Ro-vibrational fine structure of CF_3H by frequency-comb-assisted saturated spectroscopy at 8.6 µm

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Aim of high-resolution CF_3H spectroscopy

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Why CF_3H?

A great enhancement in the ultimate accuracy can be achieved if a **beam of slow and cold molecules** is used to implement **Ramsey-fringes technique**

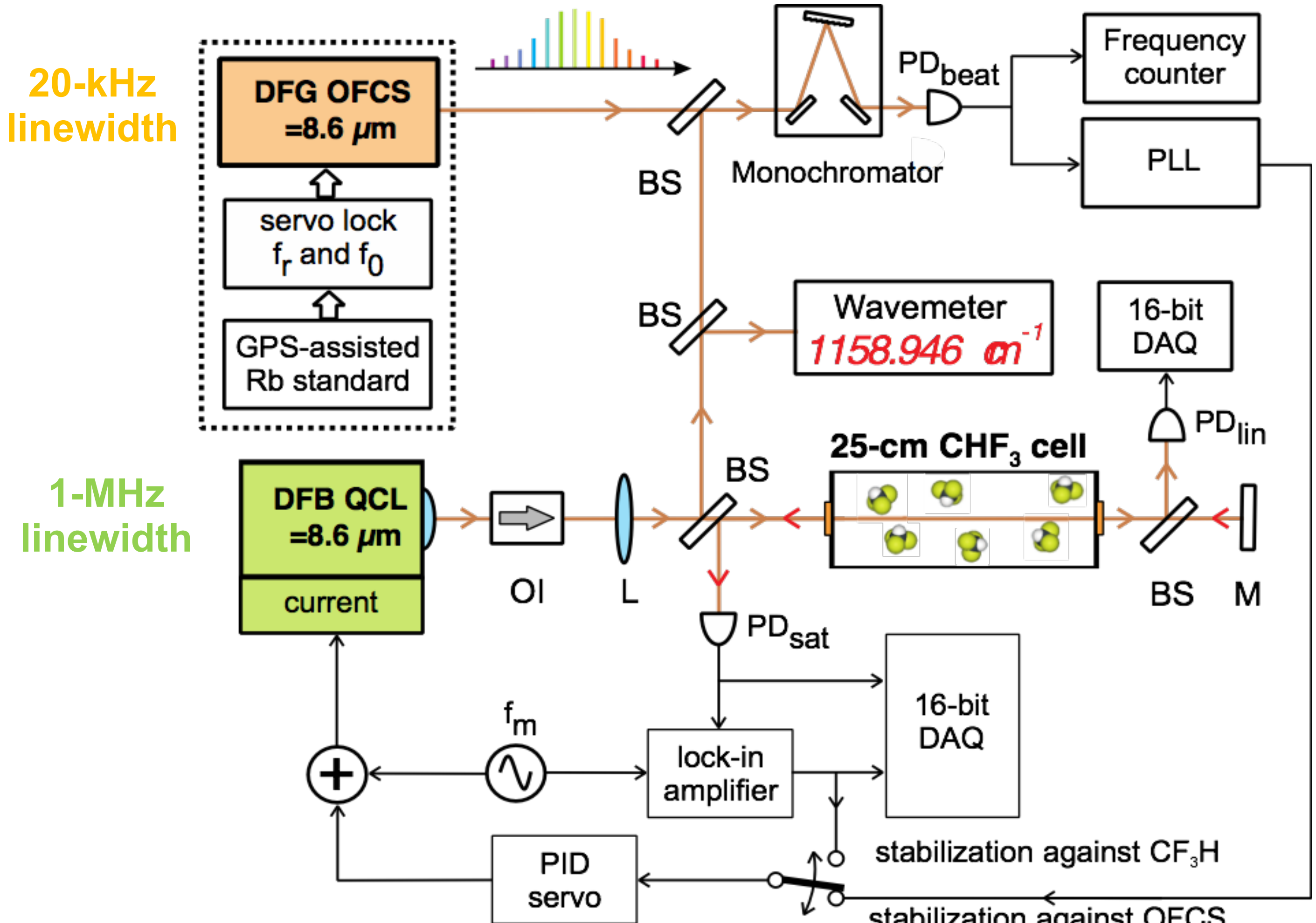
CF_3H can be easy cold down in a molecular beam (buffer gas cooling and Stark manipulation) due to a high electric dipole moment

CF_3H presents favorable two-photon transition in the mid-IR

Outline

- Sub-Doppler CF_3H spectrometer at 8.6 μm
- Comb-assisted CF_3H sub-Doppler spectroscopy
- Conclusion and further activity

Sub-Doppler CF_3H spectrometer

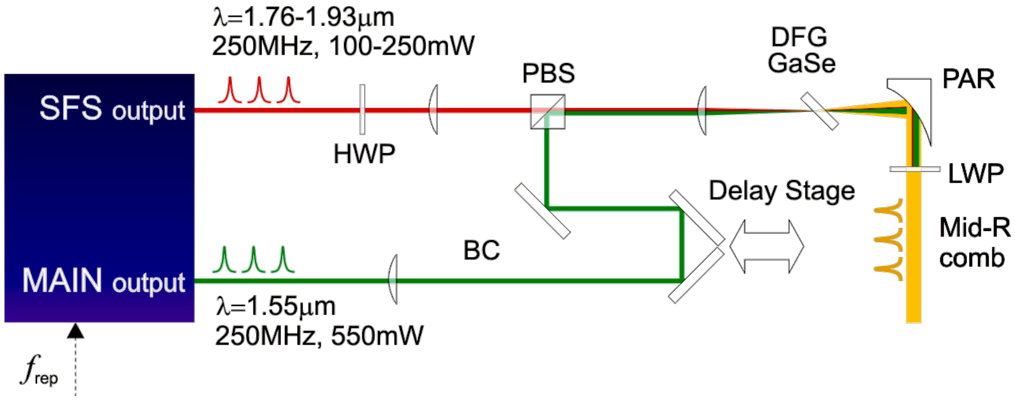


A. Gambetta, *et al.*, Opt. Lett. **38**, 1155 (2013)

E. Fasci, et al., Opt. Lett. **39**, 1155 (2014)

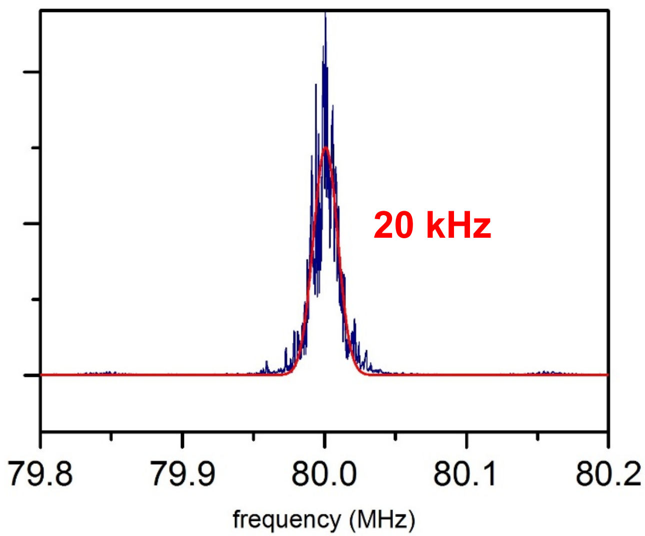
Optical Frequency Comb (OFC) synthesizers

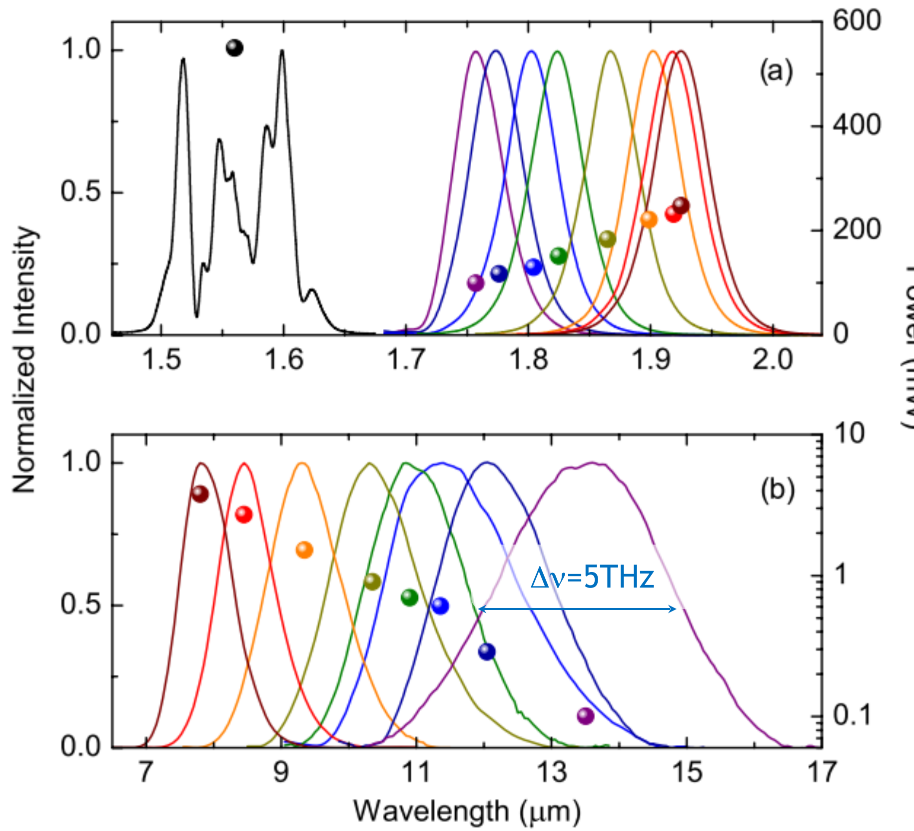
DFG OFC at 8 – 14 μm



GaSe CRYSTAL
High nonlinearity (50 pm/V)
Broad transparency range

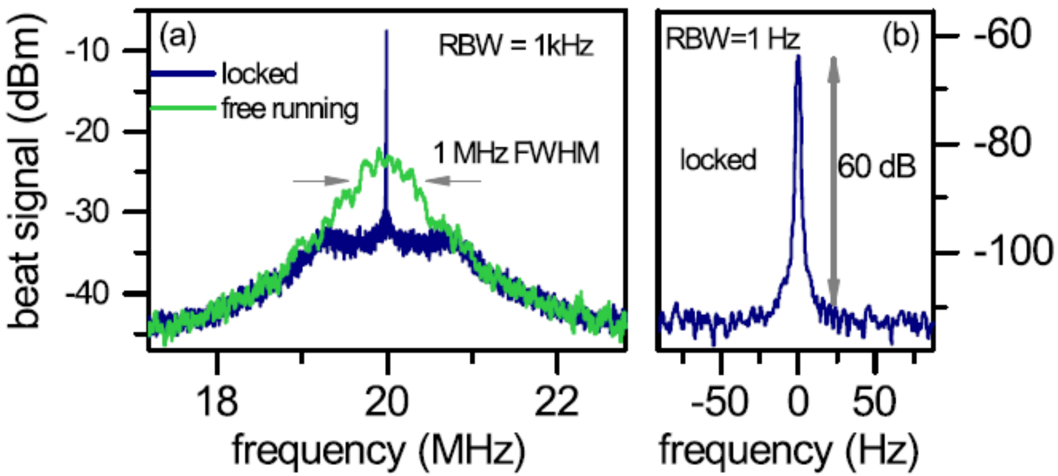
Beat with 4-kHz linewidth QCL

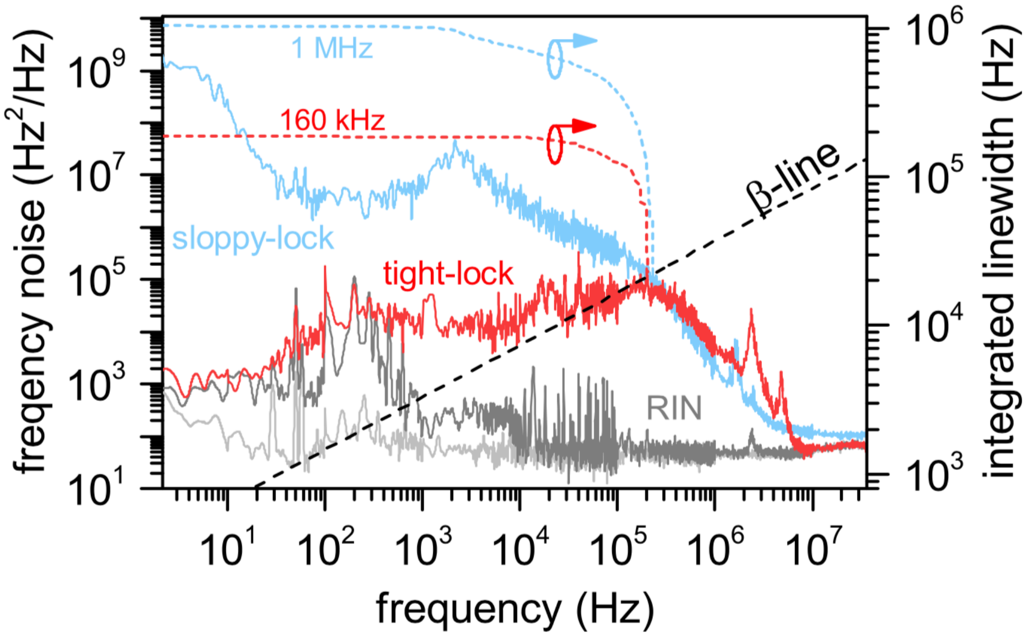




A. Gambetta, *et al*., Opt. Lett. **38,** 1155 (2013)

QCL to DFG comb phase-locking





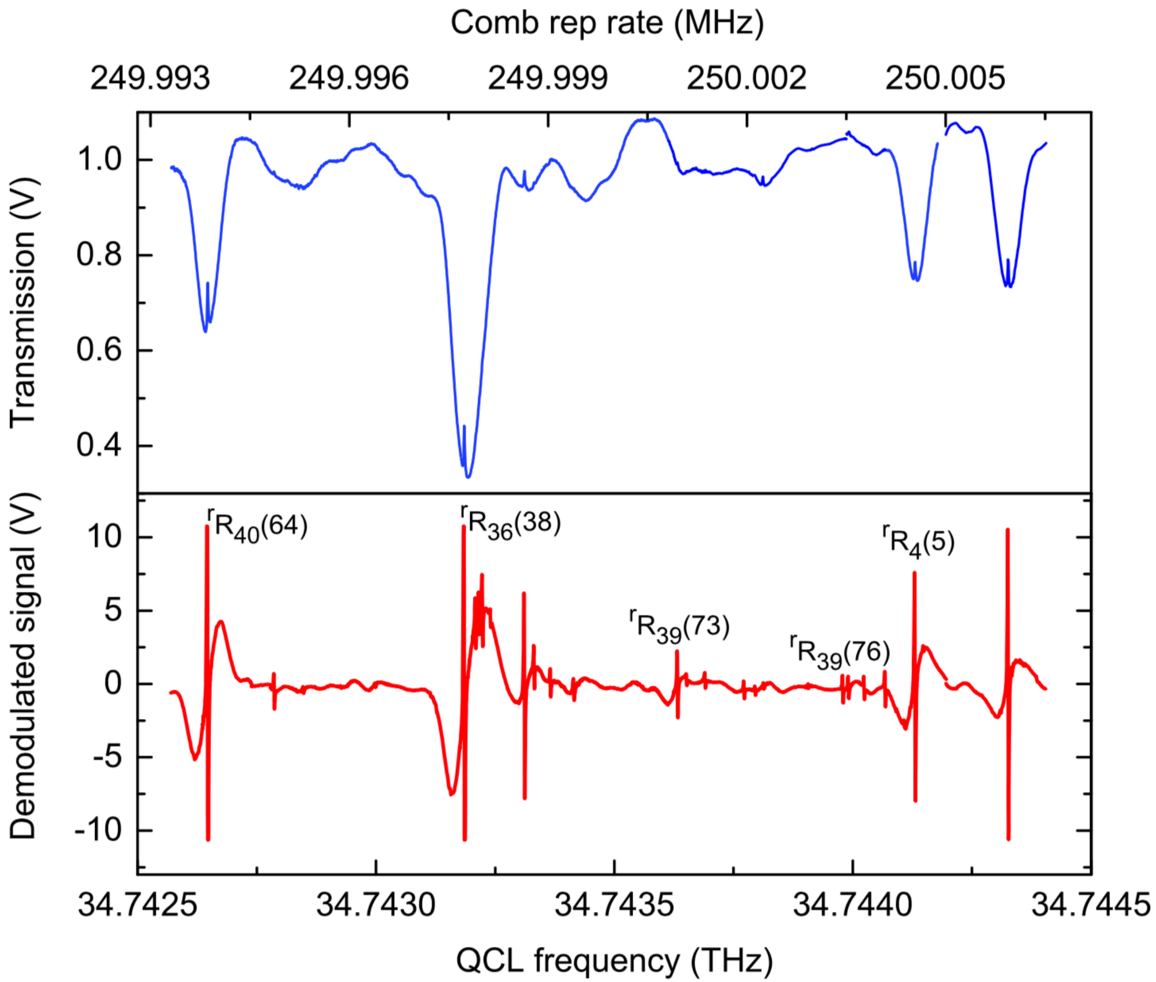
Free running beatnote
Linewidth: 1 MHz
SNR: 30 dB (1-kHz RBW)

Phase-locked beatnote
Linewidth: < 1 Hz
SNR: 60 dB/Hz
54% of the RF power in the carrier

QCL linewidth
160 kHz at 1-s integration
Servo-loop BW: 250 kHz

A. Gambetta, et al., APL Phot. **3**, 4 (2018)

Comb-assisted scanning



CHF_3 pressure
6.0(1) Pa

Pump beam power
5.0(3) mW

Pump beam radius
1.0(0) mm

Modulation parameters
f_m = 10 kHz
a_m = 320 kHz

Tens of saturated absorption lines in 2-GHz frequency span

Lamb-dip linewidths and contrasts ranging from 1.5 to 2 MHz and from 1% to 20%

Highest SNR: 58 dB in 0.1 kHz integration bandwidth

CF_3H line-center frequency measurements

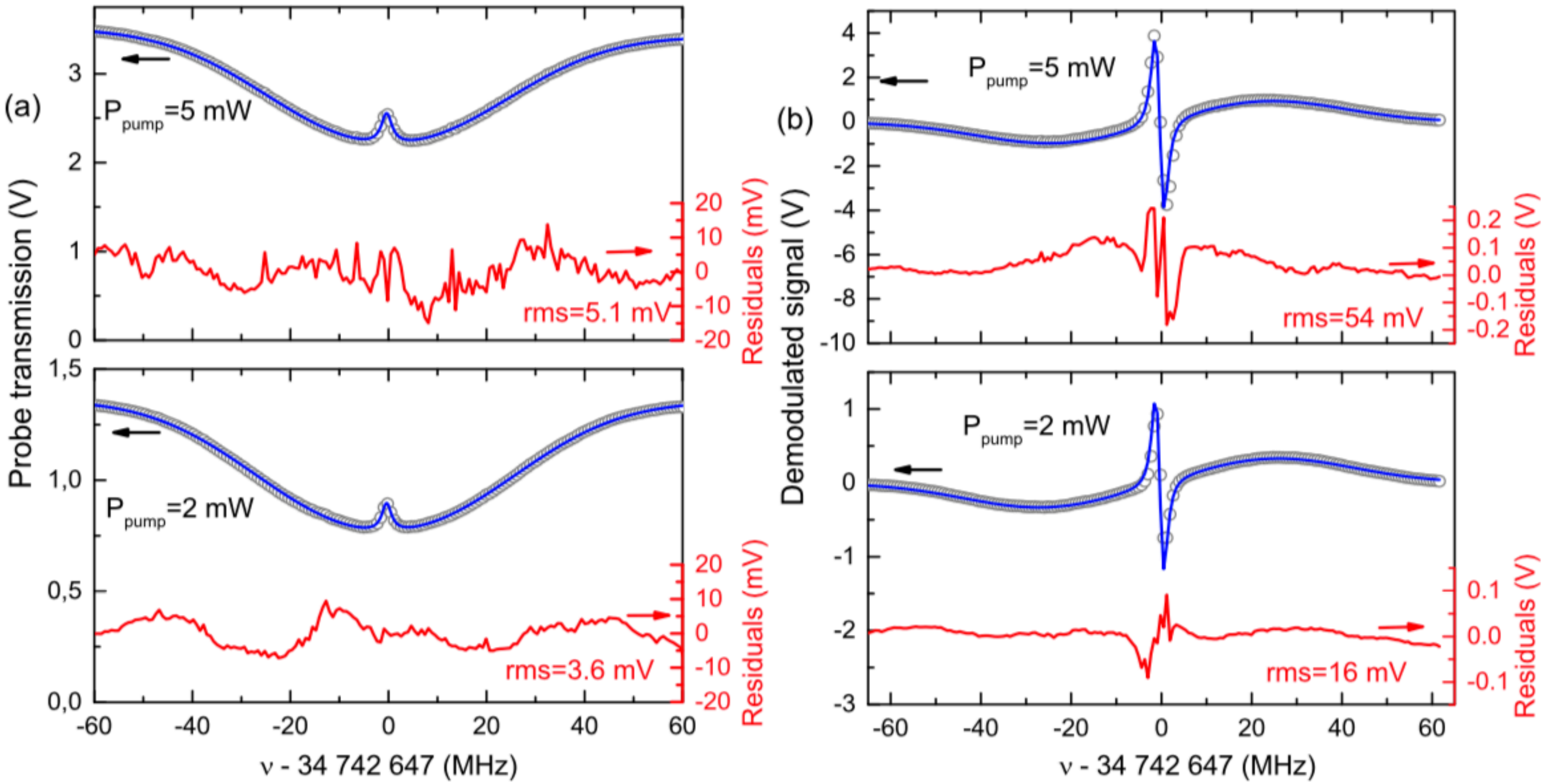
Line	Frequency (MHz)	Wave number (cm^{-1})	Assignment ν_5 band	Assignment ν_2 band	Obs. - Calc. (10^{-4} cm^{-1})
1	34 742 647.0(1)	1158.889 961(3)	$^{r}R_{40}(64)$		7.5
2	34 742 745(1)	1158.893 23(3)	–	–	
3	34 742 786.4(5)	1158.894 61(2)	–	–	
4	34 742 833(1)	1158.896 17(3)	$^{r}R_{39}(66)$		4.1
5	34 742 847(1)	1158.896 63(3)	–	–	
6	34 742 926(1)	1158.899 27(3)	$^{r}R_{41}(61)$		5.5
7	34 742 998(1)	1158.901 67(3)		$^{q}R_{36}(79)$	3.1
8	34 743 185.1(1)	1158.907 910(3)	$^{r}R_{36}(38)$		0.3
			$^{r}R_{37}(77)$		4.9
			$^{r}R_{41}(75)$		5.5
9	34 743 208.8(5)	1158.908 70(2)	–	–	
10	34 743 216.1(5)	1158.908 94(2)	–	–	
11	34 743 223.2(3)	1158.909 18(1)	–	–	
12	34 743 240.2(5)	1158.909 75(2)	–	–	
13	34 743 311.3(1)	1158.912 120(3)	–	–	
14	34 743 331.6(3)	1158.912 80(1)	–	–	
15	34 743 366.0(5)	1158.913 94(2)		$^{q}R_{36}(80)$	6.4
16	34 743 415.0(5)	1158.915 58(2)	–	–	
17	34 743 423(1)	1158.915 85(3)	–	–	
18	34 743 632.5(3)	1158.922 83(1)	$^{r}R_{39}(73)$		7.2
19	34 743 652(1)	1158.923 48(3)		$^{q}R_{36}(81)$	6.3
20	34 743 676(1)	1158.924 29(3)	$^{r}R_{37}(78)$		1.3
21	34 743 690.2(5)	1158.924 76(2)	$^{r}R_{40}(74)$		-2.0
22	34 743 771.4(5)	1158.927 48(2)	–	–	
23	34 743 795(1)	1158.928 25(3)	–	–	
24	34 743 813(1)	1158.928 86(3)		$^{q}R_{36}(87)$	0.4
25	34 743 907(1)	1158.931 99(3)	–		
26	34 743 978.9(5)	1158.934 39(2)	$^{r}R_{43}(78)$		7.8
			$^{r}R_{39}(76)$		4.0
				$^{q}R_{36}(86)$	3.2
27	34 743 991.0(5)	1158.934 79(2)		$^{q}R_{36}(83)$	0.7
28	34 744 023.8(5)	1158.935 89(2)	–	–	
29	34 744 067.9(5)	1158.937 36(2)		$^{q}R_{36}(84)$	3.7
				$^{q}R_{36}(85)$	5.7
30	34 744 107(1)	1158.938 66(3)	$^{r}R_{37}(79)$		3.5
31	34 744 110(1)	1158.938 76(3)	–	–	
32	34 744 130.6(1)	1158.939 449(3)	$^{r}R_{4}(5)$		-1.4
33	34 744 148(1)	1158.940 03(3)	$^{r}R_{38}(77)$		-7.3
34	34 744 310(1)	1158.945 43(3)	$^{r}R_{39}(67)$		7.1
			$^{r}R_{40}(46)$		9.0
35	34 744 326.0(1)	1158.945 967(3)	–	–	
36	34 744 333(1)	1158.946 20(3)	–	–	

The line center frequency is measured for 36 lines, with a best fractional precision of **2×10^{-9}** in a single scan acquisition of 1000 s

E. Vicentini et al, submitted to JQRT March 2018

Comb-assisted sub-Doppler spectroscopy

Isolated $^{r}R_{40}$ (64) line

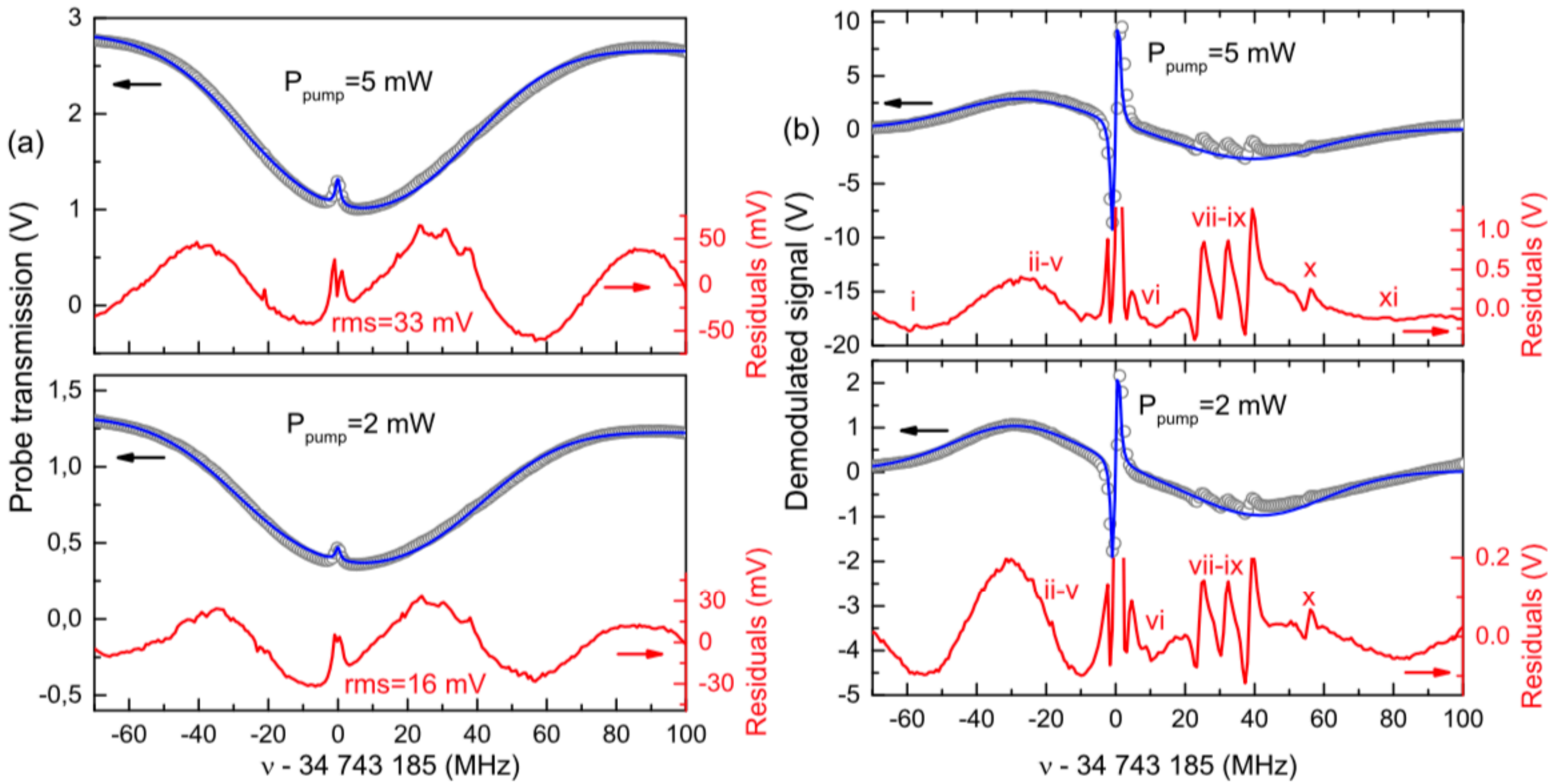


$$\alpha(\omega) = \alpha_0(\omega - \omega_{0,G}) \times \left\{1 - \left(1 - \frac{1}{\sqrt{1+S}}\right) N\, g_V(\omega - \omega_{0,V})\right\}$$

$$Z_D(\omega) = (Z_0 + Z_1\,\omega)\exp\left[-\alpha(\omega)L\right] \qquad Z_{WM}(\omega) = (Z_0^{WM} + Z_1^{WM}\omega) + \left[S_{1,G}(\omega) + S_{1,V}(\omega)\right]$$

Comb-assisted sub-Doppler spectroscopy

Multi ${}^{r}R_{36}$ (38) line

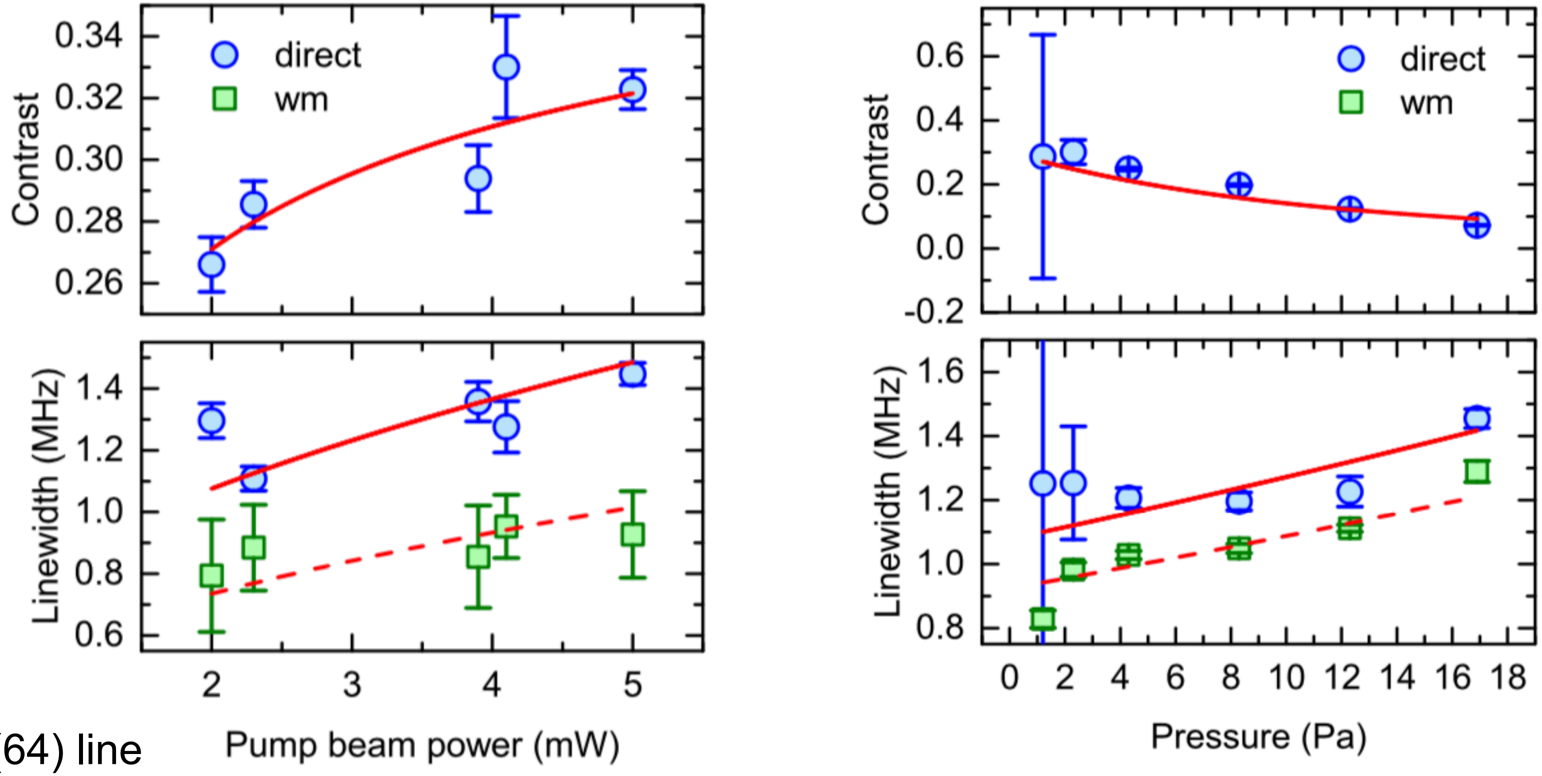


$$\alpha(\omega) = \alpha_0(\omega - \omega_{0,G}) \times \left\{1 - \left(1 - \frac{1}{\sqrt{1+S}}\right) N\, g_V(\omega - \omega_{0,V})\right\}$$

$$Z_D(\omega) = (Z_0 + Z_1\,\omega)\exp\left[-\alpha(\omega)L\right]$$

$$Z_{WM}(\omega) = (Z_0^{WM} + Z_1^{WM}\omega) + \left[S_{1,G}(\omega) + S_{1,V}(\omega)\right]$$

Rovibrational fine structure of CF_3H



$^{r}R_{40}$ (64) line

	DS	WM
Saturation power, P_s, (mW)	0.3(1)	0.3(1)
Lorentzian linewidth, γ_0, (MHz)	0.29(1)	0.20(1)
Rabi frequency, Ω_0, (MHz)	0.7(2)	0.8(2)
Pressure broadening, B_p, (MHz/Pa)	0.016(2)	0.013(2)

Transition electric dipole moment

$(7 \pm 1) \cdot 10^{-31}$ C m

(0.22 ± 0.04) Debye

Conclusion

- ✓ Direct phase-locking of a QCL at 8.6 μm to a mid-IR frequency comb
  - ✓ QCL-line narrowing down to 160-kHz (FWHM)

- ✓ High-precision sub-Doppler spectroscopy of CF_3H at 8.6 μm
  - ✓ 0.3-kHz (10^{-11}) frequency axis accuracy
  - ✓ 150-MHz/min tuning speed
  - ✓ Best SNR 59 dB at 100 Hz integration bandwidth
  - ✓ Transition electric dipole moment measurement

Future perspectives

Non-linear spectroscopic of cold CF_3H (100-150 K) at 8.6 μm

- ✓ Two-photon spectroscopy
- ✓ Hyperfine structure measurement

Thank you for attention

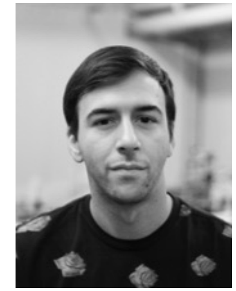
Acknowledgement


N. Coluccelli


A. Gambetta


T. T. Fernandez


Y. Wang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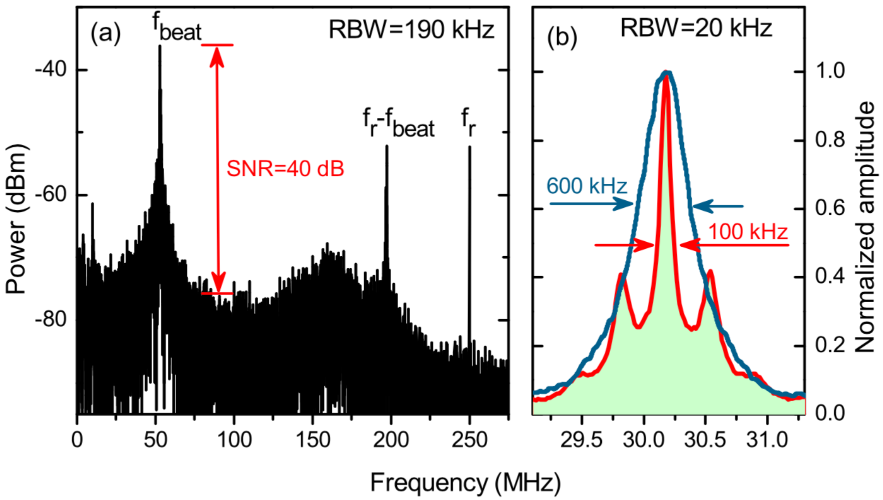
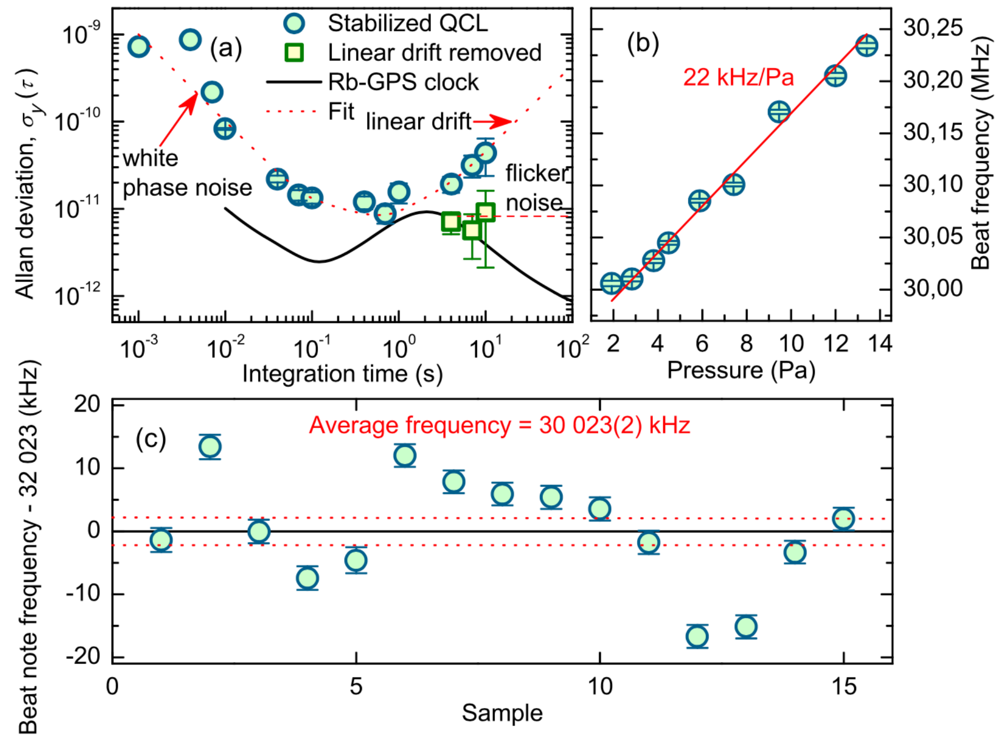
Frequency stabilization against saturated CF_3H absorption line



Uncertainty budget

Parameter	Coefficient	Type B Uncertainty (kHz)
CHF_3 pressure, p_{CHF_3}	22(1) kHz/Pa	0.75
Gas cell leakage, p_{leak}	38(2) kHz/Pa	1.5
Laser power, P_{QCL}	45 kHz/mW	0.2
Modulation frequency, f_m	100 Hz/kHz	Negligible
Modulation depth, a_m	16 kHz/MHz	Negligible
Electronic offset	0.3 kHz/mV	0.2
Etalon/interference effects		0.5
Rb-GPS clock		0.04
Total type B uncertainty		1.8 $(5 \cdot 10^{-11})$



$^rR_{36}$ (38) line center frequency

34 743 124 881(2) kHz

Fractional accuracy 6x10^{-11}

A. Gambetta et al, Opt. Lett. **42**, 1911-1914, 2017