Frequency noise and phase-locking of a QCL-pumped, 1THz gas laser using a 1560nm frequency comb

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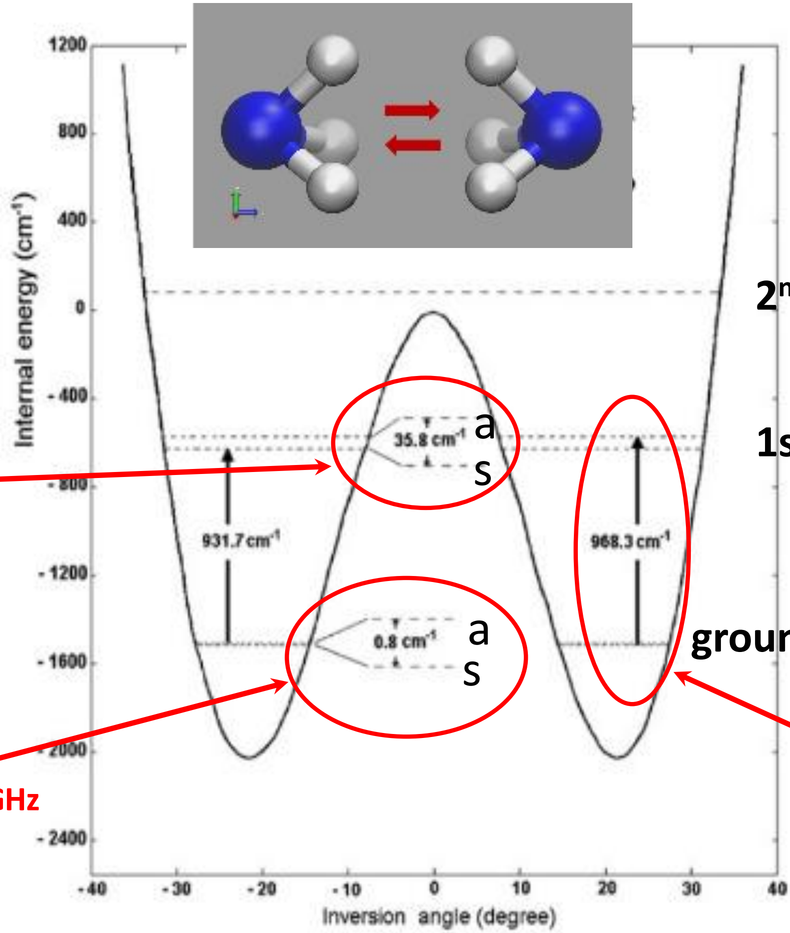
Outline

- Operation and performance of mid-IR QCL-pumped, ammonia laser at 1THz
- Measurement of frequency noise using a fs-laser comb
- Comparison with CO_2-laser pumped molecular laser at 2.5THz
- Phase locking to a fs-laser frequency comb
- Conclusions

mid-IR QCL-pumped, ammonia laser ($NH_{3)}$) at 1THz

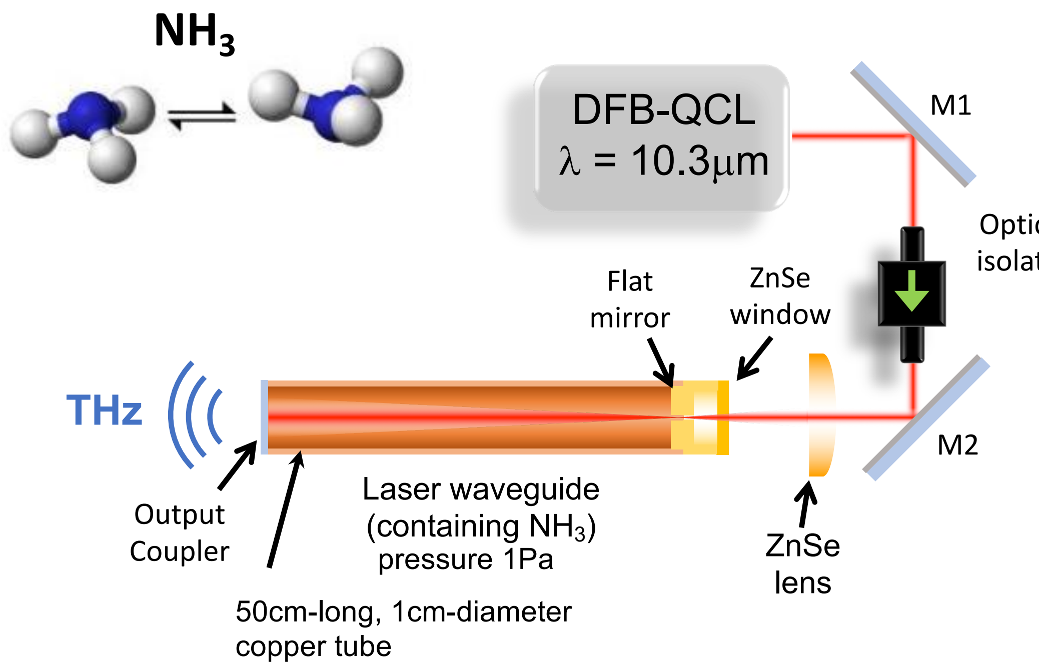
QCL-pumped FIR molecular laser at 1THz

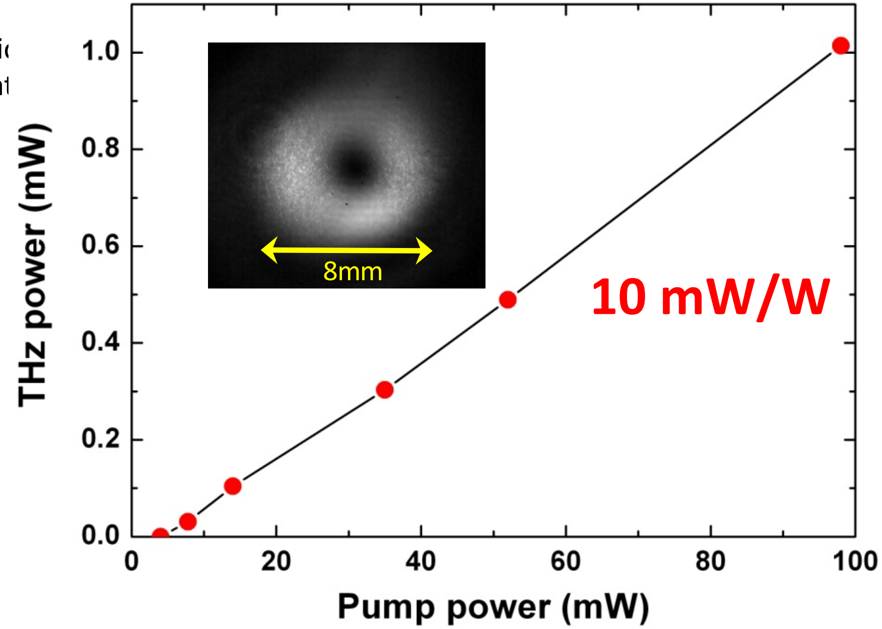
NH_3 vibration-inversion potential



- The pump *saQ(3,3)* transition **is NOT reachable by a CO_2 laser**
- Both the MIR and THz transitions present **high dipole moments of 0.2D and 1.05D**
- Fast non-radiative relaxation rate: *s(3,3) 1^{st} excited→ s(3,3) ground state*

The first QCL-pumped FIR molecular laser at 1THz





A. Pagies *et al.*, APL Photonics, **1**, 031302 (2016)
M. Micica *et al.*, Opex, **26**, 21242 (2018)

Maximum theoretical efficiency: 18.5 mW/W

- **55% of max. efficiency (= ½ ν_{THz}/ $\nu_{mid\text{-}IR}$) reached**
- need to optimize the design to increase further the output power

NH_3 : a large number of laser transitions in the THz range

In collaboration with: Laboratoire LPCA, Université du Littoral Cote d'Opale – Laboratoire ISMO, Université Paris Sacaly, SOLEIL synchrotron

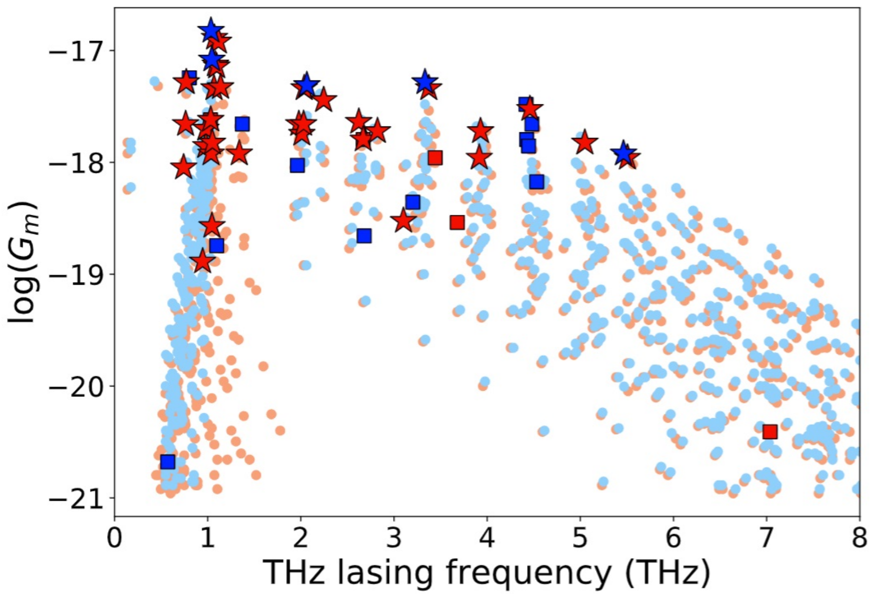
Table S3 – continued from previous page

THz laser		IR pump											
		P				Q				R			
Freq. (GHz)	QN	Freq. (cm^{-1})	QN	Pola.	G_m (uG_m)	Freq. (cm^{-1})	QN	Pola.	G_m (uG_m)	Freq. (cm^{-1})	QN	Pola.	G_m (uG_m)
1138.2110	sQ(7,7)	805.7790	sP(8,7)	⊥	3.9e-19	964.4240	sQ(7,7)	‖	4.7e-18	-	-	-	-
1166.1643	sQ(8,8)	784.9805	sP(9,8)	⊥	2.2e-19	963.3627	sQ(8,8)	‖	3.3e-18	-	-	-	-
1171.5755	sQ(13,12)*	680.8321	sP(14,12)	⊥	1.1e-20	956.3968	sQ(13,12)	‖	1.7e-19	1213.4613	sR(12,12)	⊥	4.3e-20
1199.1625	sQ(9,9)	764.0774	sP(10,9)	⊥	2.4e-19	962.1714	sQ(9,9)	‖	4.4e-18	-	-	-	-
1209.9839	sQ(14,13)*	659.6231	sP(15,13)	⊥	2.2e-21	954.5783	sQ(14,13)	‖	4.4e-20	1231.2408	sR(13,13)	⊥	9.6e-21
1237.4652	sQ(10,10)	743.0731	sP(11,10)	⊥	6.0e-20	960.8522	sQ(10,10)	‖	1.3e-18	-	-	-	-
1253.6581	sQ(15,14)*	-	-	-	-	952.6354	sQ(15,14)	‖	1.7e-20	1248.8648	sR(14,14)	⊥	4.0e-21
1281.3702	sQ(11,11)	721.9708	sP(12,11)	⊥	2.8e-20	959.4071	sQ(11,11)	‖	7.2e-19	-	-	-	-
1302.8895	sQ(16,15)*	-	-	-	-	950.5708	sQ(16,15)	‖	1.3e-20	1266.3350	sR(15,15)	⊥	3.2e-21
1331.2132	sQ(12,12)	700.7745	sP(13,12)	⊥	2.6e-20	957.8390	sQ(12,12)	‖	7.5e-19	-	-	-	-
1338.6790	aR(3,3)	832.6348	aP(5,3)	‖	1.1e-18	931.7736	aQ(4,3)	⊥	2.1e-18	1011.2036	aR(3,3)	‖	1.2e-18

Small part (>1000 laser lines!)

Based on HITRAN database

M.-H. Mammez *et al.*, Submitted



Blue: $^{15}NH_3$
Red: $^{14}NH_3$

0.7-5.5 THz

Other recent works

RESEARCH

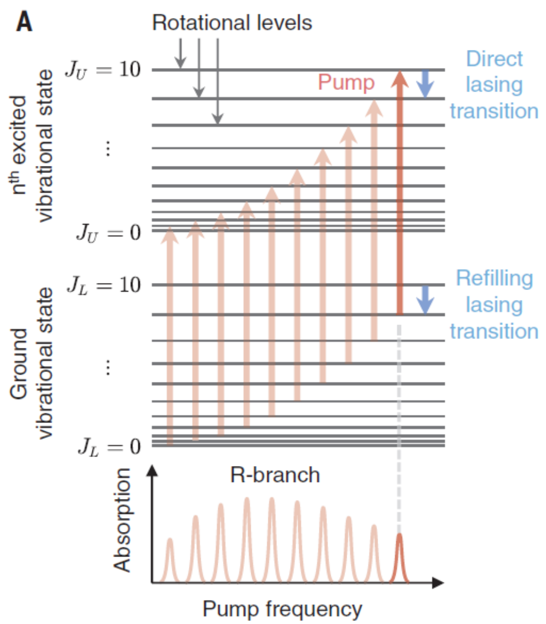
REPORT

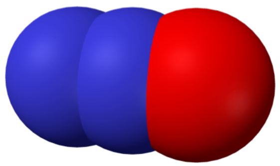
LASER PHYSICS

Widely tunable compact terahertz gas lasers

Paul Chevalier[1], Arman Armizhan[1], Fan Wang[2], Marco Piccardo[1], Steven G. Johnson[3,4], Federico Capasso[1]*, Henry O. Everitt[5,6]*

The terahertz region of the electromagnetic spectrum has been the least utilized owing to inadequacies of available sources. We introduce a compact, widely frequency-tunable, extremely bright source of terahertz radiation: a gas-phase molecular laser based on rotational population inversions optically pumped by a quantum cascade laser. By identifying the essential parameters that determine the suitability of a molecule for a terahertz laser, almost any rotational transition of almost any molecular gas can be made to lase. Nitrous oxide is used to illustrate the broad tunability over 37 lines spanning 0.251 to 0.955 terahertz, each with kilohertz linewidths. Our analysis shows that laser lines spanning more than 1 terahertz with powers greater than 1 milliwatt are possible from many molecular gases pumped by quantum cascade lasers.

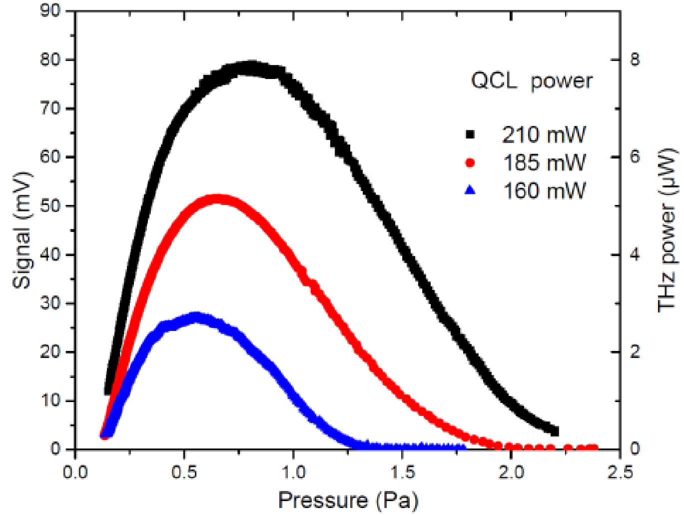




N_2O

P. Chevalier *et al.* Science **366**, 856 (2019)

$^{15}NH_3$



M. Wienold *et al.* Optics Express **28**, 23114 (2020)

QCL-pumped *vs* CO_2 pumped THz lasers

iemn

Why pumping with a QCL ?

A comparison with CO_2 laser-pumped FIR lasers:

Advantages:
- Compactness
- Low power consumption
- *Continuous tunability*

Drawbacks:
- Lower power
- More sensitive to feedback

↓

1) Can pump ***light molecules*** with **high dipolar moment**:
   → High gain
   → Fast relaxation
   → Frequencies OK for applications
2) Can access frequencies previously not accessible with CO_2 lasers

☺

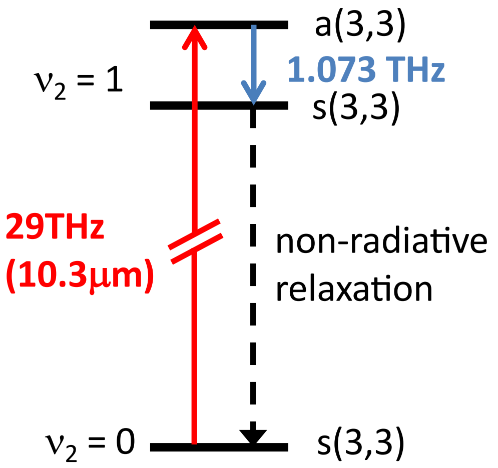
What about the spectral purity??

Mid-IR QCLs linewidth (1MHz–10MHz) typically ~10^3 times larger that CO_2 lasers (1kHz-100kHz)

↓

Larger linewidth (higher frequency noise) expected

MIR → THz modulation transfer



At 1Pa transitions are Doppler broadened:

$$f = \left(1 + \frac{v}{c}\right) f_0 \;\rightarrow\; f_{THz},\; f_{MIR}$$

$$\rightarrow \quad \delta f_{THz} = \delta f_{MIR} \times \left(\frac{f_{THz}}{f_{MIR}}\right)$$

Doppler-induced
Modulation transfer

Pump transition
Doppler broadening

- **NH_3 linewidth:** $$\Delta f_{THz} = \Delta f_{MIR} \times \frac{f_{THz}}{f_{MIR}} \sim 90MHz \times \frac{1\ THz}{30\ THz} \sim 3MHz$$

see → M. Micica *et al.*, Opex, **26**, 21242 (2018)

- **THz frequency noise:** $$\delta f_{THz} = \delta f_{QCL} \times \frac{f_{THz}}{f_{MIR}}$$

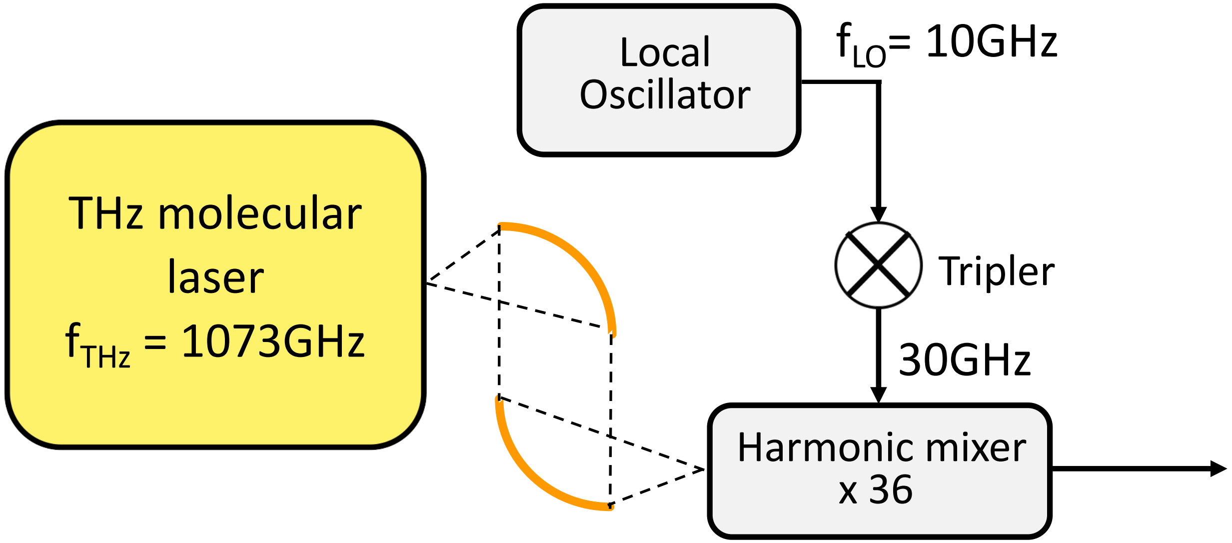
Noise limit from QCL driver: $\delta f_{THz} \sim 3 \frac{Hz^2}{Hz}$

obtained from:

- QCL tuning coeff. $\frac{\Delta f}{\Delta I} = 180 \frac{GHz}{A}$
- Current driver noise $300 \frac{pA}{Hz^{1/2}}$

Measurement of frequency noise using a fs-laser comb

Beatnote generation using a harmonic mixer



$f_{beat} = (3 \times 36) \times f_{LO} - f_{THz} =$

$= (1080 – 1073)$ GHz = 7GHz

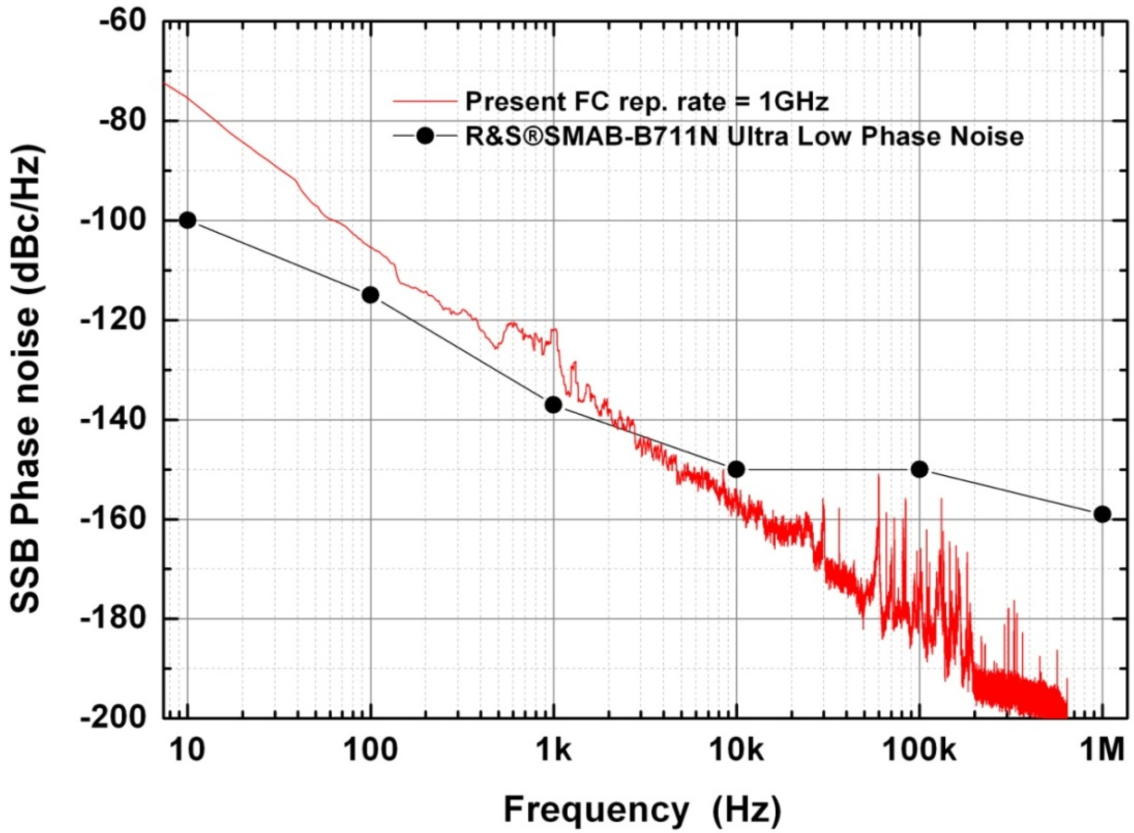
$$|\delta f_{beat}|_{rms} = 108 \times |\delta f_{LO}|_{rms} + |\delta f_{THz}|_{rms}$$

- The frequency noise (FN) of f_{beat} is a copy of the FN of f_{THz}

if $108 \times \delta f_{LO} << \delta f_{THz}$

→ Need a low phase noise LO (δf_{LO} is multiplied by 108!)

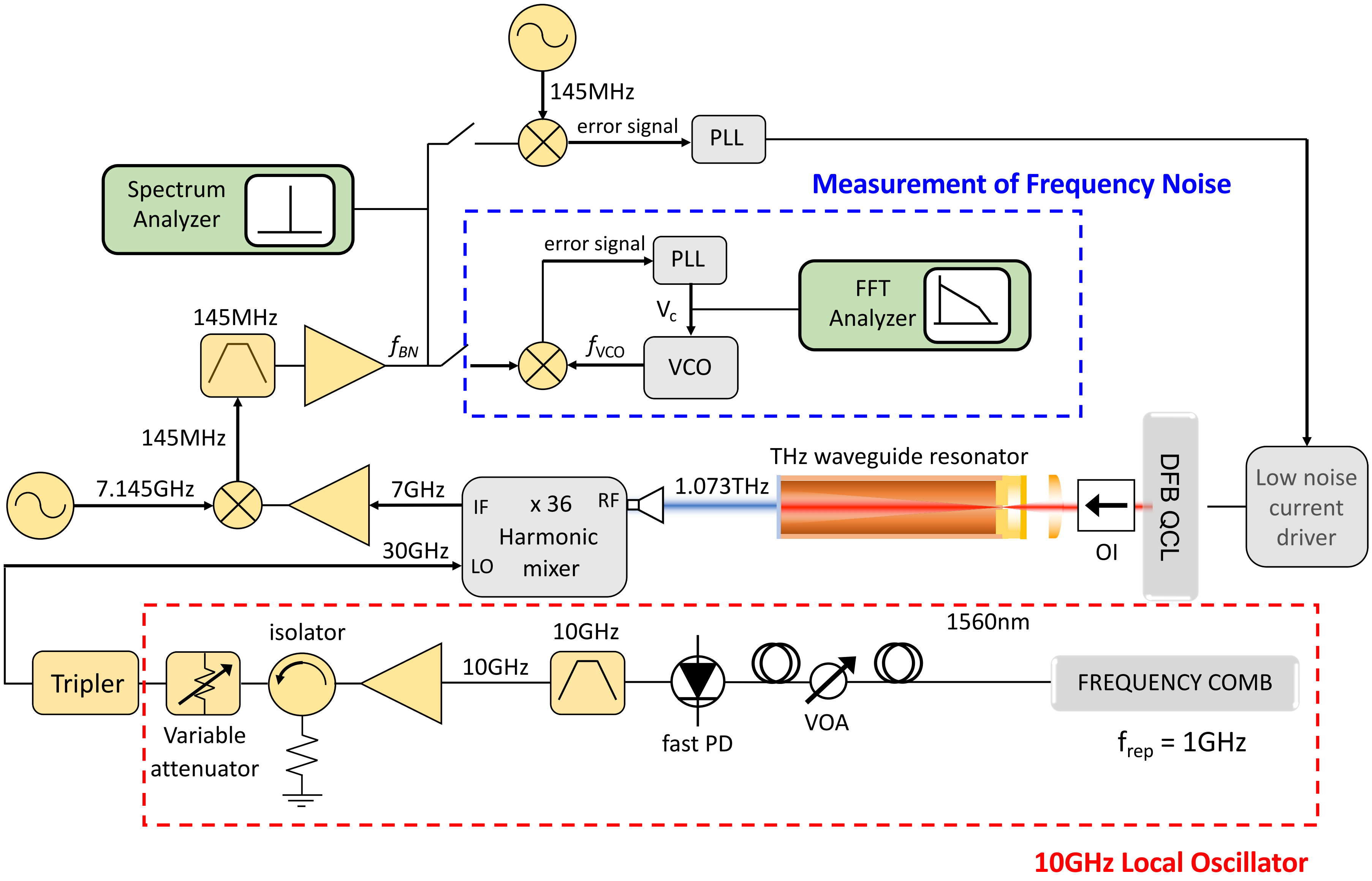
Phase noise of fs-laser comb repetition rate



- Harmonically mode-locked 1560nm laser comb, with 1GHz repetition rate
- Phase noise at 1GHz extrapolated from a beating with a fiber laser (195THz) stabilised on a high-finesse cavity

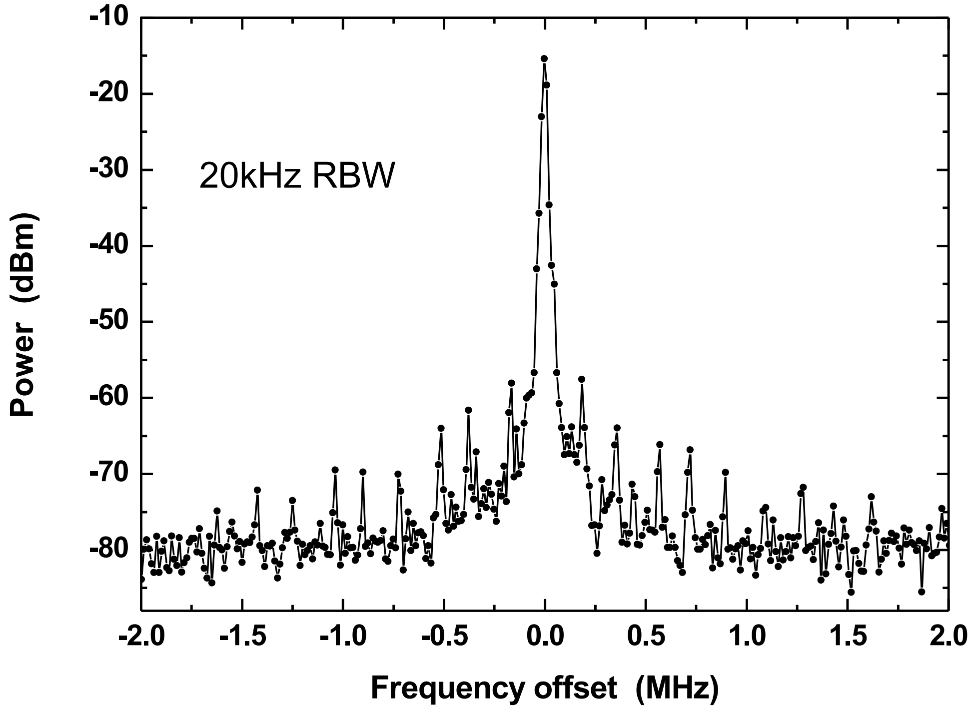
Very low phase-noise fundamental oscillator → suitable for our experiment

Experimental setup: measurement of frequency noise PSD



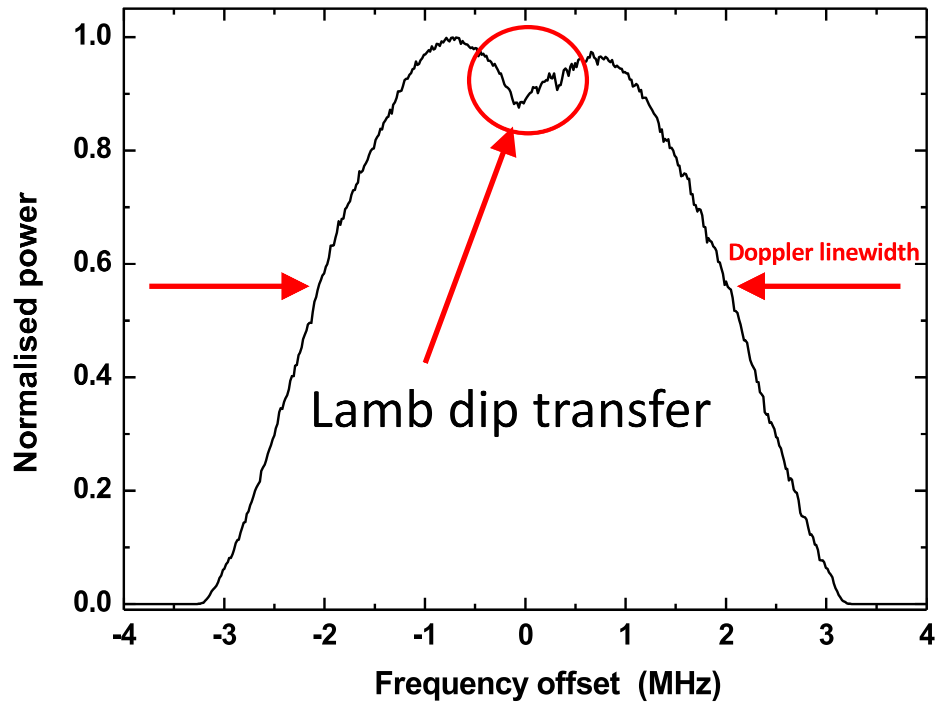
Beatnote spectra

Single-shot



Max hold spectrum

Obtained by scanning the QCL frequency across the pump transition



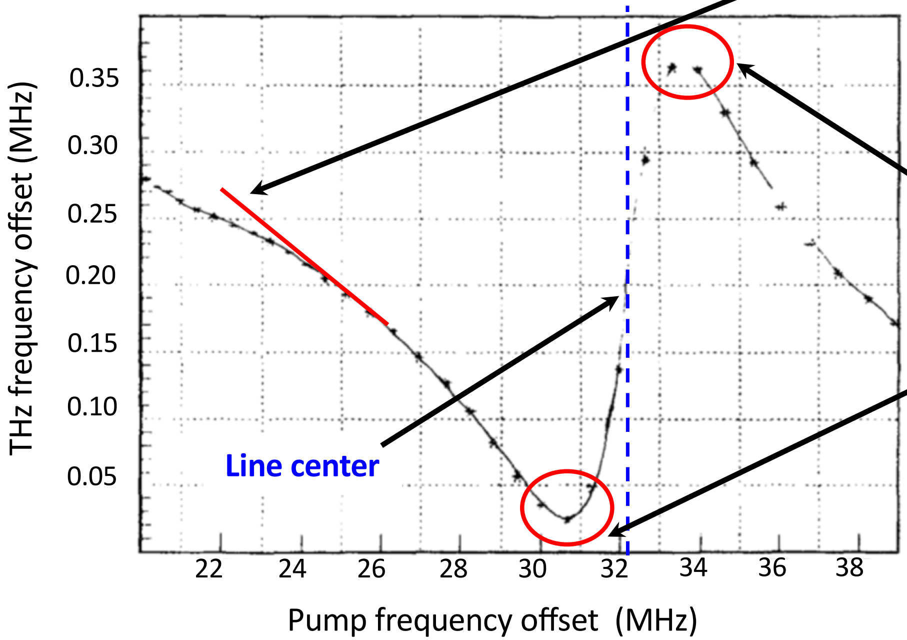
- <u>Lamb dip transfer</u>: well known effect in THz molecular lasers produced by stronger saturation of the mid-IR transition when the QCL is pumping at the line center

See for example T. A. De Temple *et al.*, J. IR MM Waves, **7**, 1-41 (1983)

Effect of Lamb-dip transfer on mid-IR → THz modulation transfer

Doppler limited: $\left|\frac{\delta f_{THz}}{\delta f_{mid-IR}}\right| = \left|\frac{f_{THz}}{f_{mid-IR}}\right| = 0.025$

0.73THz line of formic acid

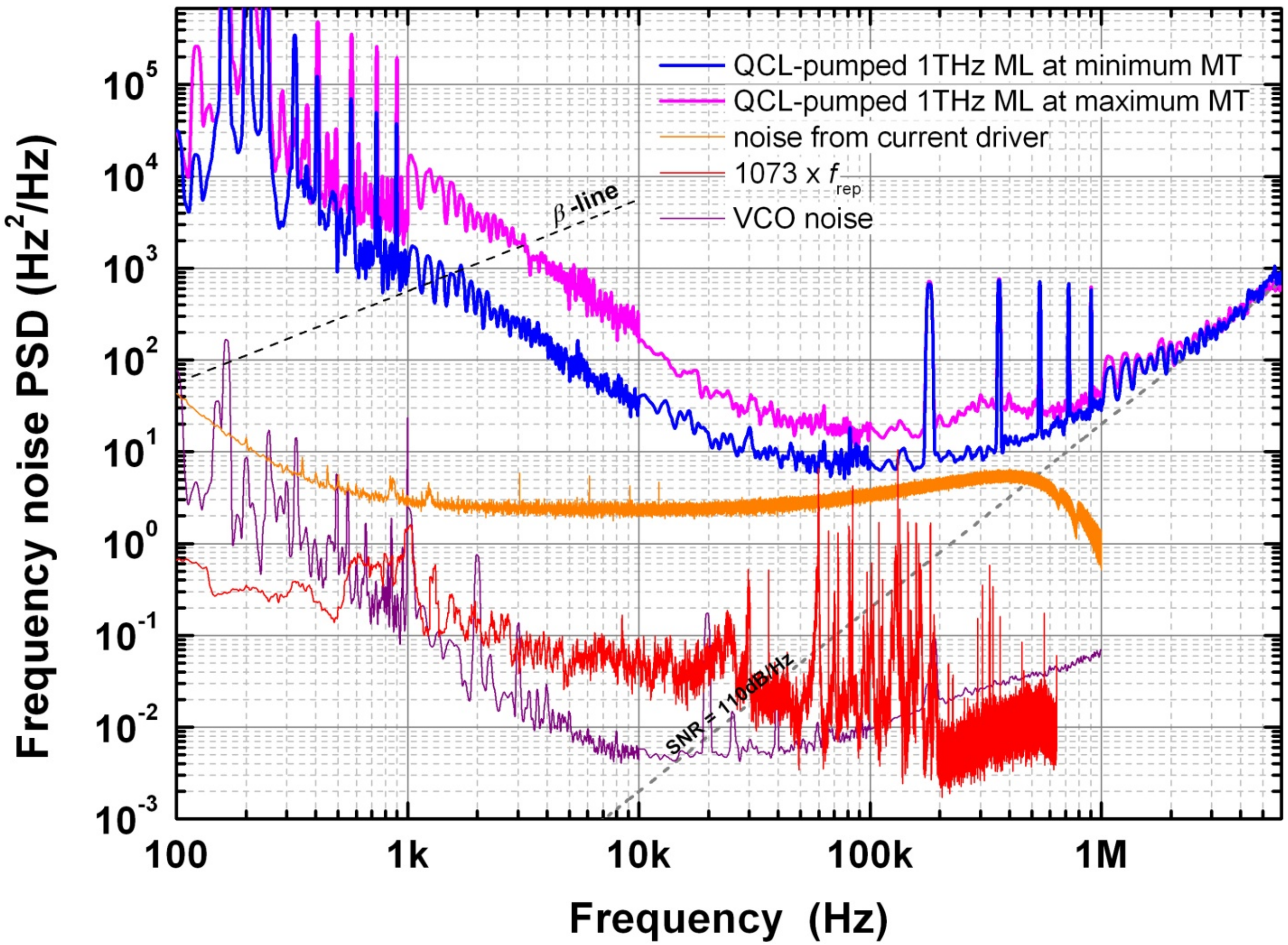


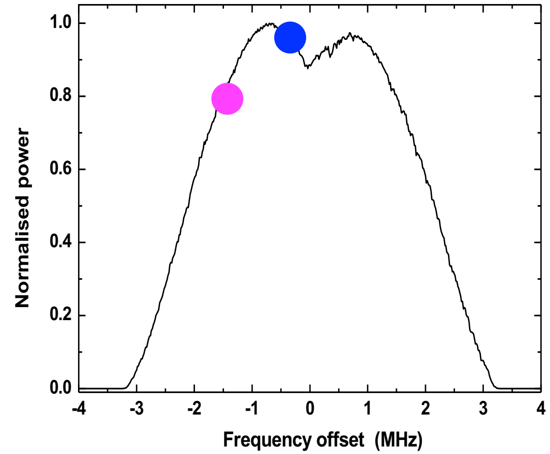
$\left|\frac{\delta f_{THz}}{\delta f_{mid-IR}}\right| \approx 0$

B. Dahmani *et al.*, Int. J. IR MM Waves, **5**, 1052 (1984)

- Close to the center of the line → 2 points of minimum noise-transfer
- Away from line center → noise-transfer ruled by Doppler effect

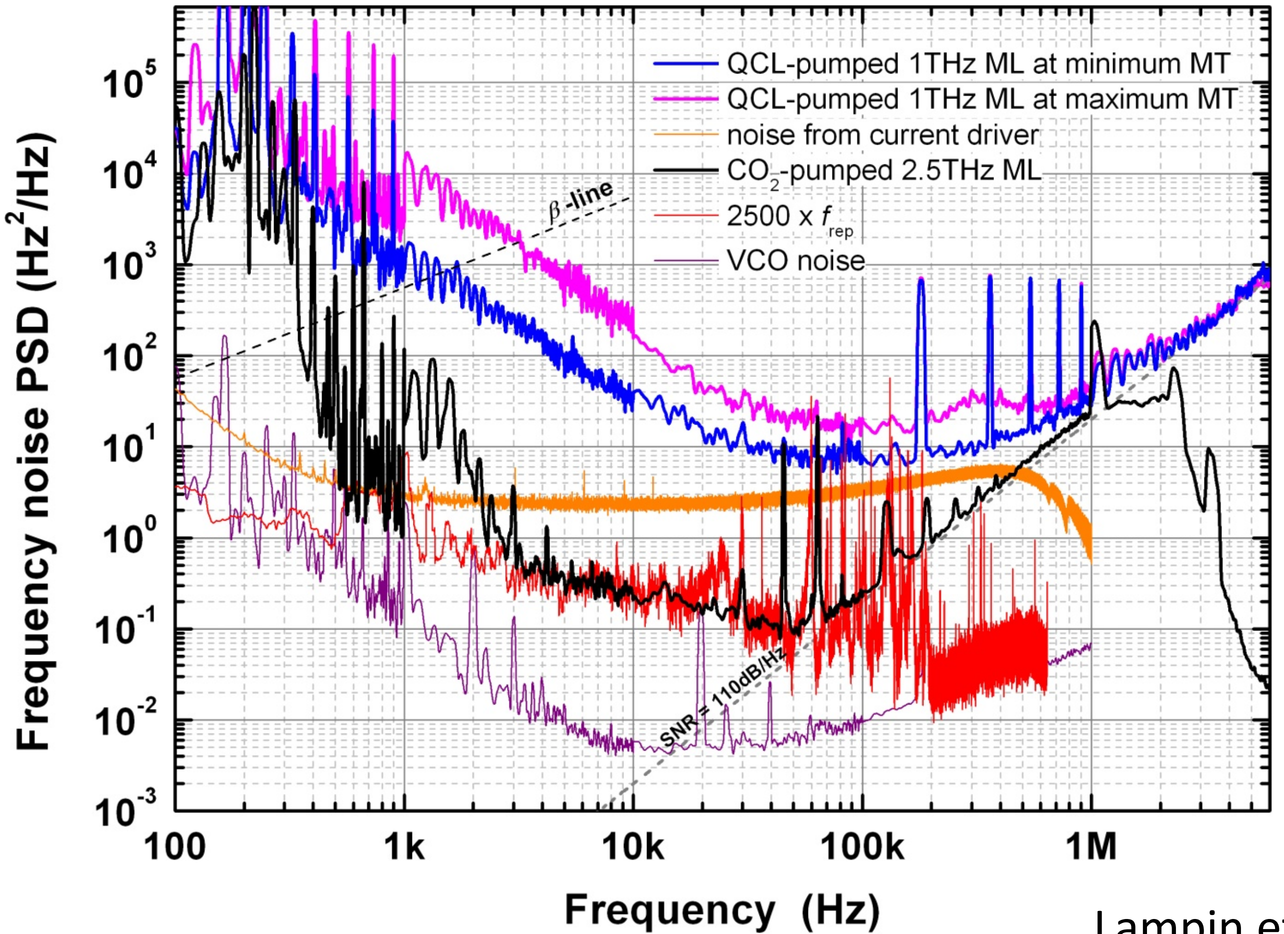
Frequency noise of QCL-pumped THz laser

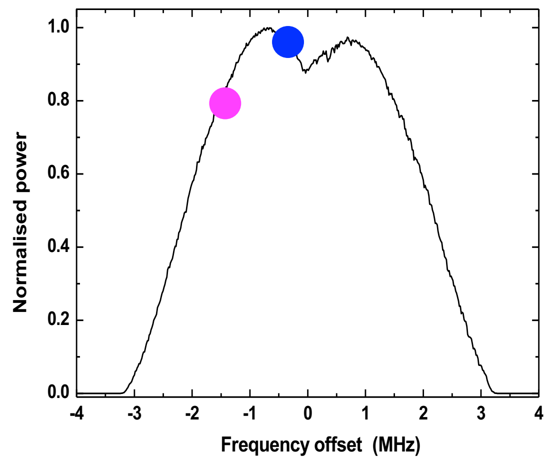




- Variation of modulation transfer observed on noise spectra
- from β-line crossing → linewidth < 5kHz on 10ms integration time
- Below ~30kHz → ~ "technical noise" compatible with mid-IR QCL frequency noise
- Above 30kHz → THz laser frequency noise (possibly limited by current driver noise)

Comparison with frequency noise of CO_2-laser pumped FIR laser

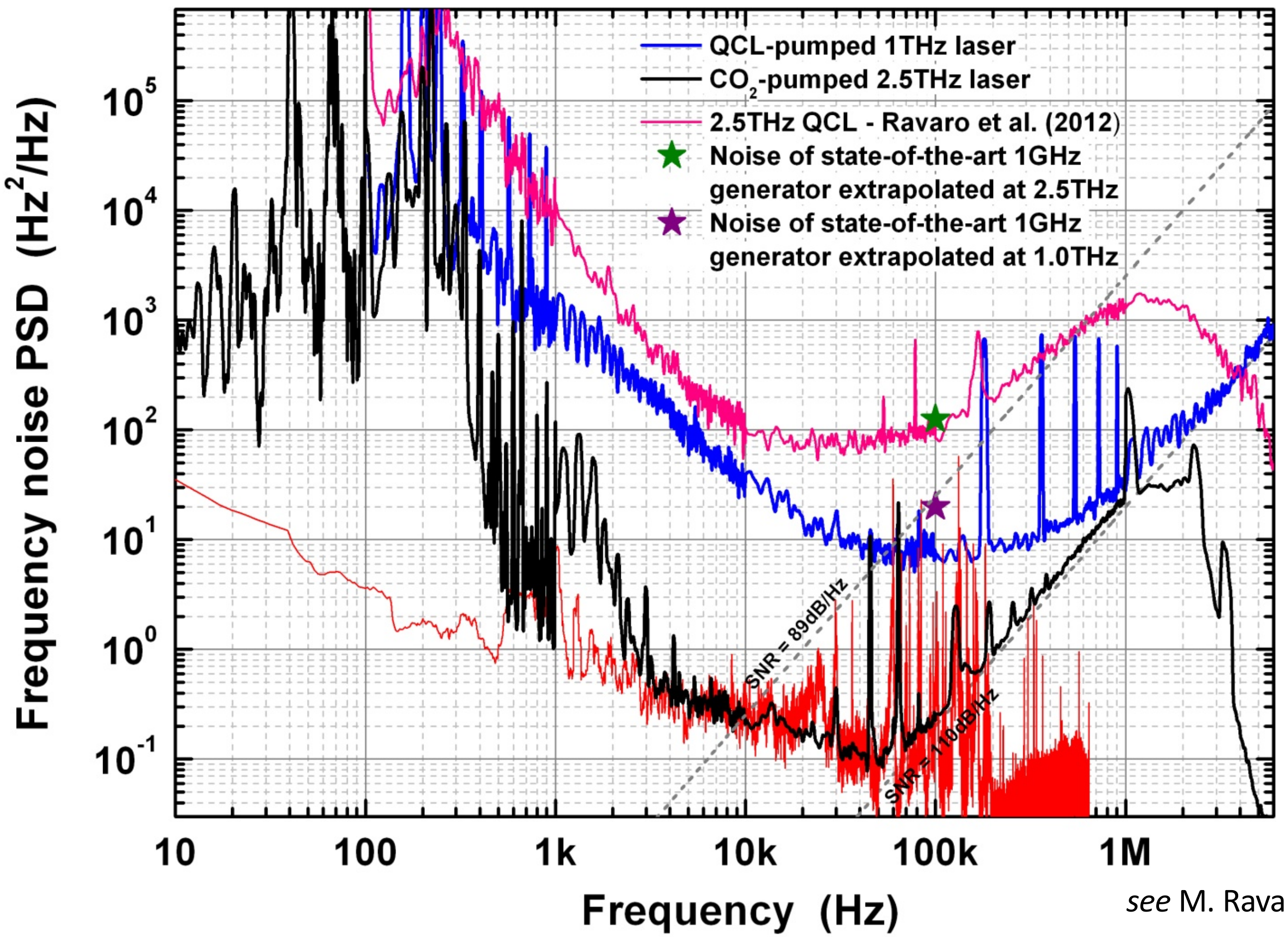




Lampin et al. Opt. Expr. **28**, 2091 (2020)

- CO_2-laser pumped 2.5THz laser PSD measured using electro-optic detection
- Noise-analysis limited by the noise of frequency comb rep. rate harmonic (2500 x f_{rep})

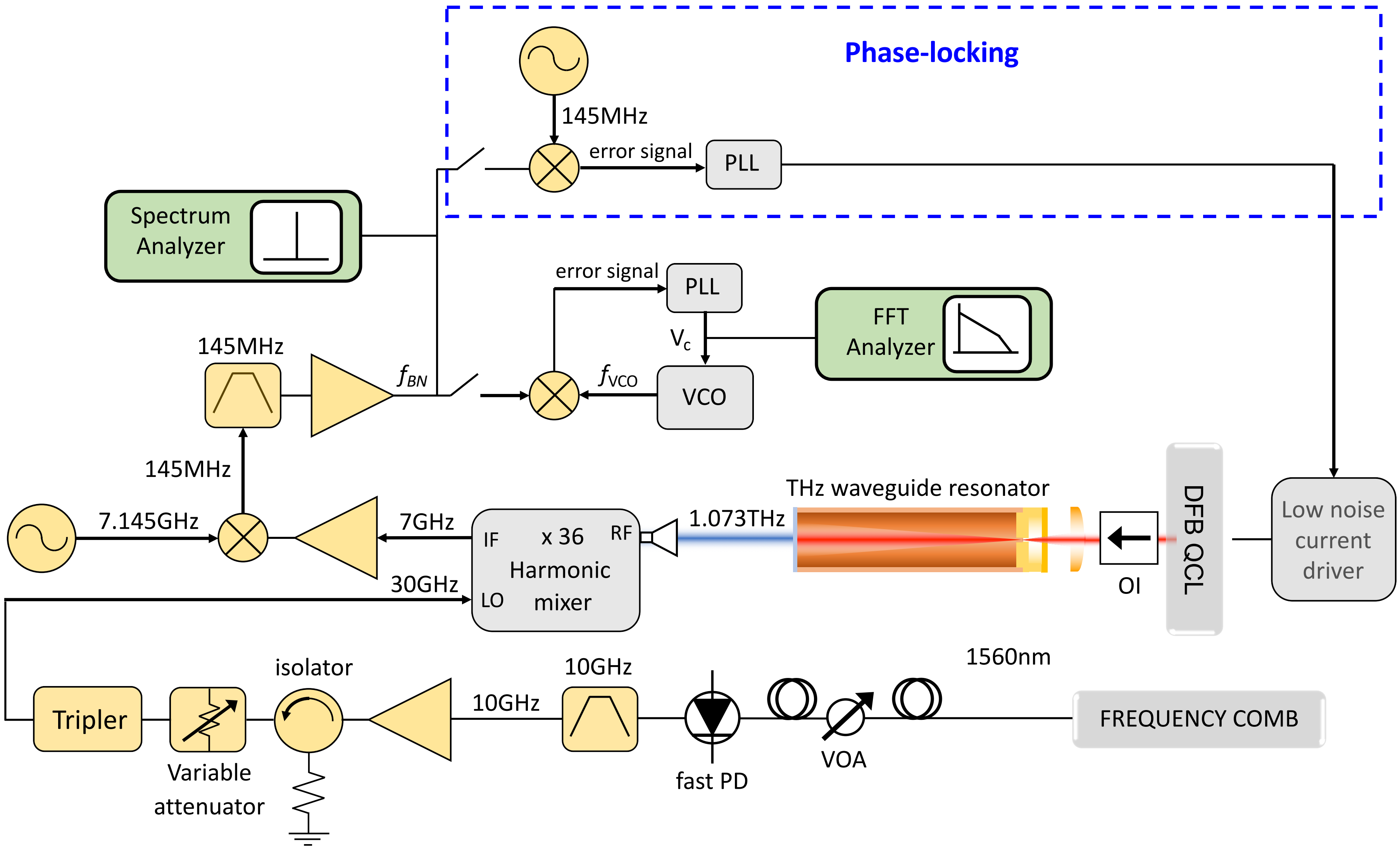
Comparison with CO_2-pumped 2.5THz laser and 2.5THz QCL



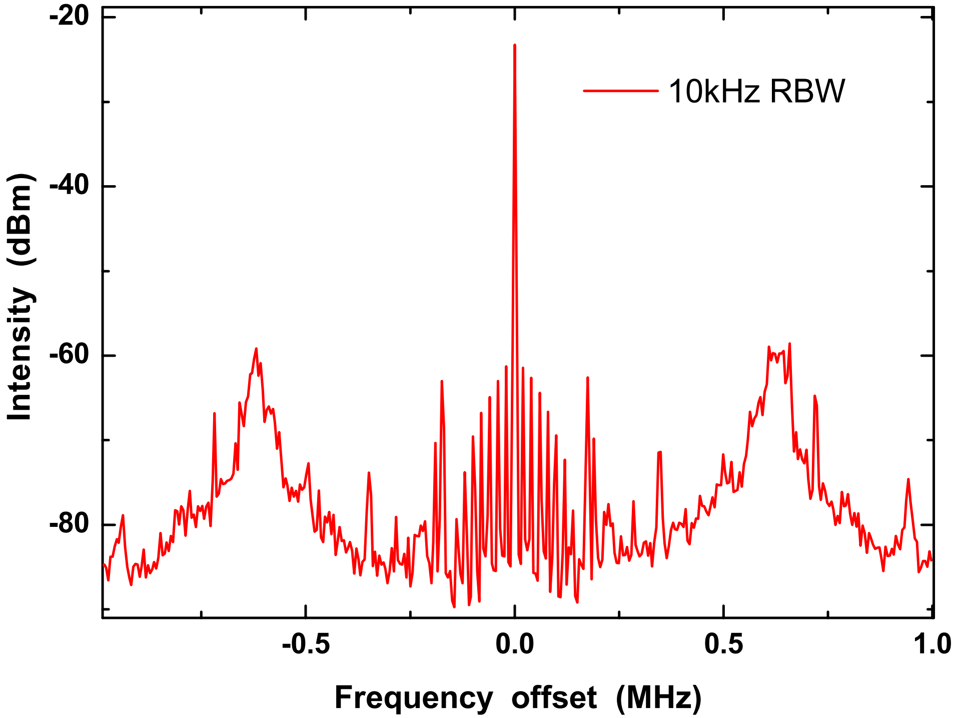
see M. Ravaro et al., Opt. Expr. **5**, 25654 (2012)

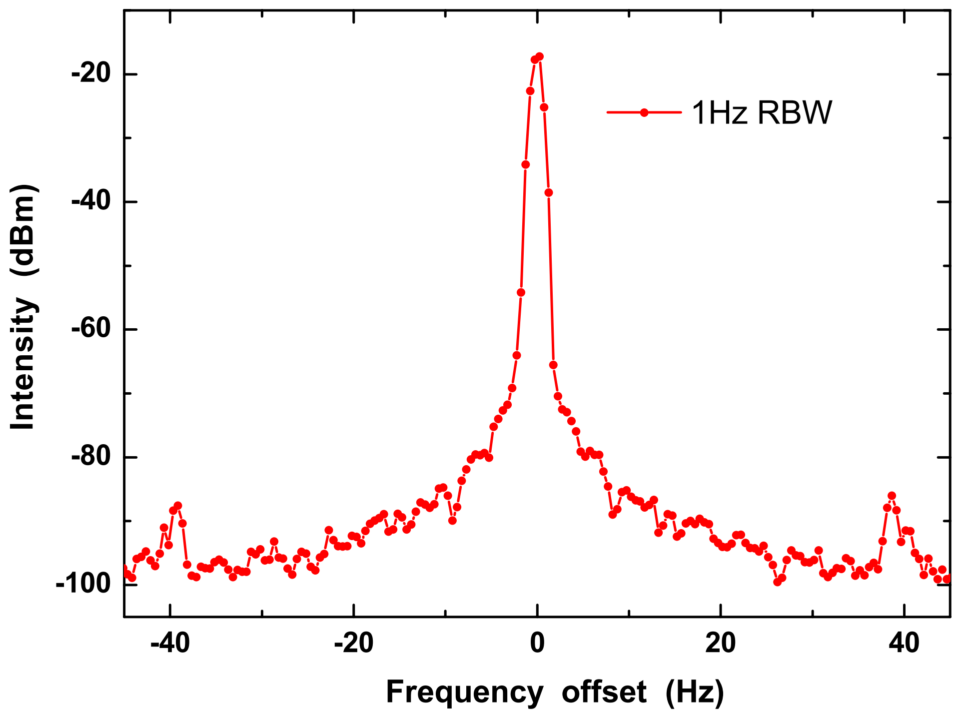
- ➢ The pump makes a difference…!
- ➢ The frequency comb too…!
- ➢ <u>The QCL-pumped FIR laser is a spectrally pure source</u>

Phase-locking to the 1080th harmonic of f_{rep}



Phase-locking to frequency comb





- Phase-locking successful: sub-Hz linewidth
- ~ 0.04rad residual rms phase noise → > 99.5% of BN signal power coherently locked

Conclusions

- **First measurement of frequency noise of mid-IR QCL-pumped 1THz ammonia laser**
  - → 7Hz^2/Hz at 100kHz, limited QCL current driver noise
  - → 10-fold improvement compared to 2.5THz THz QCL
  - → comparable to multiplied state-of-the-art GHz oscillator

- **Phase-locking of mid-IR QCL-pumped 1THz ammonia laser to frequency comb demonstrated, with sub-Hz linewidth**

Lampin et al. Opt. Expr. **28**, 2091 (2020)

- **QCL-pumped molecular lasers have a bright future as THz sources for applications such as high-resolution spectroscopy, radio-astronomy, free space THz communications, THz radar imagery...**