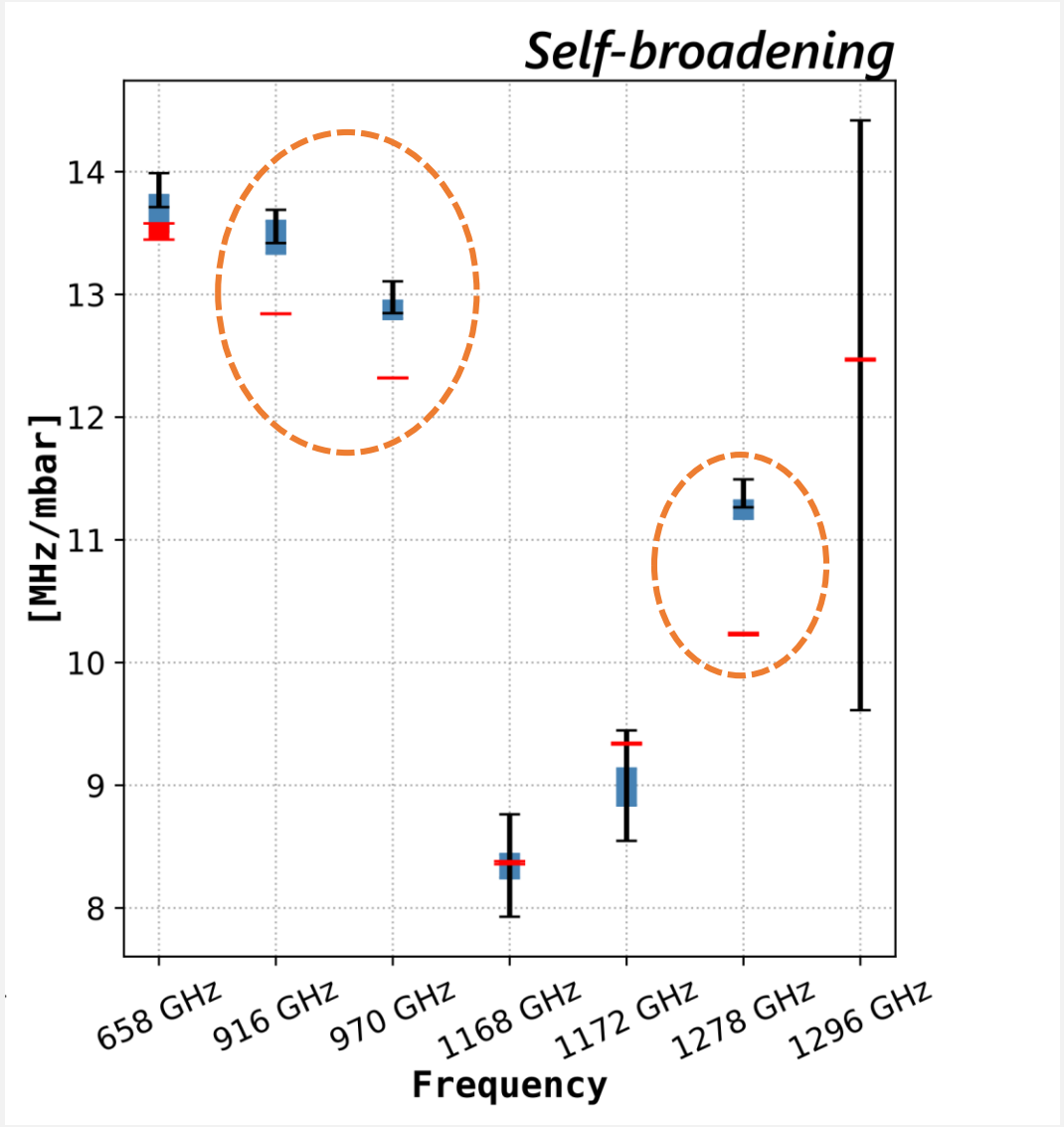
*[9] G. Cazzoli,, Journal of Quantitative Spectroscopy and Radiative Transfer, nov. 2008,*

*[10] R.R. Gamache, (2020) private communication. CRB calculation for different isotopologues of water vapor*

# Results overview : self broadening

*preliminary results*





*[6] G. Yu. Golubiatnikov, Journal of Quantitative Spectroscopy and Radiative Transfer, juill. 2008*

***[7] V.B. Podobedov, Journal of Quantitative Spectroscopy and Radiative Transfer, 2004***

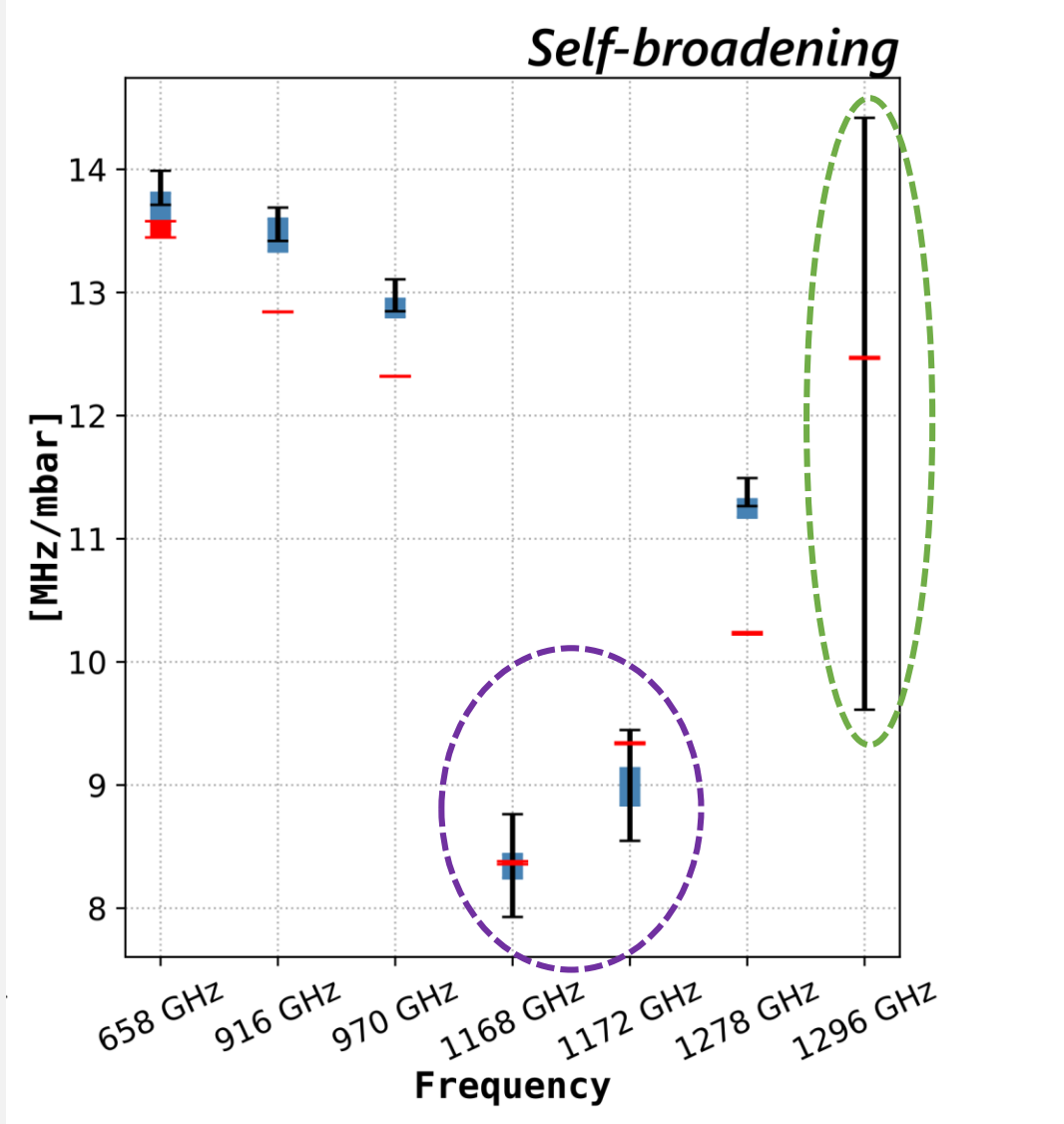
*[8] G. Cazzoli, Journal of Quantitative Spectroscopy and Radiative Transfer, juin 2008*

*[9] G. Cazzoli,, Journal of Quantitative Spectroscopy and Radiative Transfer, nov. 2008,*

*[10] R.R. Gamache, (2020) private communication. CRB calculation for different isotopologues of water vapor*

# Results overview : self broadening

*preliminary results*





*[6] G. Yu. Golubiatnikov, Journal of Quantitative Spectroscopy and Radiative Transfer, juill. 2008*

*[7] V.B. Podobedov, Journal of Quantitative Spectroscopy and Radiative Transfer, 2004*

***[8] G. Cazzoli, Journal of Quantitative Spectroscopy and Radiative Transfer, juin 2008***

***[9] G. Cazzoli,, Journal of Quantitative Spectroscopy and Radiative Transfer, nov. 2008,***

***[10] R.R. Gamache, (2020) private communication. CRB calculation for different isotopologues of water vapor***

# Conclusion & perspective

## Source THz

**Photomixing**

**Continuous frequency scan**

**Hz** level up to 1.2 THz

## $H_2O$ vapour study

**Line profile recordings**

**Pressure broadening**

Disagreement with literature

**Confirm Pressure shift trends**

## Next

**Detection limit**

**Higher frequencies**

**New molecular targets**

$NH_3$

$CH_3OH$

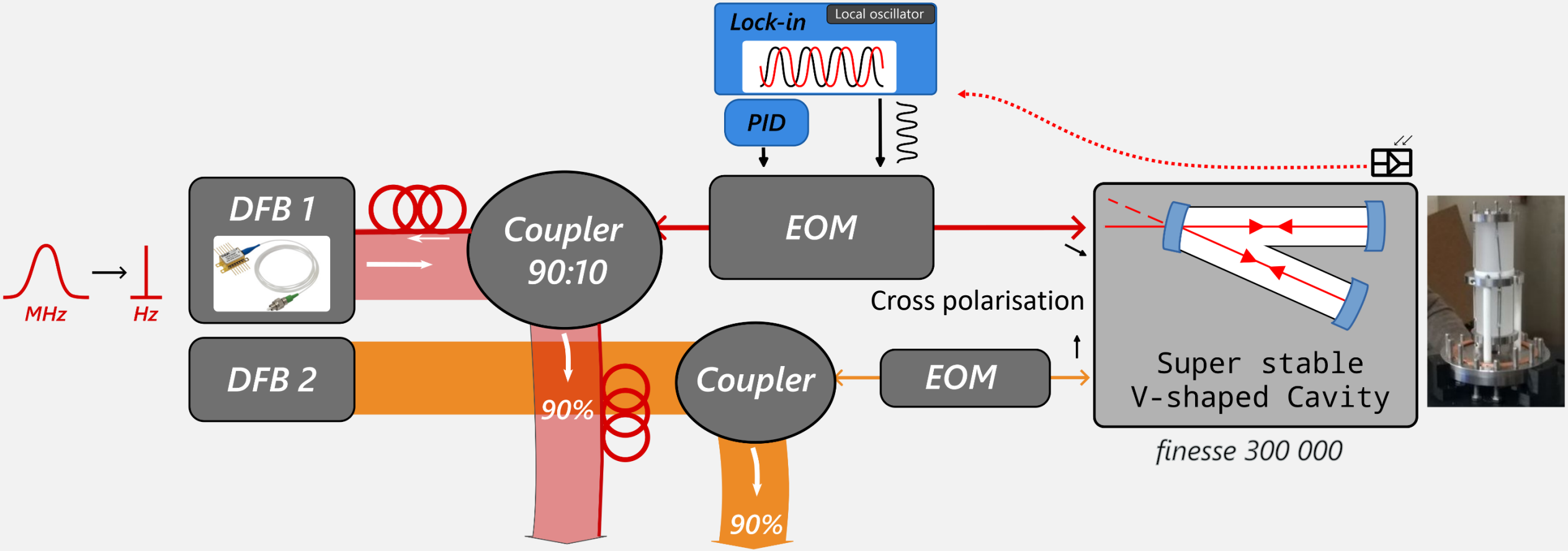
$N_2O$


# Thank you

Question ?

# Annexes

# Optical feedback technique with DFB laser : Two lasers



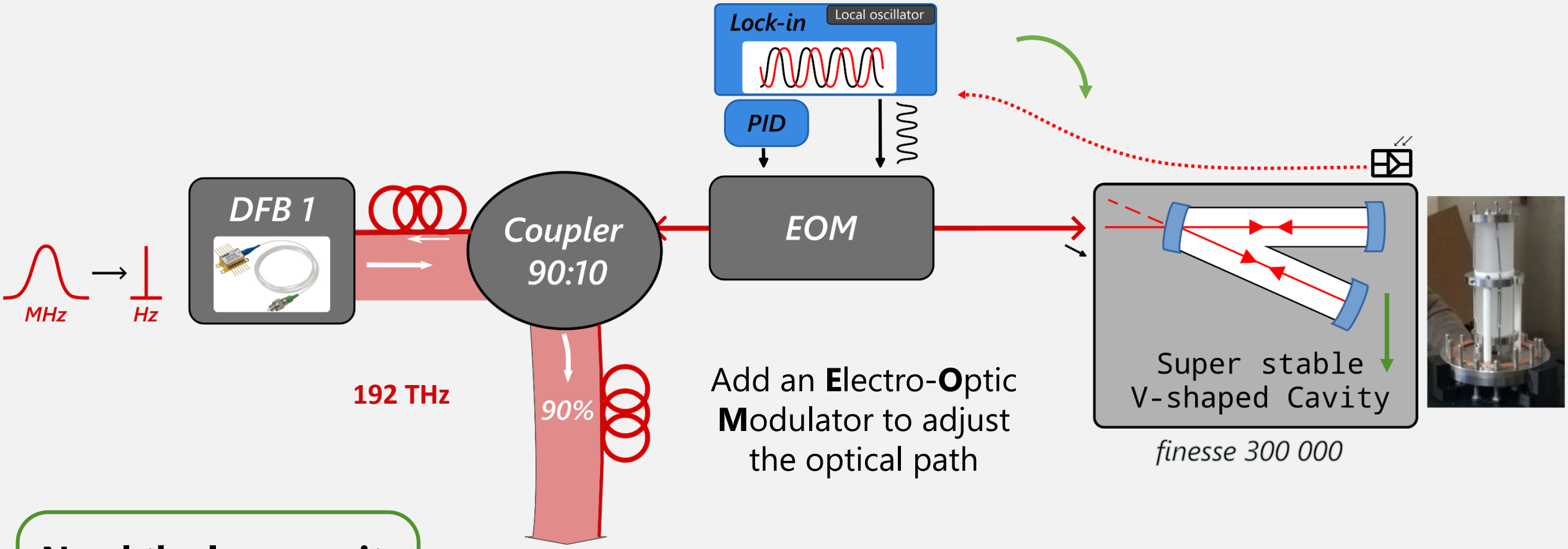

Photomixing

**192 THz** & **193 THz**

**Δ1 THz**

**Common fluctuation**

# Optical feedback technique with DFB laser : Phase control




Add an **E**lectro-**O**ptic **M**odulator to adjust the optical path

**Need the laser-cavity distance to be a multiple of λ/2**

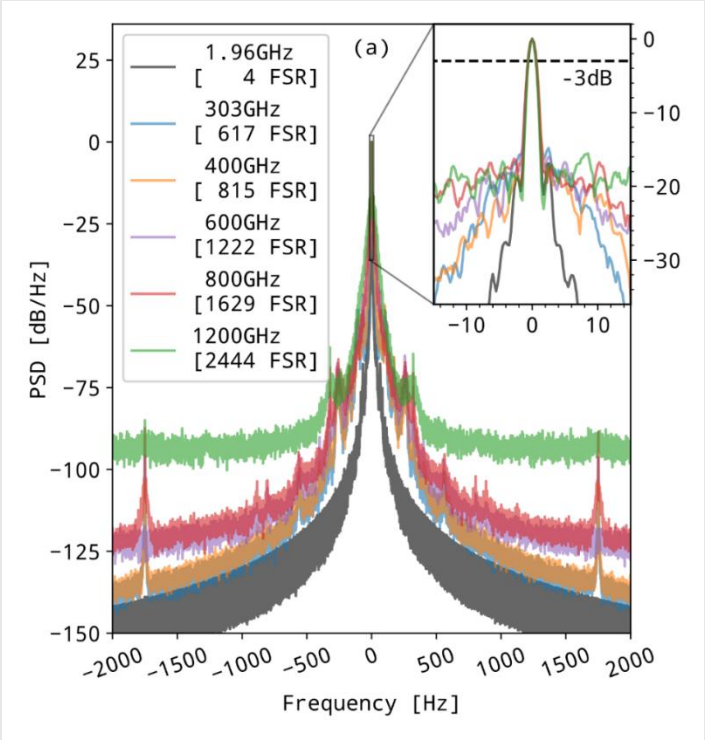
**DC** voltage
+
Weak **modulation**

Stay locked on top of the cavity mode

# Source performance : purity and stability

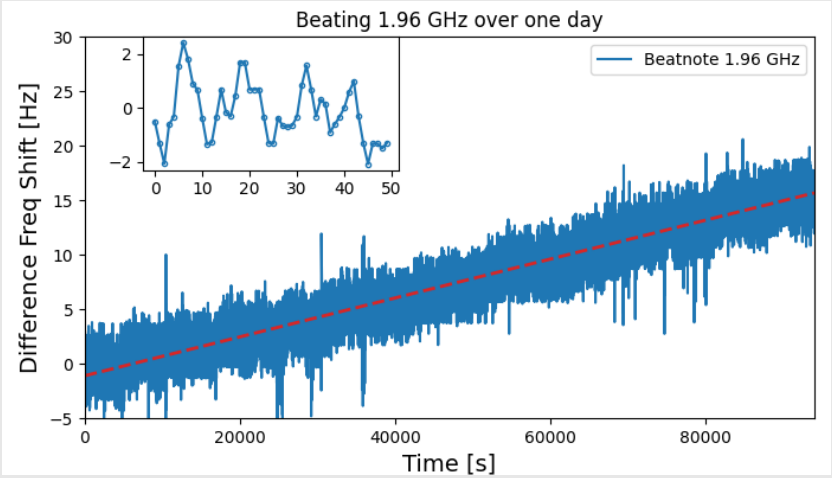
*[5]L. Djevahirdjian et al Nature Communications nov. 2023*

## Spectral purity from 2 to 1200 GHz

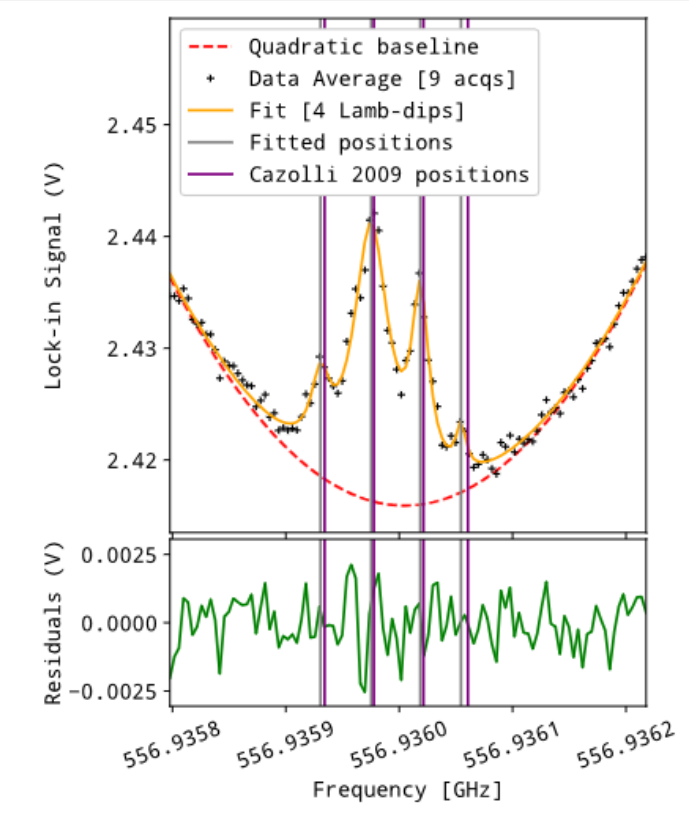



1 GHz to 1200 GHz
**Hz linewidth** emission

## Long term Stability over 1 day

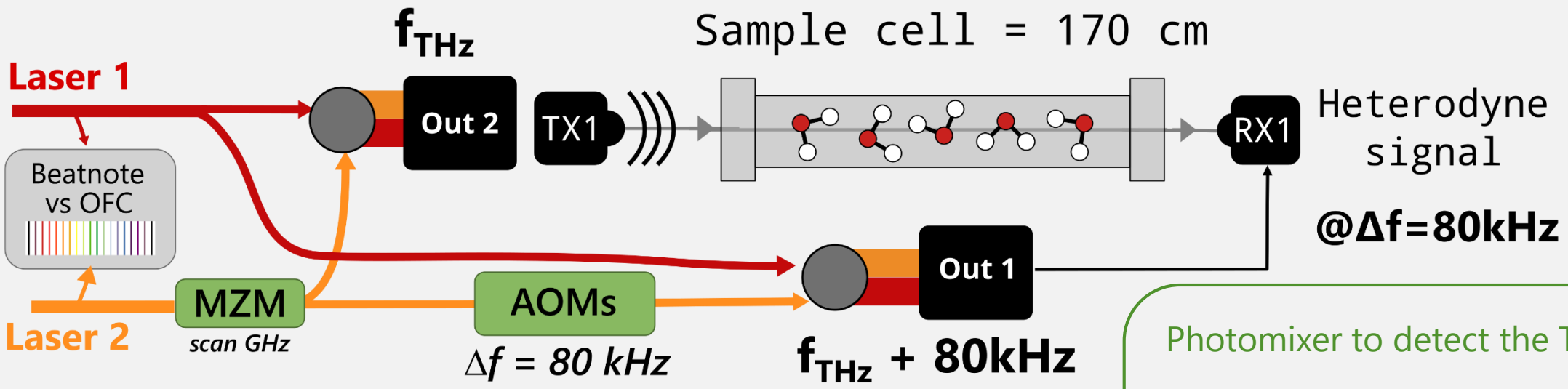



Measure @1.96GHz
**Drift < Hz** in 1 hour
Accuracy **200 mHz in 20 minute**




**Scan sub-Doppler structure**
**Linewidth ~10 kHz**

# THz detection




**Absolute** frequencies determination

**A**cousto **O**ptic **M**odulator to shift the frequency

Photomixer to detect the THz

Low response time

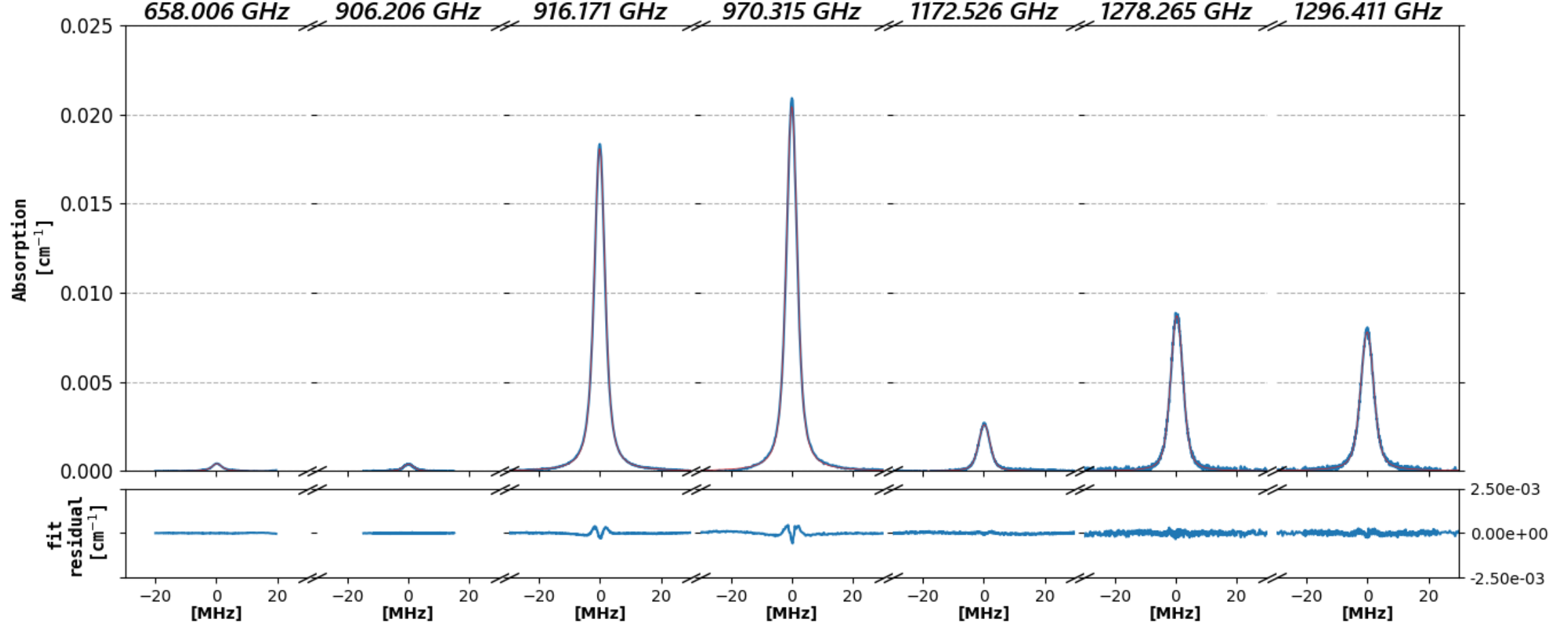
DC signal if same frequency

AC **Heterodyne** detection @80 KHz

Better SNR

# Absorption spectra overview

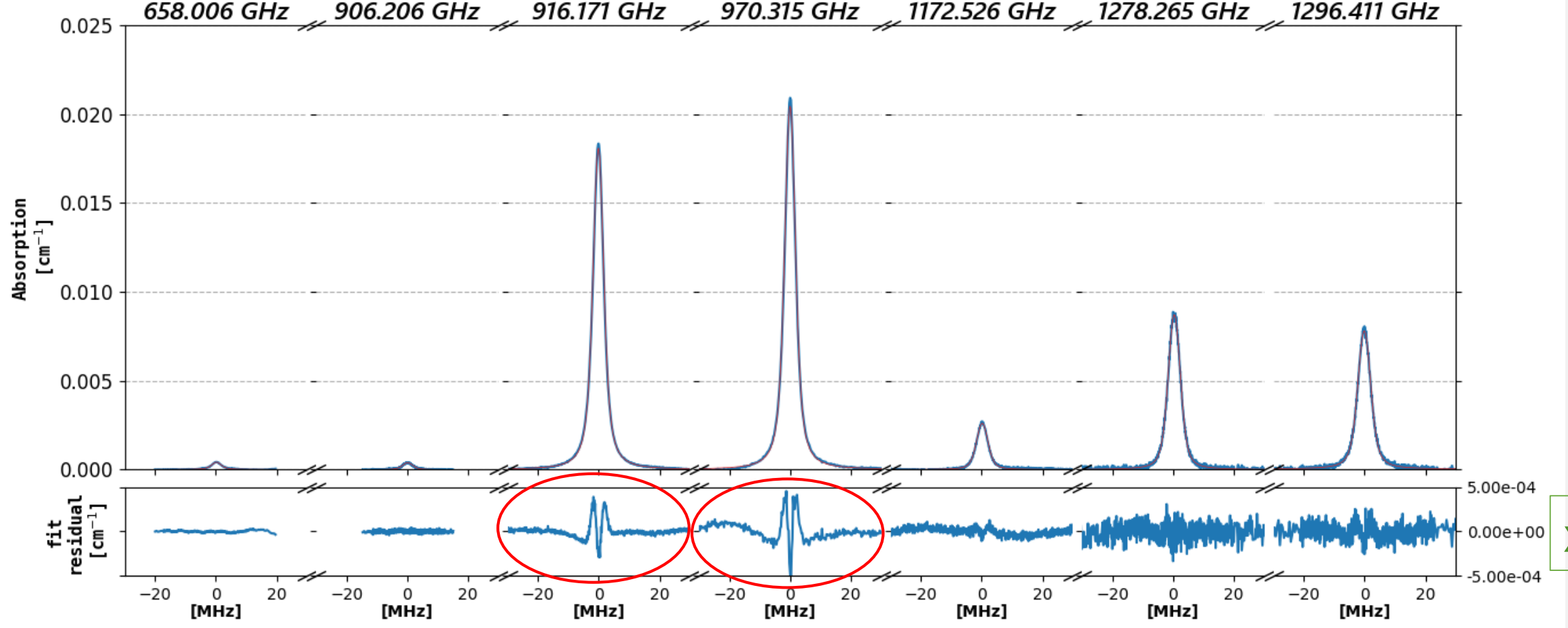
*Doppler fixed*




**@100 µbar with Voigt profile fit residual**

# Absorption spectra overview

*Doppler fixed*

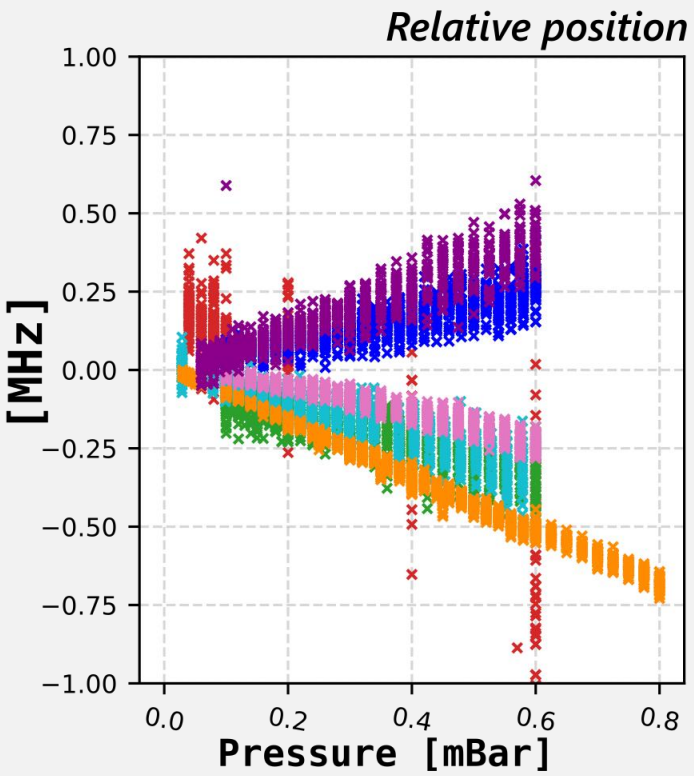


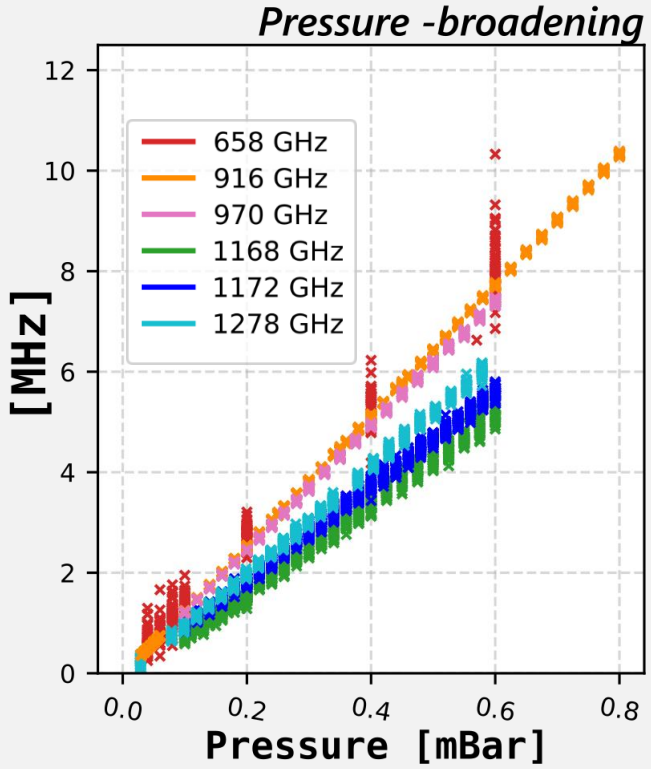

x5

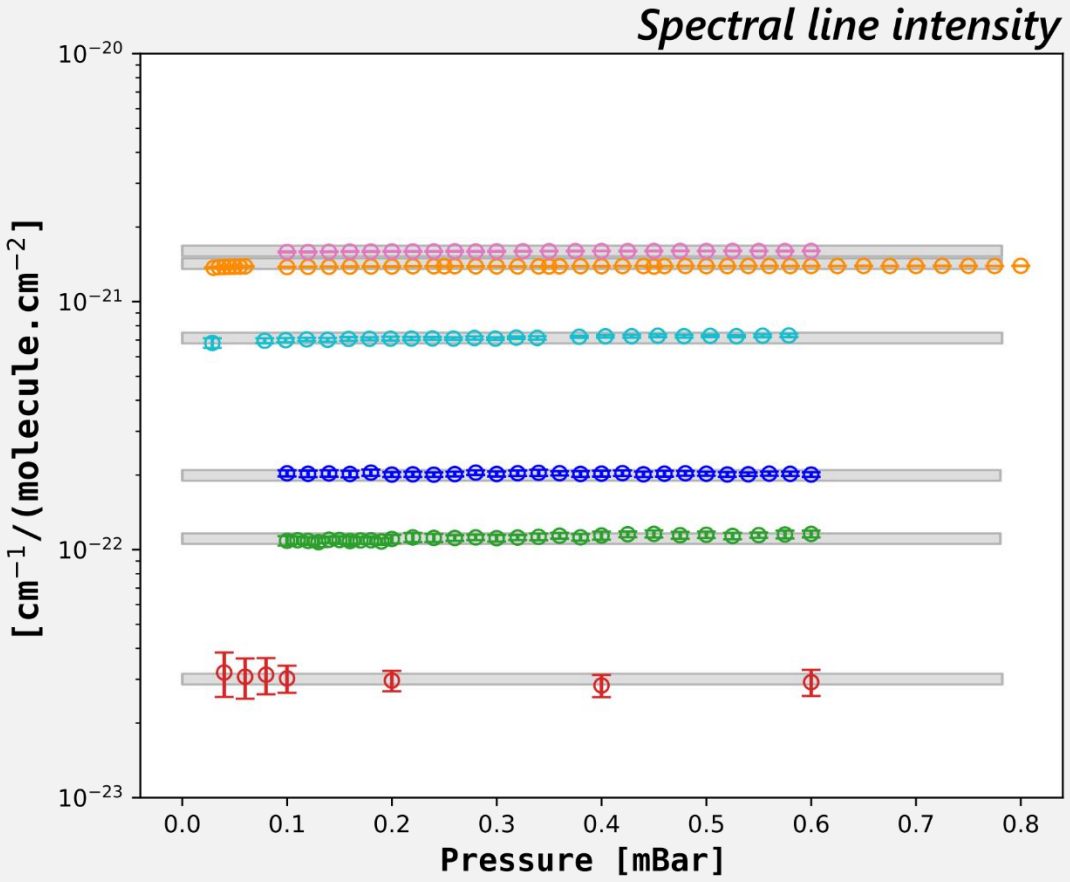
**Fit with Voigt profile, residual structure, work in progress…**

# Results overview

*Doppler fixed*




Frequency
and **pressure shift**

[MHz/mbar]

**Gamma self**
Pressure broadening

[MHz/mbar]

**Line intensity**
Good agreement with database

Instrumental validation

# Experimental position vs HITRAN

*Doppler fixed*