Stabilisation de QCL pour l'observation de la violation de parité dans les molécules

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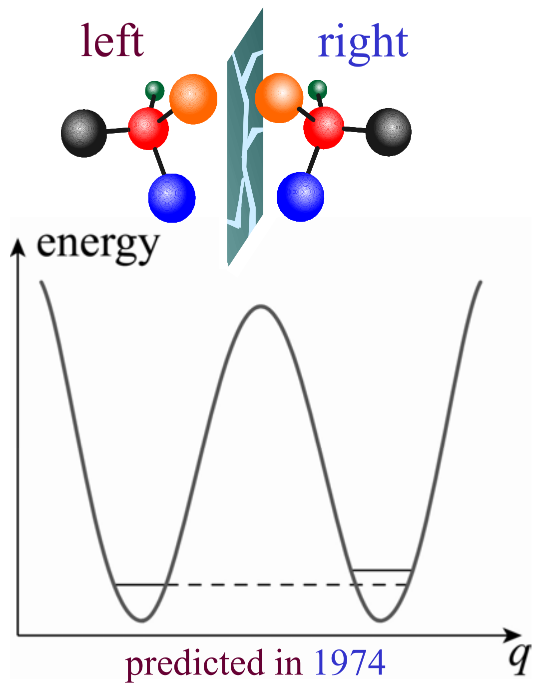
Parity, a broken symmetry

the weak nuclear force violates parity

- predicted by Lee and Yang (1956)
- 1st observation in β-decay of ^{60}Co (1957)
- emergence of the Standard Model (electro-weak theory, 1967)
- observed in high-energy physics
- observed in Cs (M.-A. Bouchiat, 1982 – C.E Wieman, 1997), effects $\propto Z^3$

never observed in chiral molecules



predicted in 1974
Rein, *J. Mol. Evol.* (1974)

→ in the long term: probe the Standard Model in the low-energy regime (enhanced effects $\propto Z^5$)… and physics beyond it

→ link to biomolecular homochirality

→ evaluate relativistic quantum chemistry

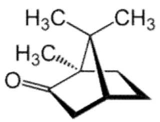
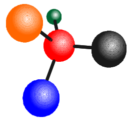
→ advanced manipulation techniques for polyatomic molecules

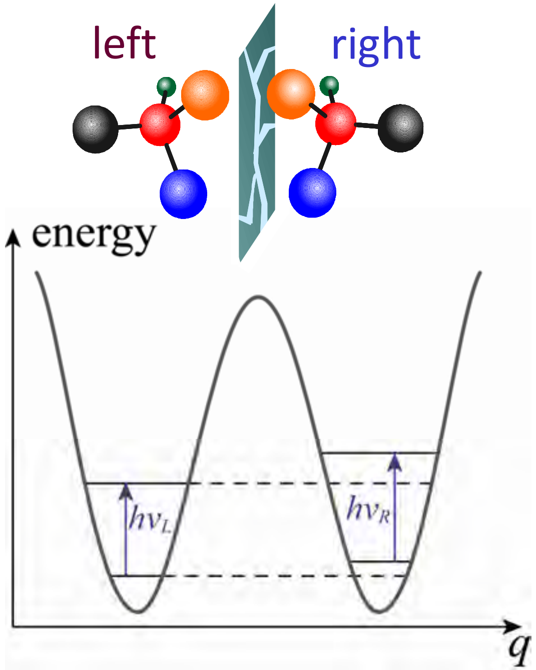
Parity violation in chiral molecules

several proposed experimental methods

- Lethokov's proposal (1975): vibrational spectroscopy (~30 THz)

The attempts so far…

molecule	experimental sensitivity	$\Delta\nu_{PV}^{calc}/\nu$
camphor 	10^{-8} (T. Oka, 1977)	3×10^{-19} (P. Schwerdtfeger, 2004)
CHFClBr 	2.5×10^{-13} (C. Chardonnet, 2002)	8×10^{-17} (P. Schwerdtfeger, 2005)



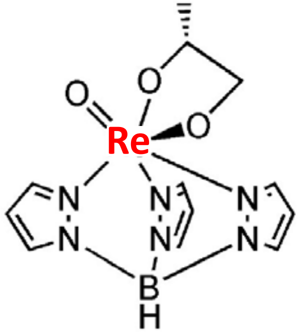
$$\Delta\nu_{PV} = |\nu_L - \nu_R| \neq 0$$

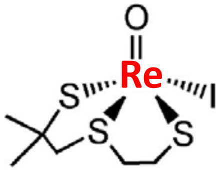
… have triggered a lot of theoretical work!

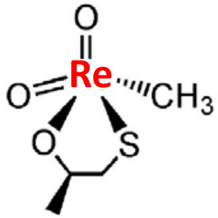
PV in chiral molecules: our strategy

Molecules with measurable PV:

Z_{Re} = 75







- $\Delta\nu_{PV} \sim 10^{-14} - 10^{-13}$ for the Re=O stretch of rhenium complexes
- synthesized but in solid form

Darquié et al, *Chirality* (2010)
Saleh et al, Phys. Chem. Chem. Phys. (2013)

Université Paul Sabatier TOULOUSE III
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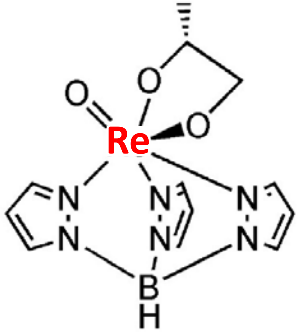
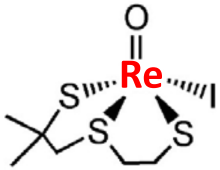
Centre for THEORETICAL Chemistry and Physics
P. Schwerdtfeger

UNIVERSITÉ DE RENNES 1
J. Crassous

PV in chiral molecules: our strategy

Molecules with measurable PV:





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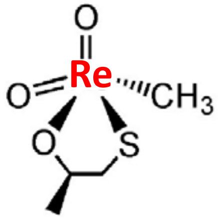

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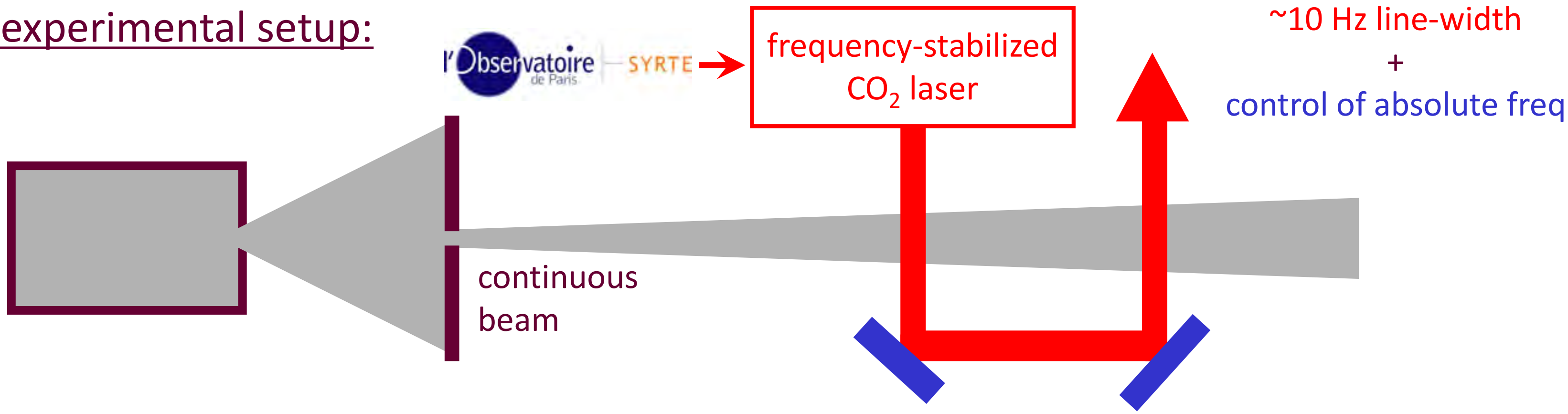
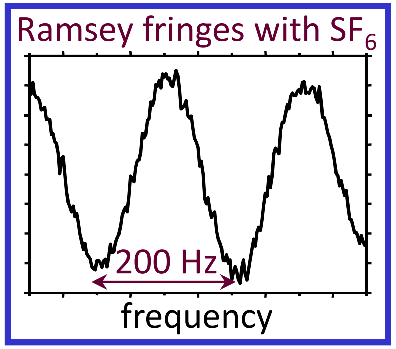
Current experimental setup:

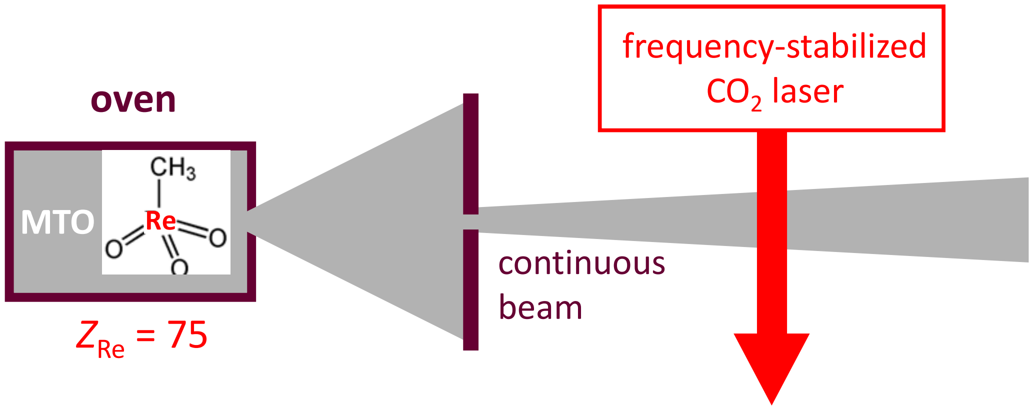


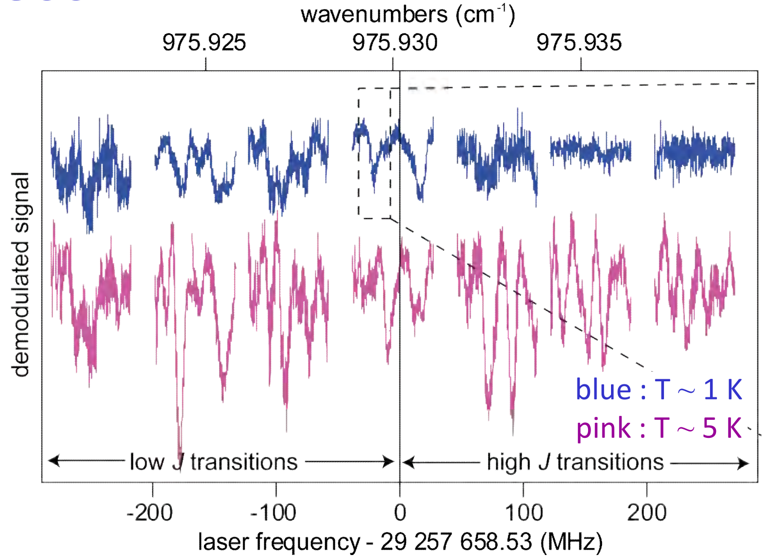
- continuous supersonic beam + CO_2 lasers + **Ramsey interferometer**
- uncertainty in determining line centre (with SF_6): 2×10^{-14}
- alternate (right/left) supersonic jet $\Rightarrow$ cancel out systematic effects



expected sensitivity for a differential measurement: < 10^{-15}

Recent progress





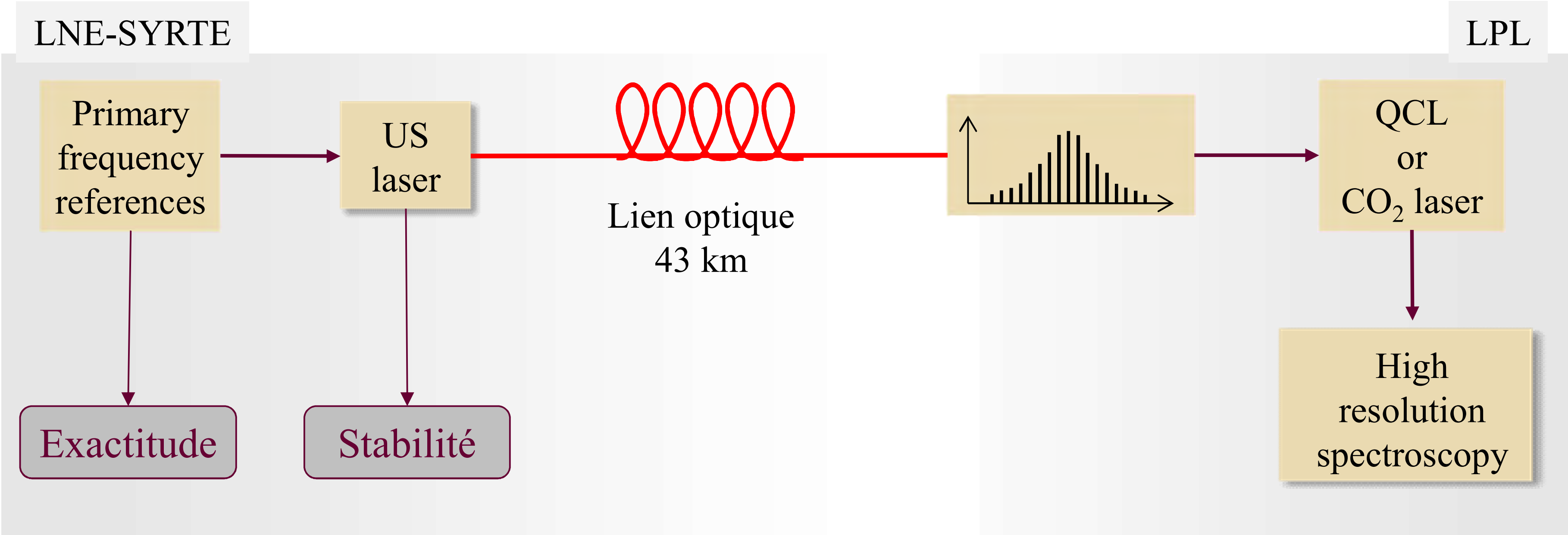
Tokunaga et al, *Mol. Phys.* (2013)

successfully created a continuous beam of MTO + high resolution spectroscopy

<u>Limitations of this experimental setup:</u>

1. weak flux ⟶ observation of Ramsey fringes difficult (rhenium complexes have little to no vapour pressure)
2. CO_2 lasers are not tunable enough
3. direct detection of mid-IR laser absorption is not sensitive enough

Transfert de la stabilité de fréquence du NIR => MIR



- Référence optique : cavité ultra-stable (US)
  - stabilité à 1 s de ~10^{-15}
  - dérive de fréquence ~Hz/s

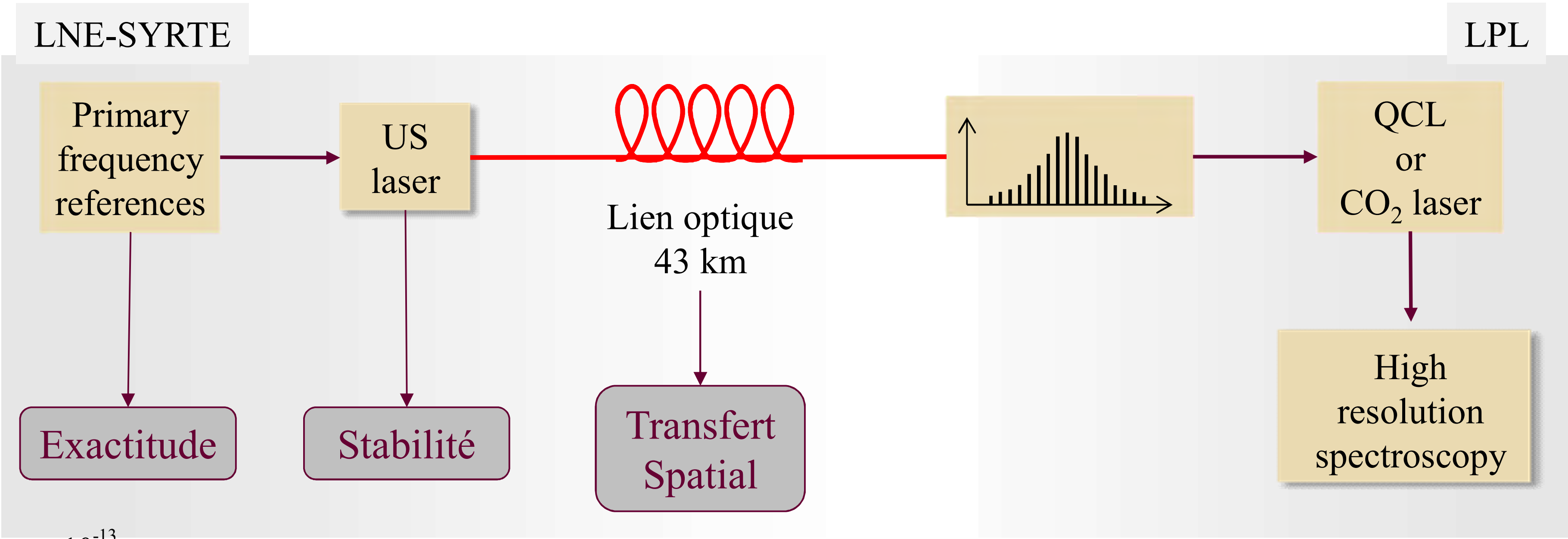
l'Observatoire de Paris — SYRTE
Systèmes de Référence Temps-Espace
Daniele Nicolodi
Michel Abgrall
Yann Le Coq

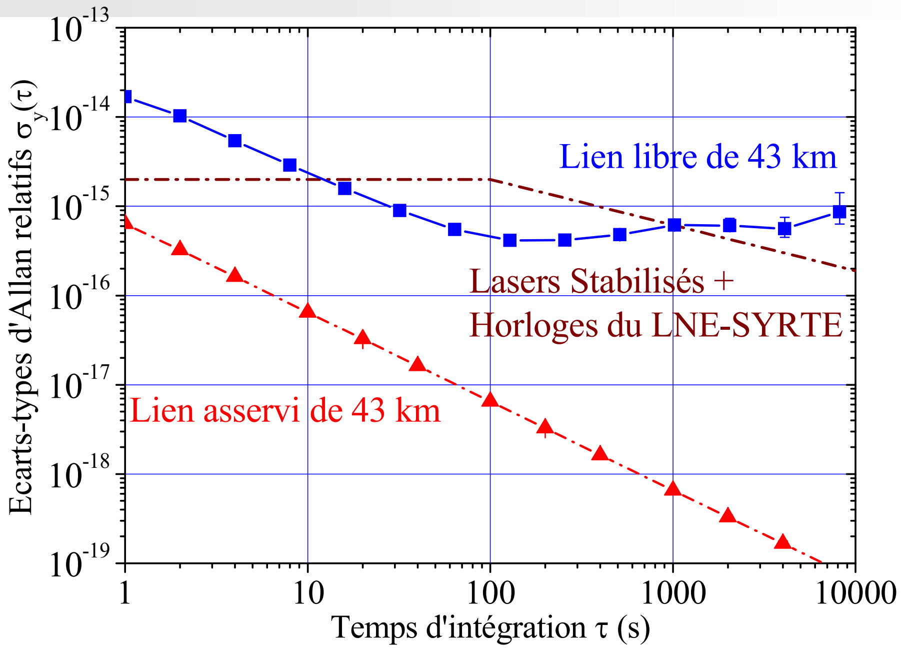
- Horloges primaires : maser H
  - Stabilité ~ $2.10^{-14}\,\tau^{-1}$
  - exactitude ~ 10^{-14} à 100s

=> Mesure de la fréquence absolue

=> Correction des dérives

Transfert de la stabilité de fréquence du NIR => MIR



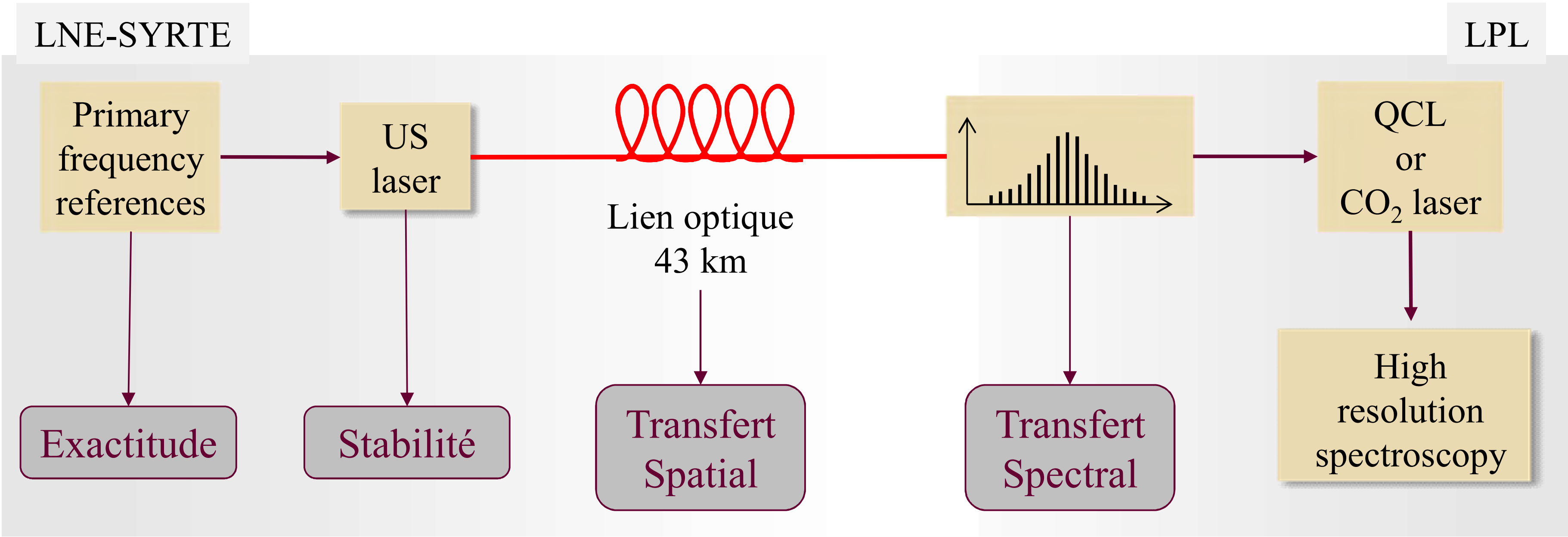


correction du bruit de phase permet le transfert du laser US sans dégradation de la stabilité

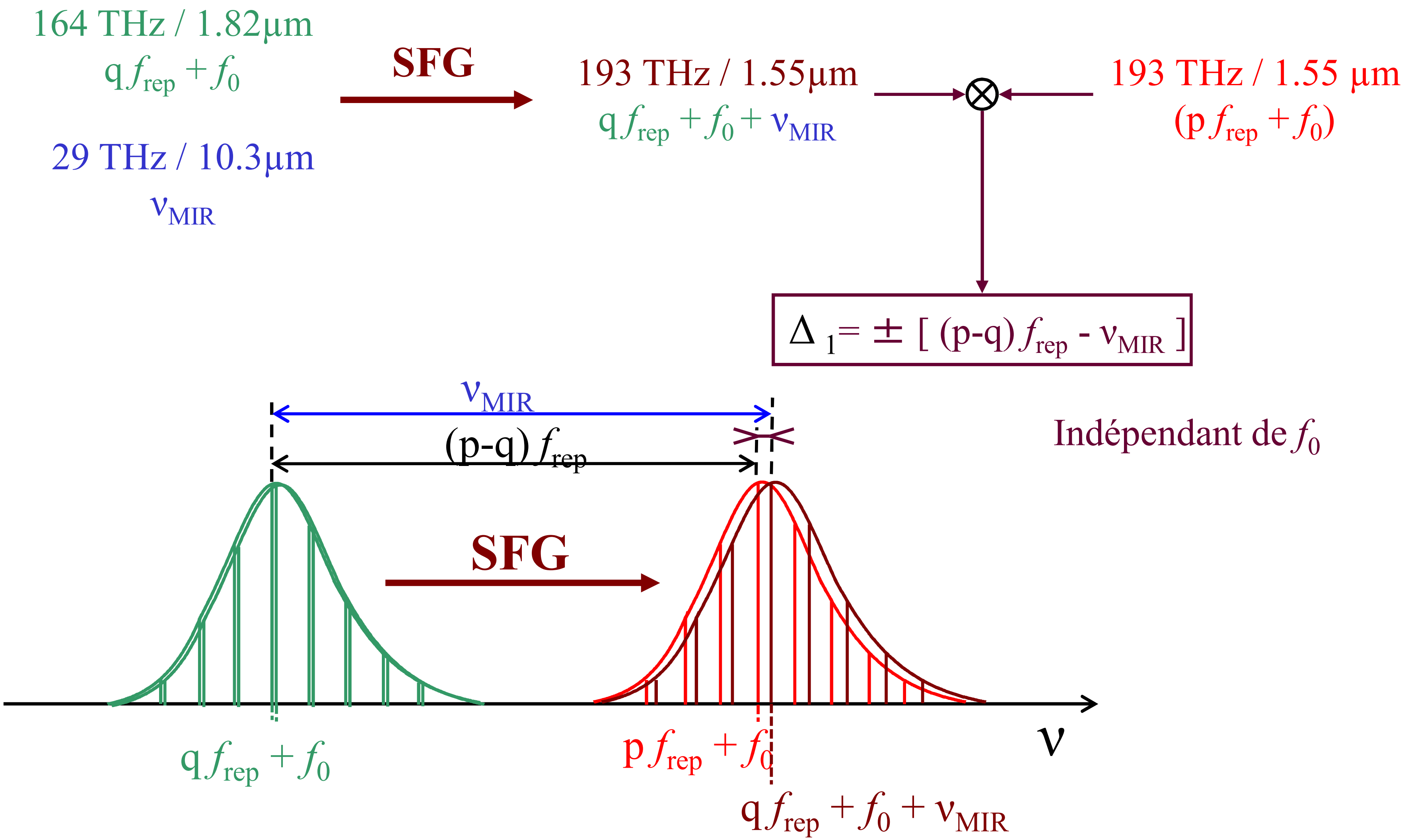
En collaboration avec G. Santarelli et P.-E. Pottie

Ecart-type d'Allan avec recouvrement
Compteur Π sans temps mort

Transfert de la stabilité de fréquence du NIR => MIR



Transfert de stabilité du NIR au MIR

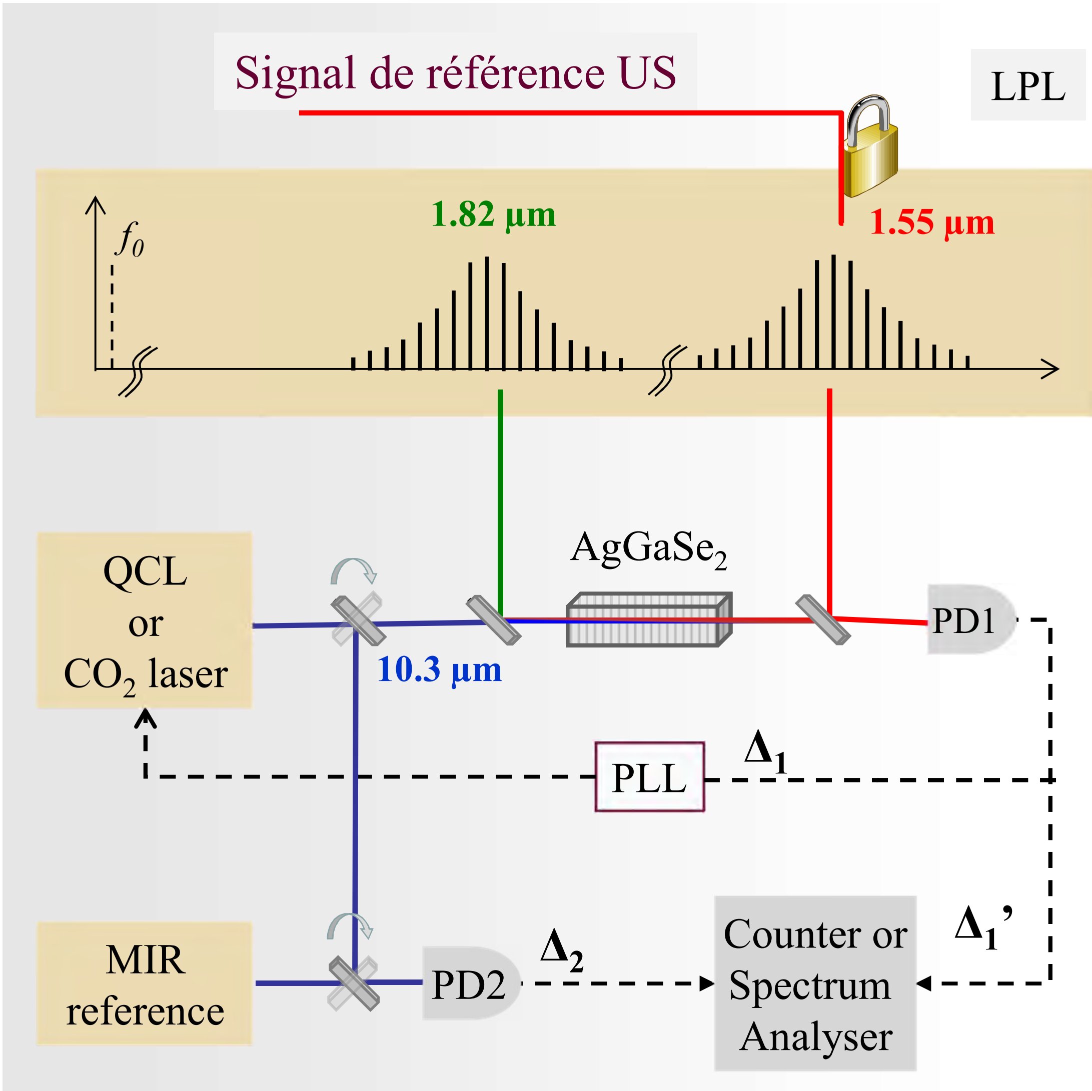


Stabilisation du QCL

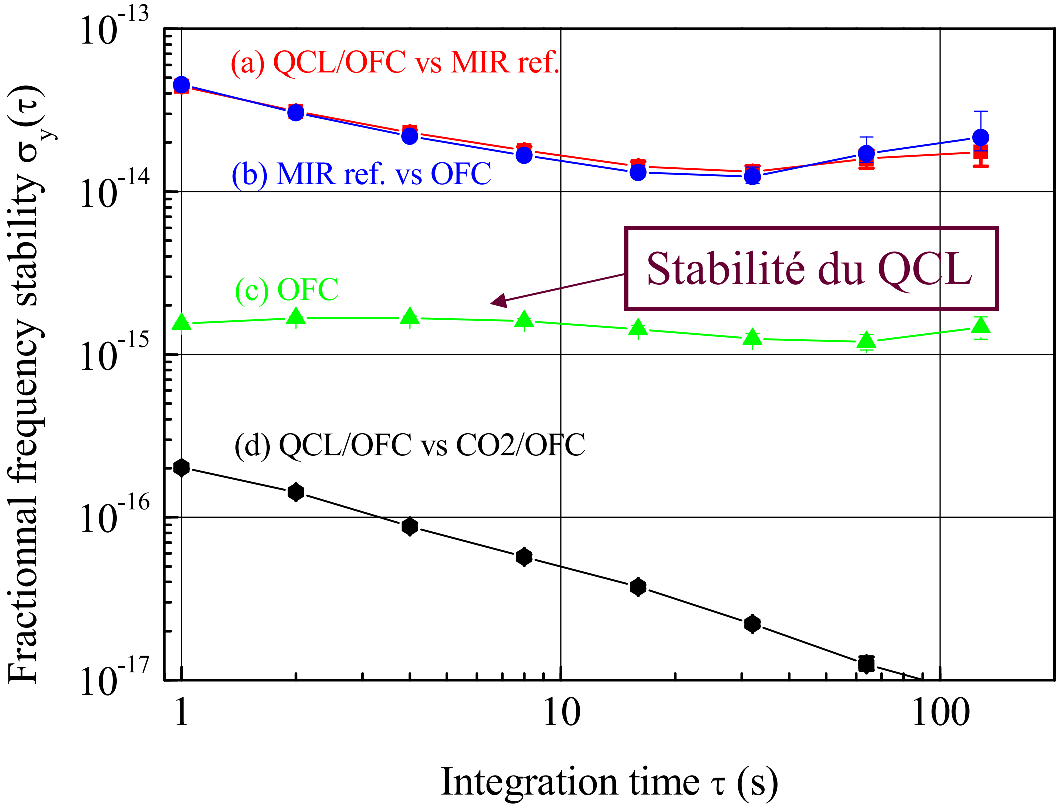
- Asservissement de f_{rep}
- Somme de fréquence
  - 30 nW
  - Δ_1 : asserv. QCL

  25 dB in 100 kHz RBW
- MIR référence :
  - Laser CO_2 stab. OsO_4
  - Stabilité : 5.10^{-14} à 1s

Argence et al, *Nature Phot* (2015)



Stabilité de fréquence - Ecart-type d'Allan



Stabilité de la référence MIR
=> limite haute de la stabilité du QCL
Chanteau et al, *New J. Phys.* (2013)

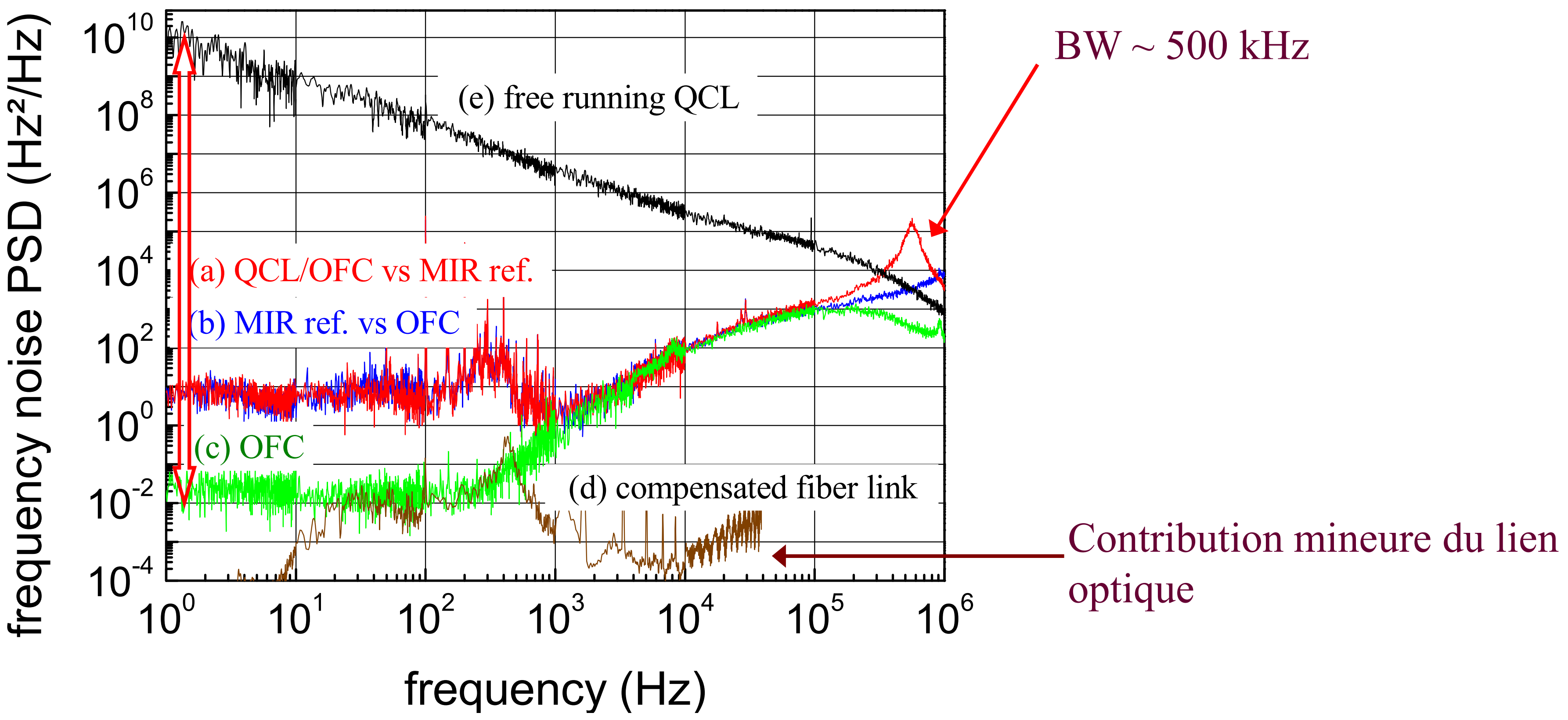
2[nd] laser US transmis via
même lien optique
=> mesure de la stabilité de l'OFC

Asservissement simultané
d'un QCL et d'un laser CO_2
=> mesure du bruit apporté par PLL
Compteur en Λ, Gate time de 1s
Excepté (d) compteur Π

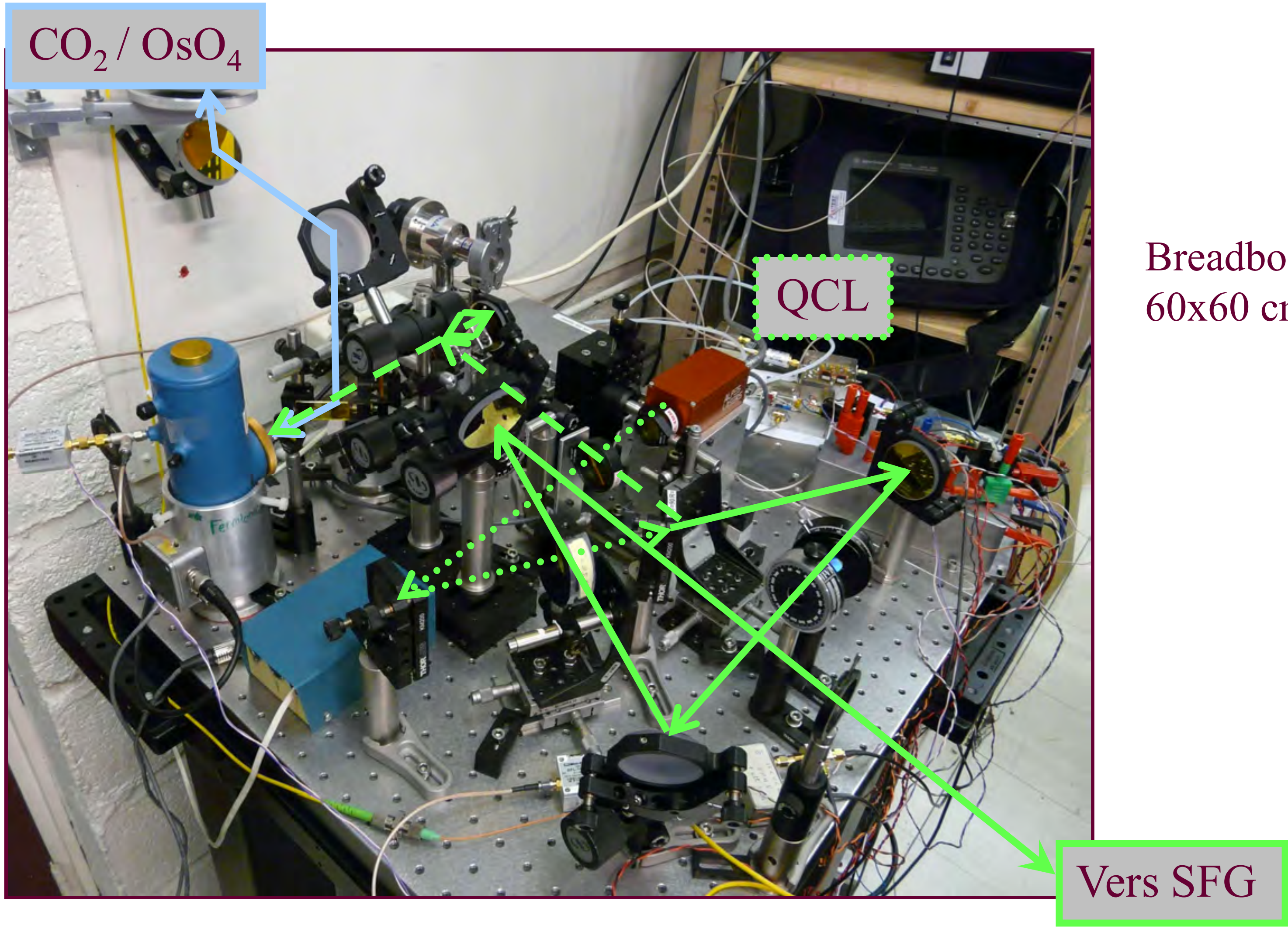
Gain de > 1 ordre de grandeur par rapport à l'état de l'art 10 Hz => 0,2 Hz

Stabilité de fréquence - PSD

Réduction du bruit ~ 12 ordres de grandeur à 1 Hz

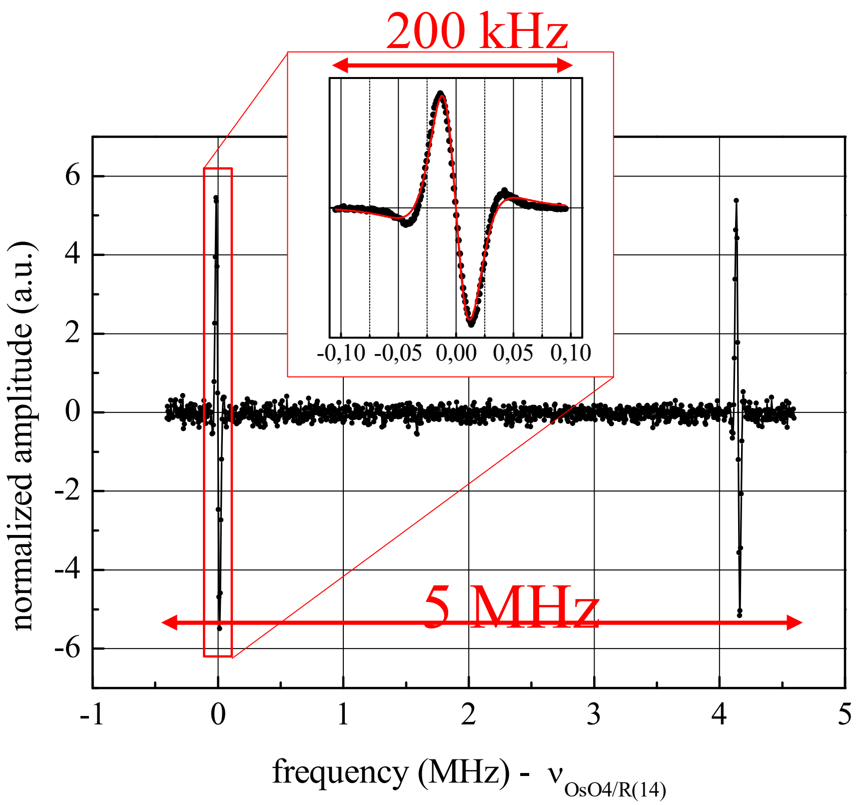


Photographie du dispositif



Breadboard
60x60 cm

Spectroscopie MIR ultra haute résolution



- Largeur de raie du QCL : 0,2 Hz

- 25 kHz pic-pic
- Incertitude sur le centre de raie ~10^{-12}

- Balayage PLL
  => 100 MHz à 10 µm

- Balayage d'un laser intermédiaire NIR équivalent à un balayage de f_{rep}
  => 1 GHz à 10 µm (6 GHz à 1,5 µm)

Table 1 | Absolute frequencies of five OsO_4 absorption lines in the vicinity of the R(14) CO_2 laser line.

OsO_4 lines in the vicinity of the CO_2 R(14) laser line at 10.3 µm	Frequency shift from $\nu_{OsO4/R(14)}$ calculated from refs 39 and 41 (kHz)	Frequency shift from $\nu_{OsO4/R(14)}$ measured in this work (kHz)
$^{190}OsO_4$ reference line (unassigned)	0.000 (40)	−0.009 (22)
Unreported line	–	+4,147.399 (23)
$^{190}OsO_4$, R(46)A_1^3(−)	+101,726.83 (5)	+101,726.821 (32)
Unreported line	–	+123,467.401 (32)
Unreported line	–	+204,269.162 (33)

The frequencies are given with respect to the OsO_4/CO_2-R(14) reference line frequency $\nu_{OsO4/R(14)}$ = 29,137,747,033.383 THz, reported in ref. 39. In the second column we report the absolute frequencies calculated from refs 39 and 41 with 1σ uncertainty. The third column displays the results of this work, where the uncertainty is the standard uncertainty of the mean. The R(46)A_1^3(−) line has previously been recorded at lower pressure[41]. Our measurement is thus expected to be pressure-shifted by approximately +10 Hz (ref. 26).

Conclusion

- Technique de stabilisation :
  - Transfert de la stabilité des meilleurs lasers ultra-stables du proche IR vers le moyen IR

  => mesures de précision sur les molécules

- Utilisation QCL vs CO_2 :
  - Lève la contrainte du laser CO_2 ou d'une référence moléculaire
  - Toute espèce absorbant entre 3 et 25 µm

  => Augmentation considérable du nombre de molécules candidates potentielles

NCPChem (2011-2014), LIOM (2012-2014), ASTRID QUIGARDE (2013-2015), PVCM (2015-2019)