Développements instrumentaux micro-onde et optiques pour les mesures spectroscopiques de précisions avec des molécules

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AG 2021, Paris

Precision measurements with molecules

- Complementary to measurements in atoms for precision tests of fundamental physics:

measure constants	m_e/m_p (Schiller, Hilico/Karr, Ubachs, Koelemeij – $HD^{(+)}$, $H_2^{(+)}$) k_B (Gianfrani, $H_2{}^{18}O$, CO_2 - **LPL**, **NH_3**),…
measure their variations in time	α (J. Ye, OH) - m_e/m_p (Truppe/Hinds/Tarbutt, CH - Bethlem, NH_3 - **LPL**, **SF_6**)
test fundamental symmetries	parity & time-reversal symmetry (eEDM): Hinds (YbF), Cornell/Ye (HfH^+), DeMille/Doyle/Gabrielse (ThO) parity symmetry: D. DeMille (BaF), **LPL** (**chiral species**),…
QED tests, 5^{th} force	W. Ubachs (H_2, HD^+),…
test the symmetrization postulate	Tino, De Natale,… (O_3, CO_2, NH_3,…)

→ Many are based on high-resolution spectroscopy, often in the mid-infrared domain

- Frequency references for frequency metrology

provide an almost continuous set of references throughout the MW, THz, IR, visible, UV

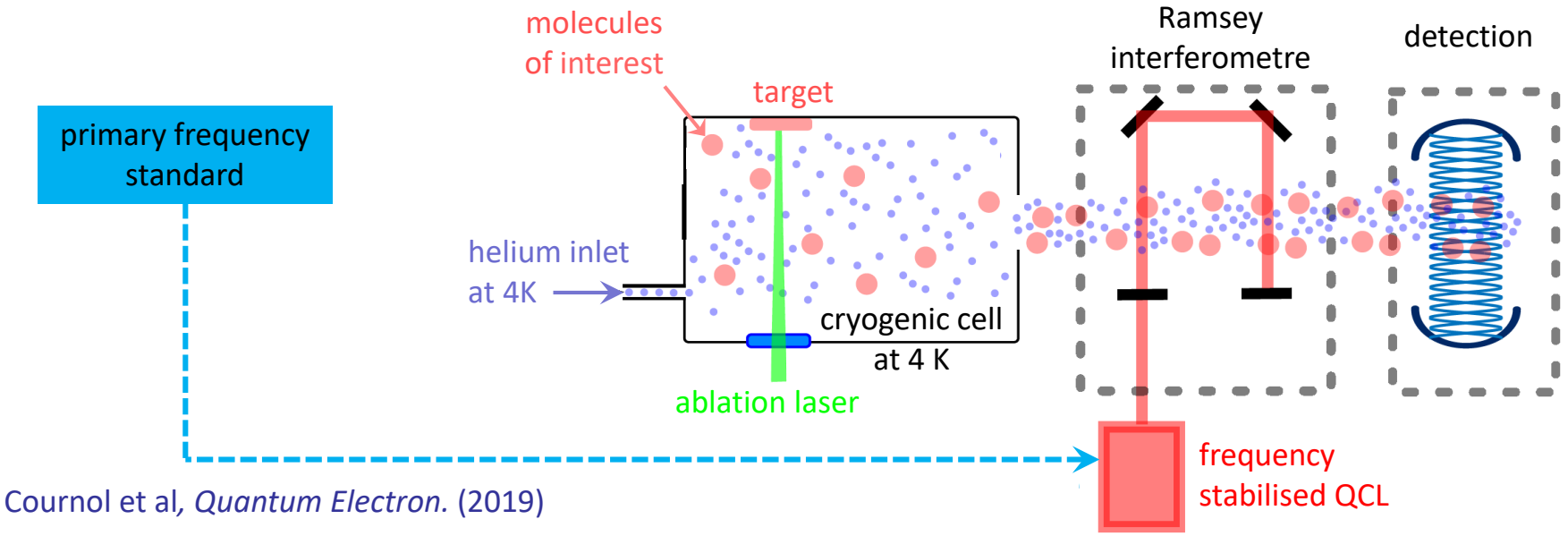
→ Require advanced manipulation techniques (already demonstrated in atomic physics): control/cooling of internal and external degrees of freedom, individual internal states addressability, state-selective high detection-sensitivity and -rate, long coherence times…

Outline

- Development of a high-sensitivity microwave detector for molecules

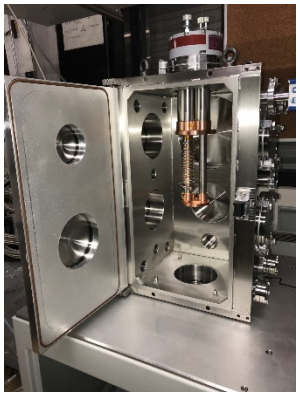
- Implementation of the new kelvin using Doppler Broadening Thermometry

A machine for reaching record 10^{-15} vibrational frequency uncertainties



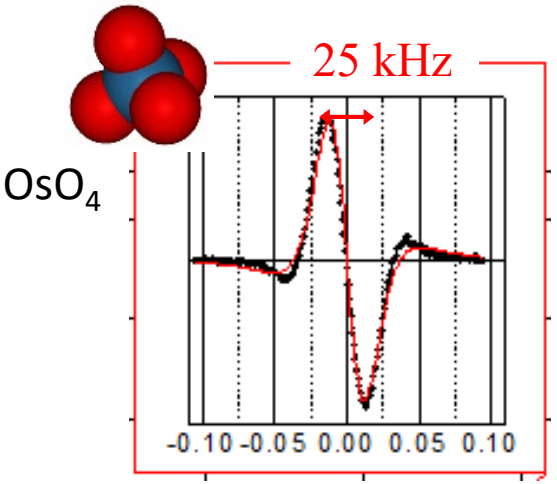
Cournol et al, *Quantum Electron.* (2019)

buffer-gas-cooled molecular beam

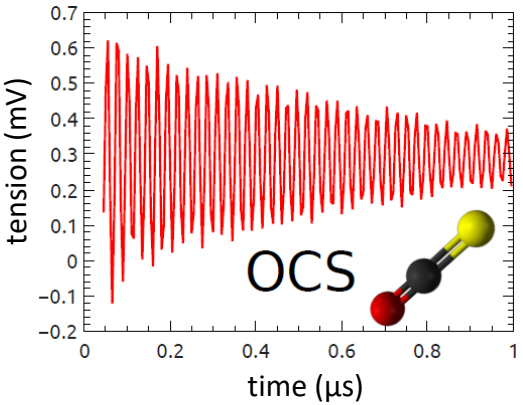


cryostat under construction

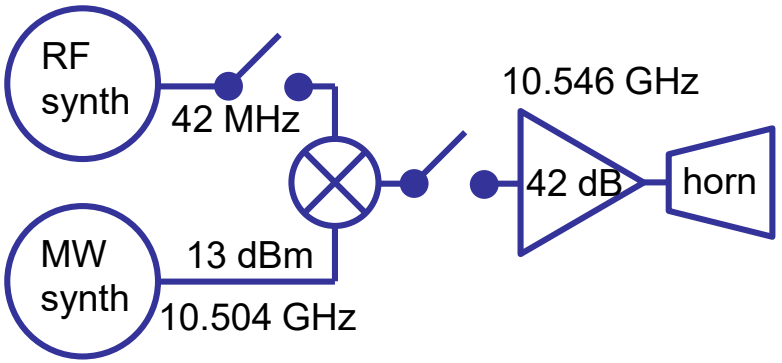
ultra-stable and SI-traceable quantum cascade laser (QCL)



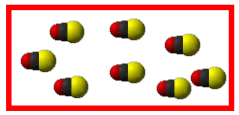
high-sensitivity microwave detection



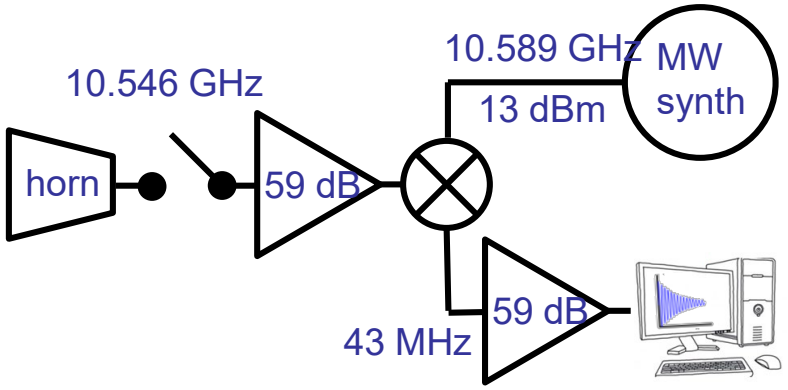
High-sensitivity microwave detector (2-18 GHz)



S=C=O



gas cell

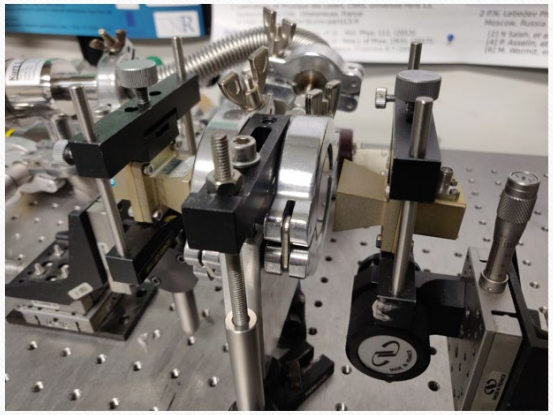


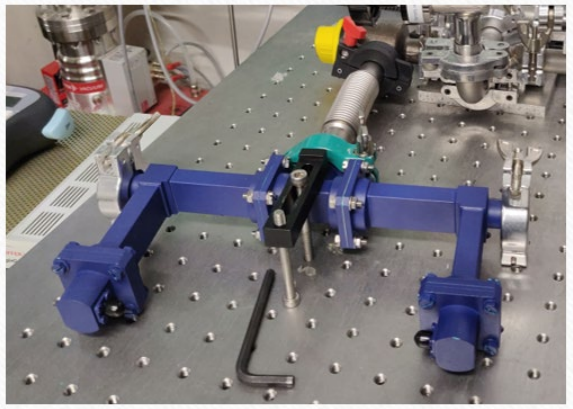
Writing:

- 100 ns min pulses
- 4 W max

Reading:

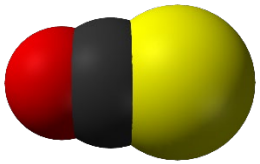
- down-conversion MW -> RF
- fast acquisition computer cards



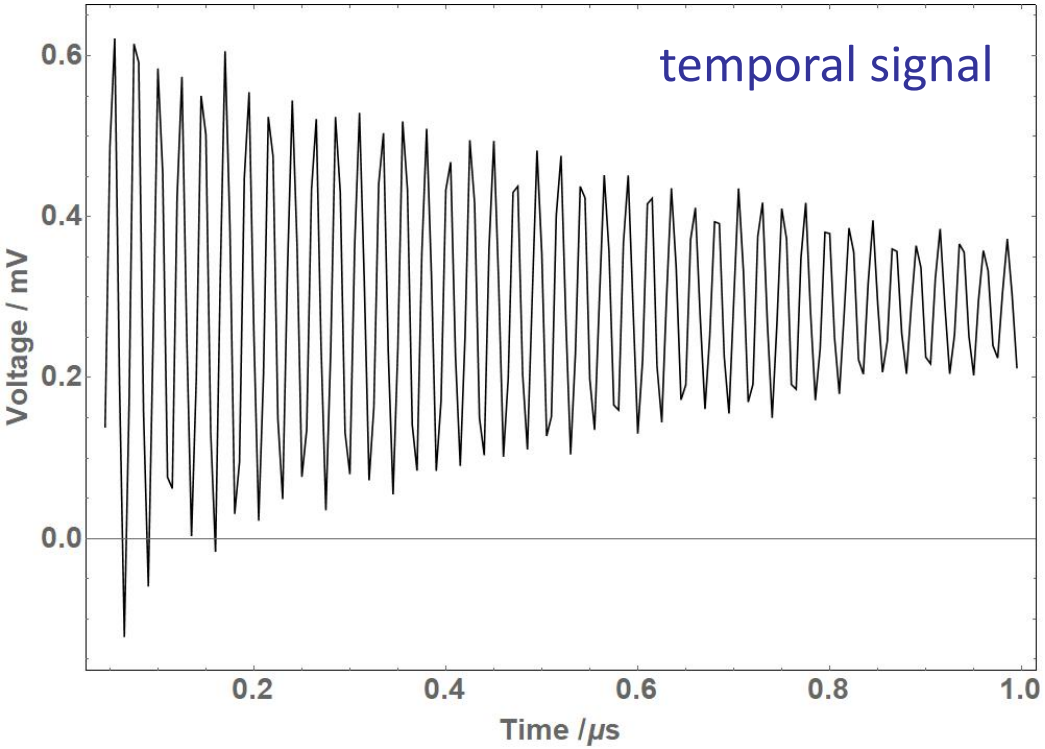


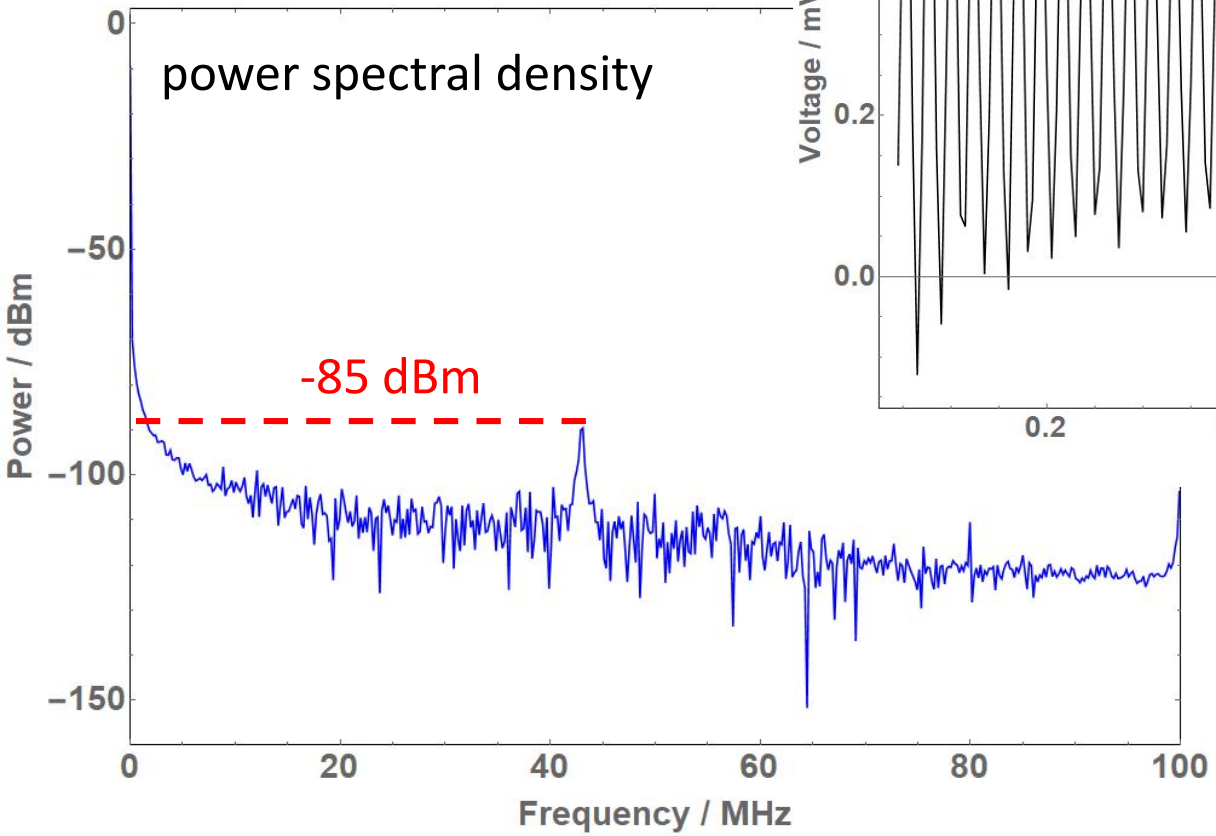
FID signals of OCS in a cell

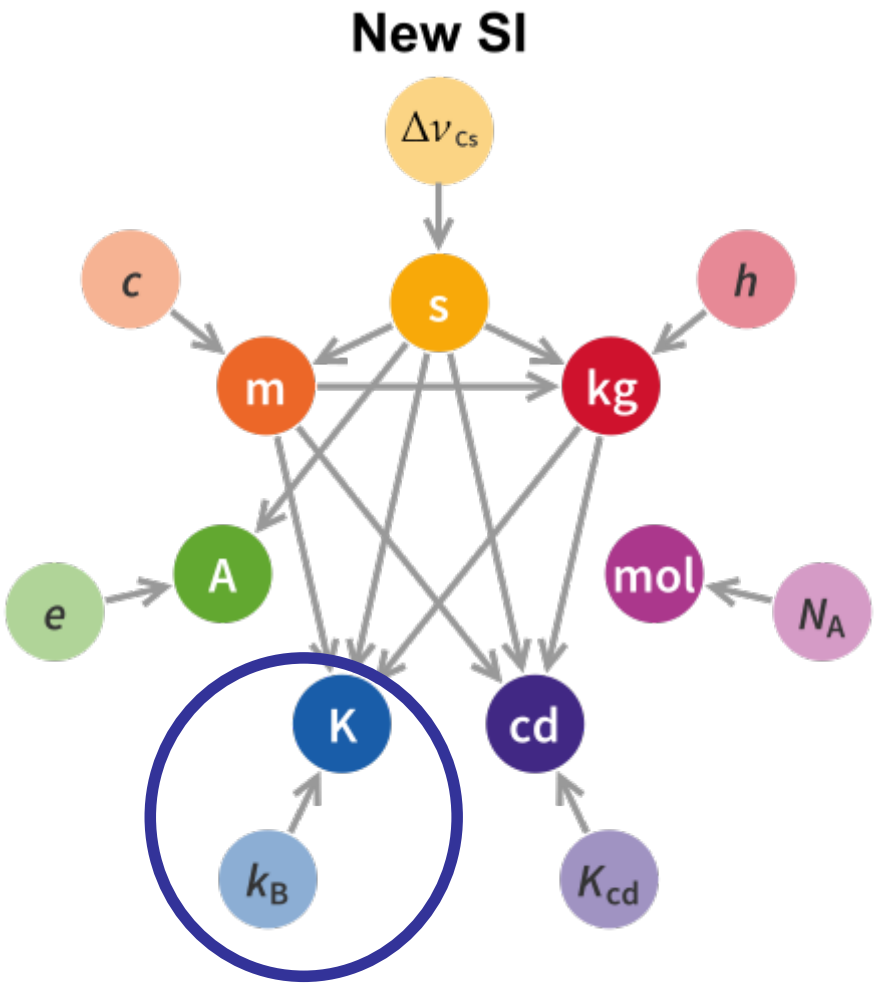
S=C=O

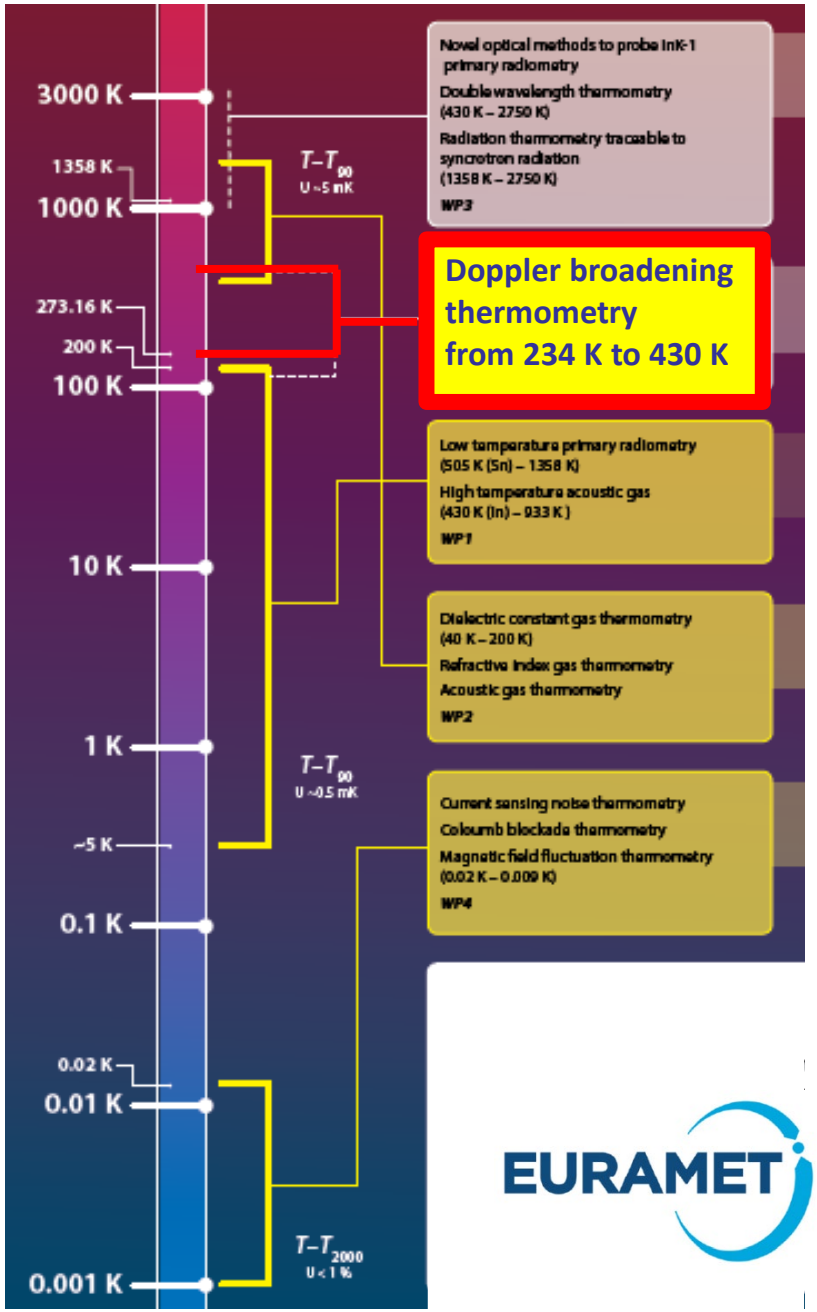


pressure ~10 Pa









Doppler Broadening Thermometry (DBT) as a novel primary method

Temperature range :
from the Hg triple point (234 K)
to In freezing point (430 K)

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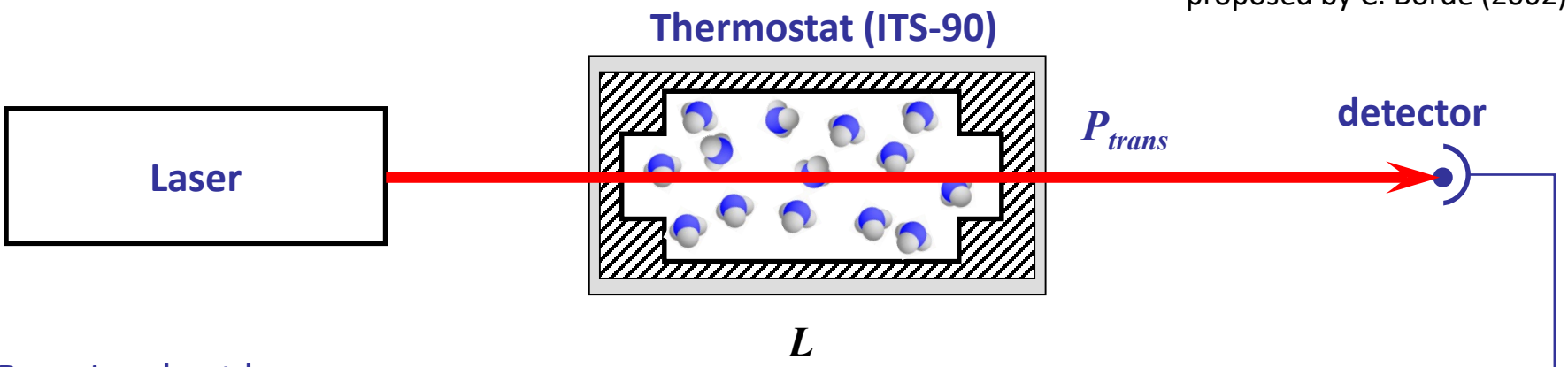
The Doppler Broadening Technique

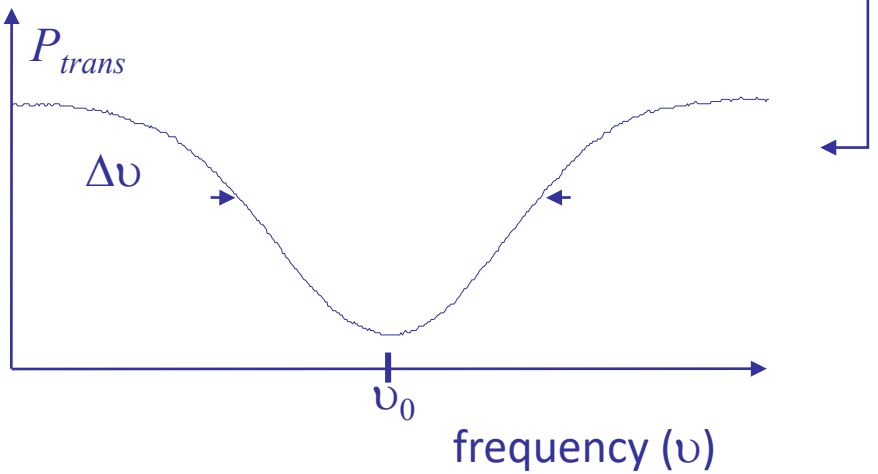
proposed by C. Bordé (2002)



Beer-Lambert law :

$$P_{trans} = P_{inc} exp(-\alpha(\upsilon)L)$$

$\alpha(\upsilon)$
- Doppler broadening
- collisional broadening
- Dicke narrowing
- hyperfine structure
- …



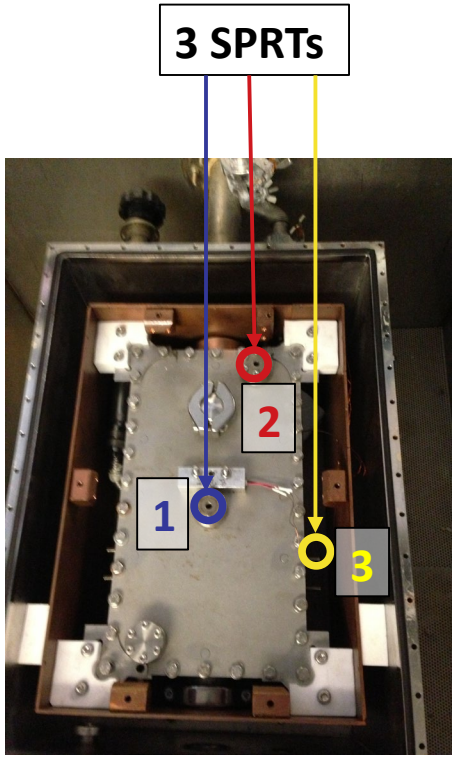
$$k_B T = \frac{m}{2}\left(\frac{\Delta\upsilon_D}{\upsilon_0} c\right)^2$$

record and model the absorption profile

once k_B fixed ⇒ extract T

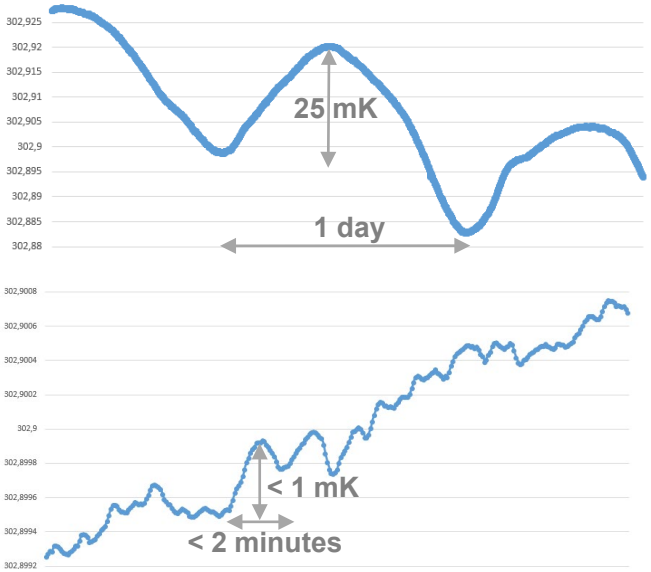
Thermostat characterization and ITS-90 measurement

Previously at AG FIRST-TF 2019!

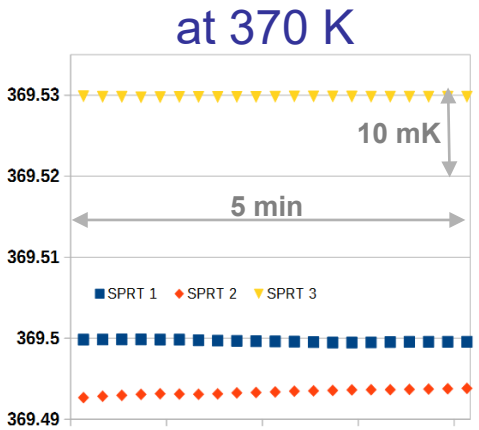


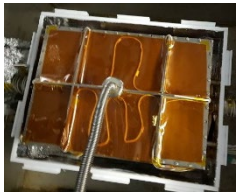
Stability measurements:

- <50mK over several days at 370K
- Short time fluctuations



Gradient measurements:





origin	uncertainty components	T = 303 K value, mK	T = 370 K value, mK	comments
cell temperature	cell homogeneity	**0.8**	**1.7**	gradient inside the cell
	cell temperature fluctuations	**10.1**	**14.4**	typical fluctuation over several days
calibration chain	SPRT calibration uncertainty	**0.4**	**1.0**	calibration with uncertainty propagation a, Ro
detection chain	measurement resolution	**negl.**	**negl**	negligible
	measurement repeatability	**0.5**	**0.5**	typical noise over 1 spectrum
	reference resistance calibration	**negl.**	**negl.**	negligible
	reference resistance stability	**0.1**	**0.1**	temperature fluctuations of the oil bath
	SPRT self-heating	**1.5**	**1.5**	not corrected for, uncertainty taken equal to the shift
Combined uncertainty k=1, mK		**10.3**	**14.7**	

few 10 ppm level

The frequency stabilized 10 µm source

DFB QCL @ 10.3 µm

Phase Lock Loop

carrier

8-18 GHz 8-18 GHz

frequency stabilised CO_2 laser

EOM

detector

- locked to a saturated absorption line of OsO_4
- < 10 Hz line width, < 100 Hz accuracy

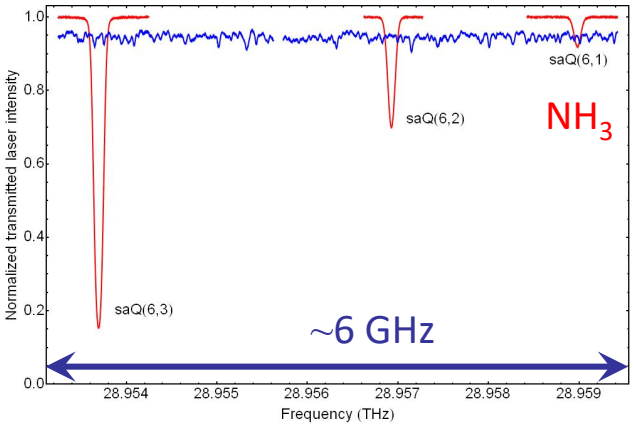
home-made 8-18 GHz synthesizer

- YIG oscillator
- Phase locked to a DDS

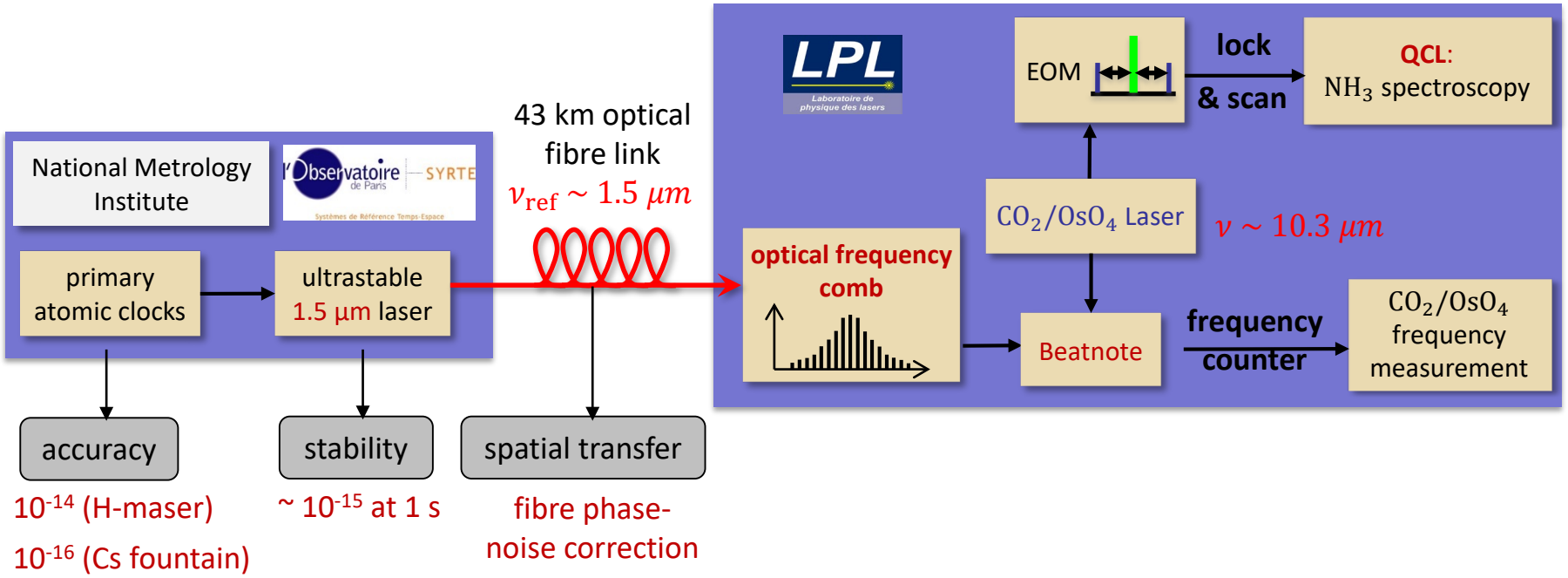
absorption cell

detector

NH_3



SI-traceability via a direct link to primary frequency standard



Traceability to the SI frequency standard:

- beatnote between CO_2 laser and comb
- count beatnote frequency

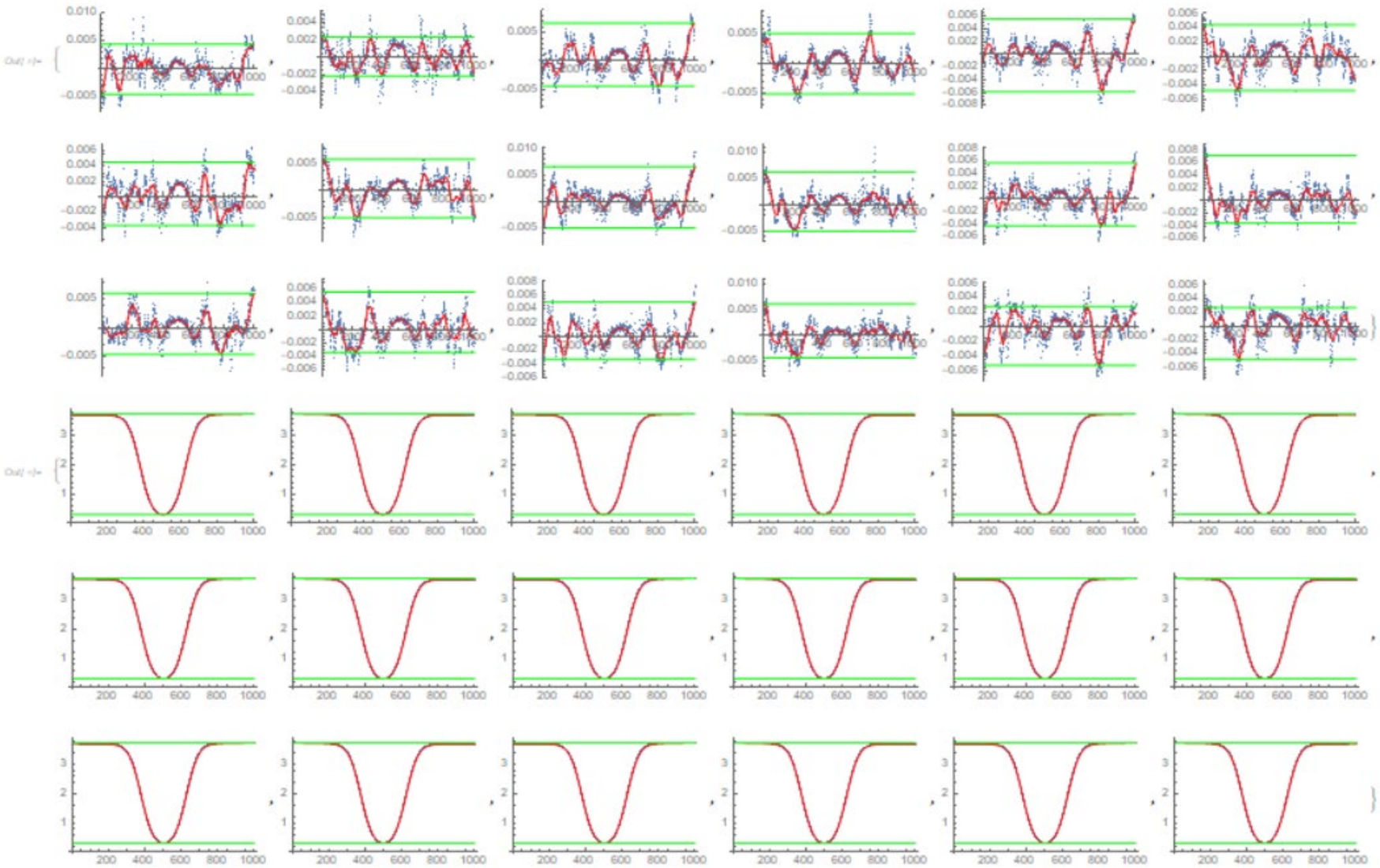
$$\nu_{OsO_4/CO_2} = \frac{n}{N}(\nu_{ref} + \Delta) + \text{Beatnote}$$

Uncertainty on the QCL absolute frequency:

- Sub-100 Hz contribution from the CO_2/OsO_4 laser
- <0.1 Hz contribution from the µW driving the EOM

~600 spectra recorded so far at 303 K and 10 Pa

preliminary analysis: Voigt profile fitted to the data



~600 spectra recorded so far at ~303 K and ~13 Pa

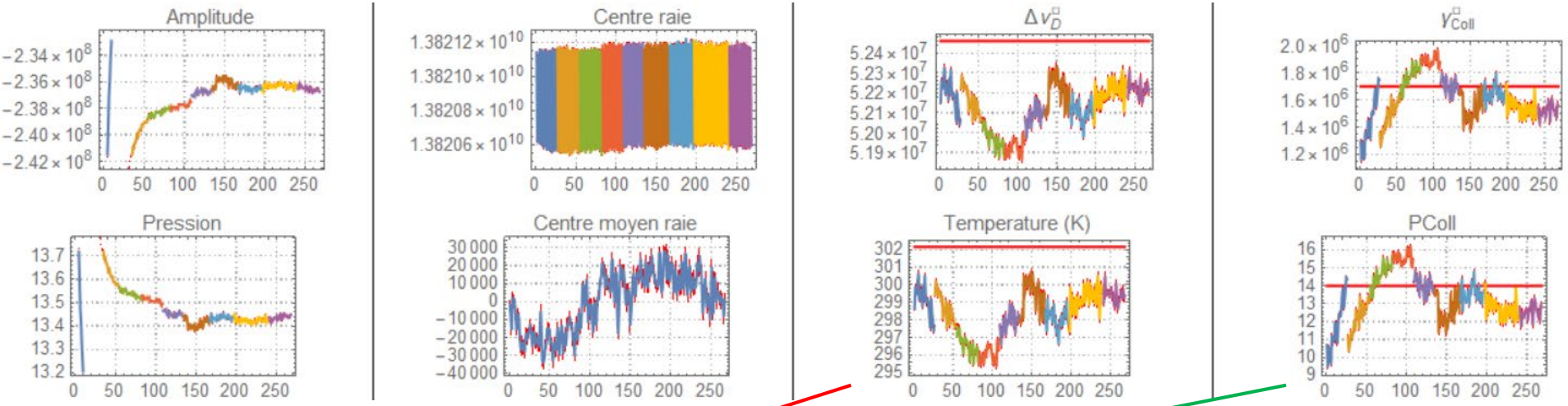
preliminary analysis: Voigt profile fitted to the data

integrated absorbance
↔ NH_3 partial pressure

centre frequency

Doppler broadening
↔ thermodynamic temperature

collisional broadening
↔ total temperature



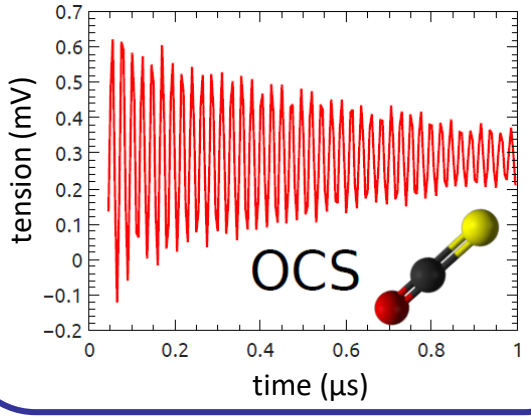
Correlations between Doppler and collisional broadening ↔ parasitic interference fringes

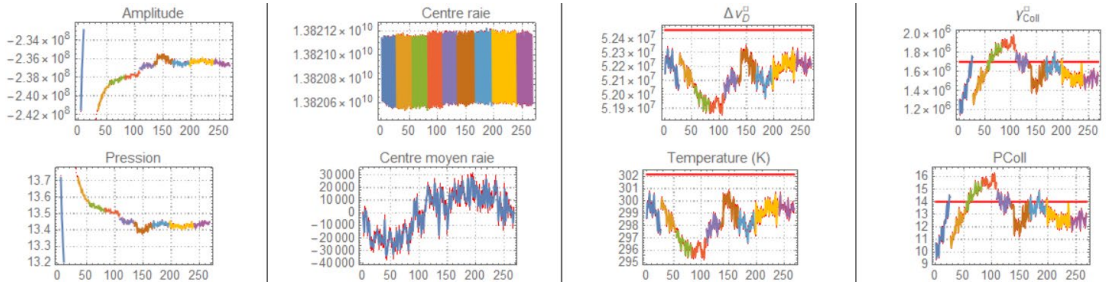
⟶ need spectra at other pressures (currently under progress)

⟶ full analysis yet to be done

sub-100 ppm temperature uncertainty expected

Summary





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