COMB-REFERENCED COHERENT RAMAN SPECTROSCOPY ON PURE H$_2$

M. Lamperti, L. Rutkowski, D. Gatti, B. Alsaif, R. Gotti, G. Cerullo, D. Polli, M. Marangoni, F. Thibault, P. Maslowski, S. Wojtewicz, P. Wcislo
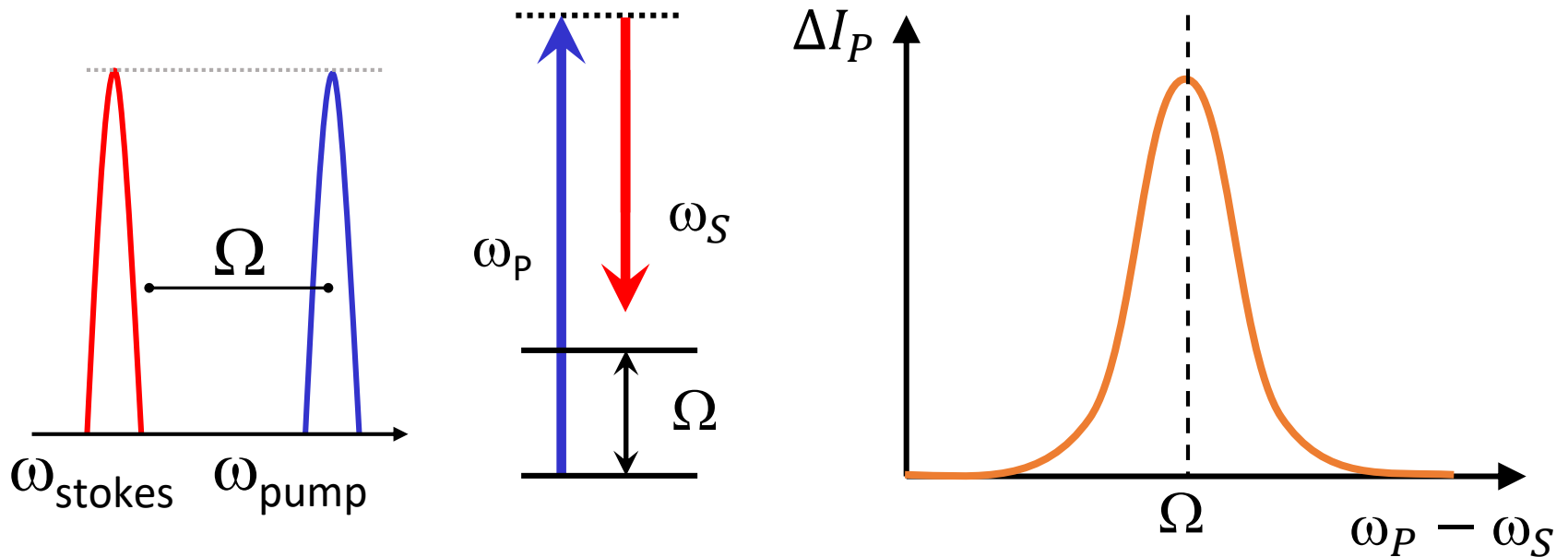
THE IMPORTANCE OF H_2

- Simplest molecule and most abundant in the universe
  - Benchmark system for *ab-initio* calculations of transitions frequencies and line profiles
  - Transition frequencies:
    - Test for new physics without the need for a particle accelerator
    - Test of invariance of fundamental constants
    - Interpretation of observational data (interstellar clouds…)

- Presence of strong collisional effects
  - Low-pressure measurements are favored to extract zero-pressure transition frequencies
  - High pressure gives much better SNR and requires better collision modeling → *Raman spectroscopy*

A. Owyoung, Opt. Lett. **2** (4), 1978; G. J. Rosasco and W. S. Hurst, PRA **32** (1), 1985

STIMULATED RAMAN SCATTERING

Third order nonlinear process:
stimulated emission from a virtual level

SRS SIGNAL

$$\Delta I_{P,max} \propto \chi^{(3)} L I_S I_P$$

- $\chi^{(3)} \to$ H$_2$ Raman cross-section is high
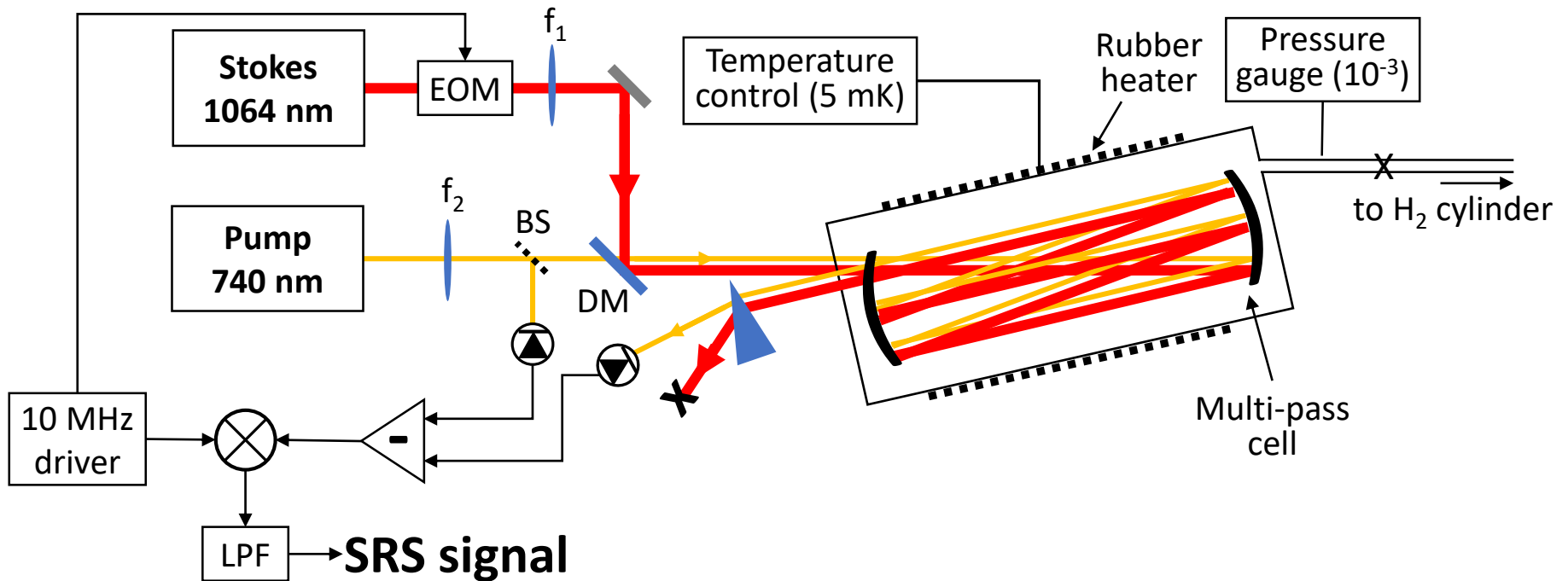- $L \quad \to$ Multi-pass cell
- $I_S \quad \to$ High-power CW laser

SRS signal estimation @ 1 atm for (1←0) Q(1) line:

- 10 mW pump at 740 nm
- 5 W Stokes at 1064 nm
- 30 m multipass cell

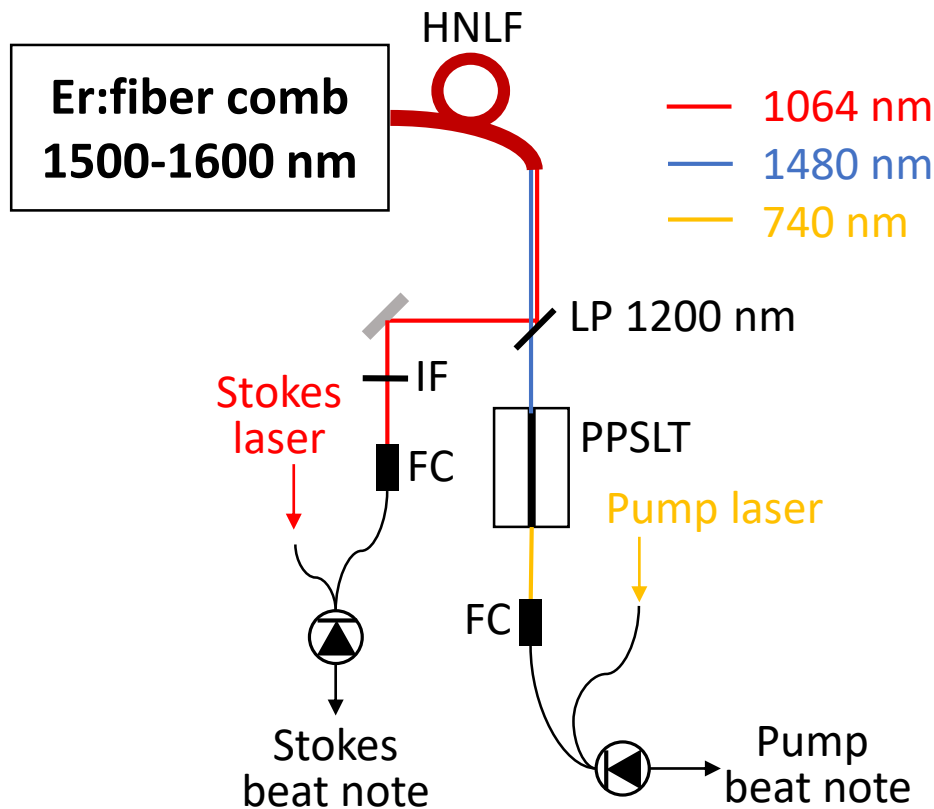$$\frac{\Delta I_{P,max}}{I_P} \approx 5 \cdot 10^{-4}$$

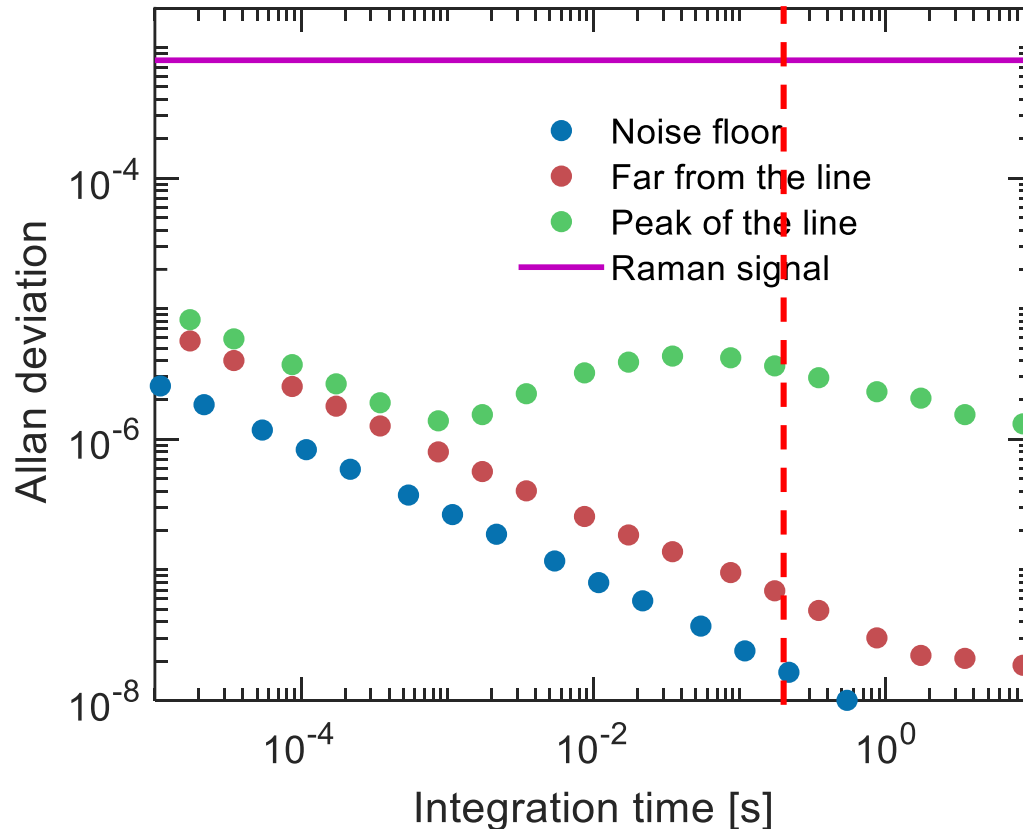Shot noise $\approx 3 \cdot 10^{-7}$ in 100 ms
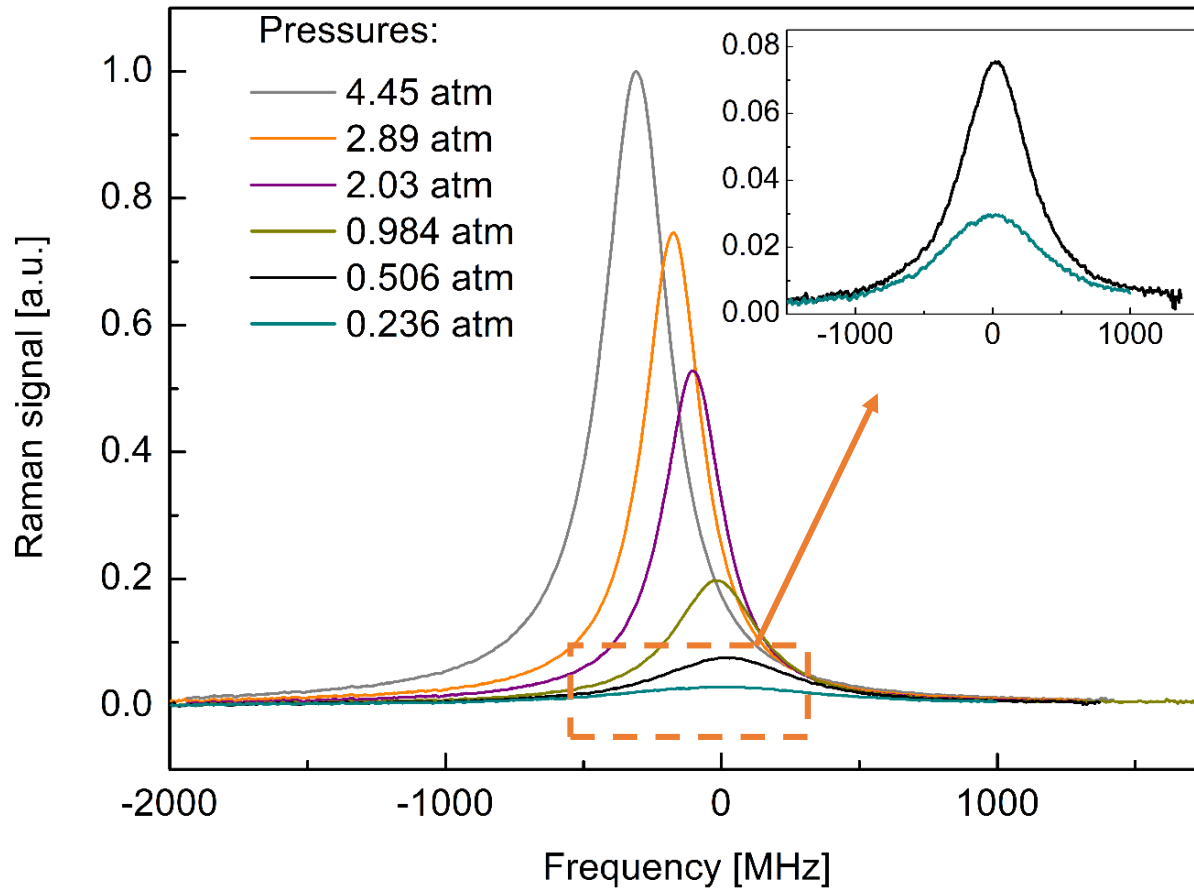
SETUP

COMB REFERENCING

NOISE ANALYSIS

Allan deviation of the demodulated signal

DATA

SNR:
- 2000 @ 5 atm
- 200 @ 0.5 atm

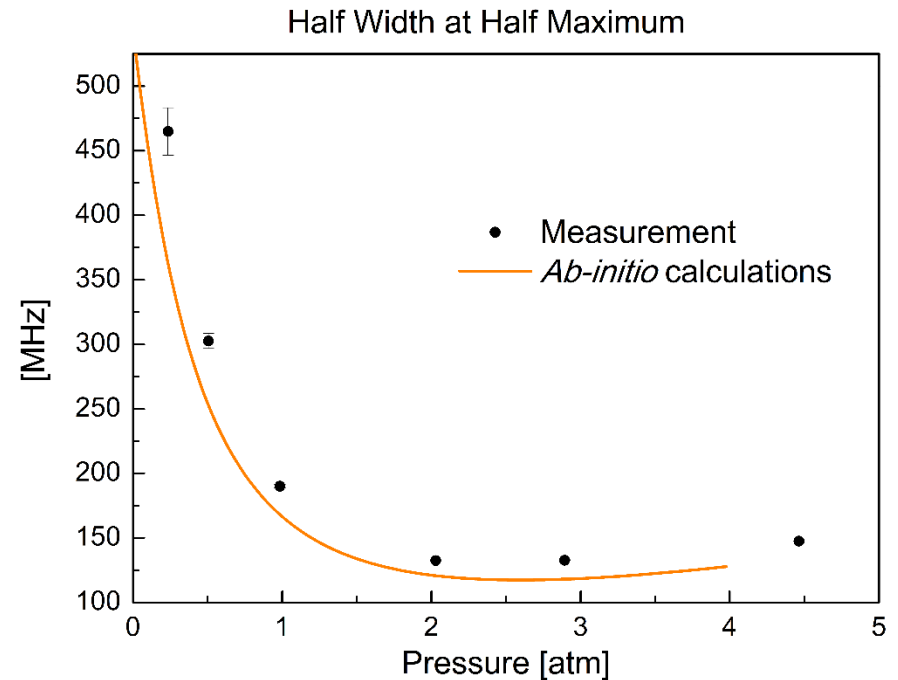| Type A	< 560 KHz
Comb	40 kHz
Pump BN	300 kHz
Stokes BN tracking	400 kHz
Temperature stability	< 10 kHz
Pressure stability	< 250 kHz
Type B	**< 1.53 MHz**
Pressure accuracy	< 1.53 MHz
Temperature accuracy	< 90 kHz
Purity of H_2	99.999%

RESULTS FROM RAW DATA

Fit: $-3.0 \cdot 10^{-3}$ cm^{-1}/atm
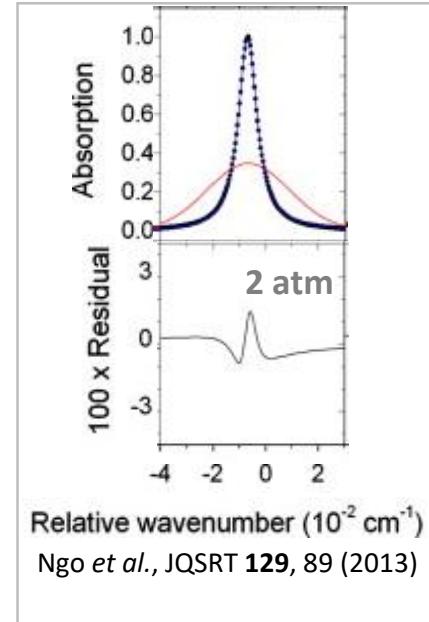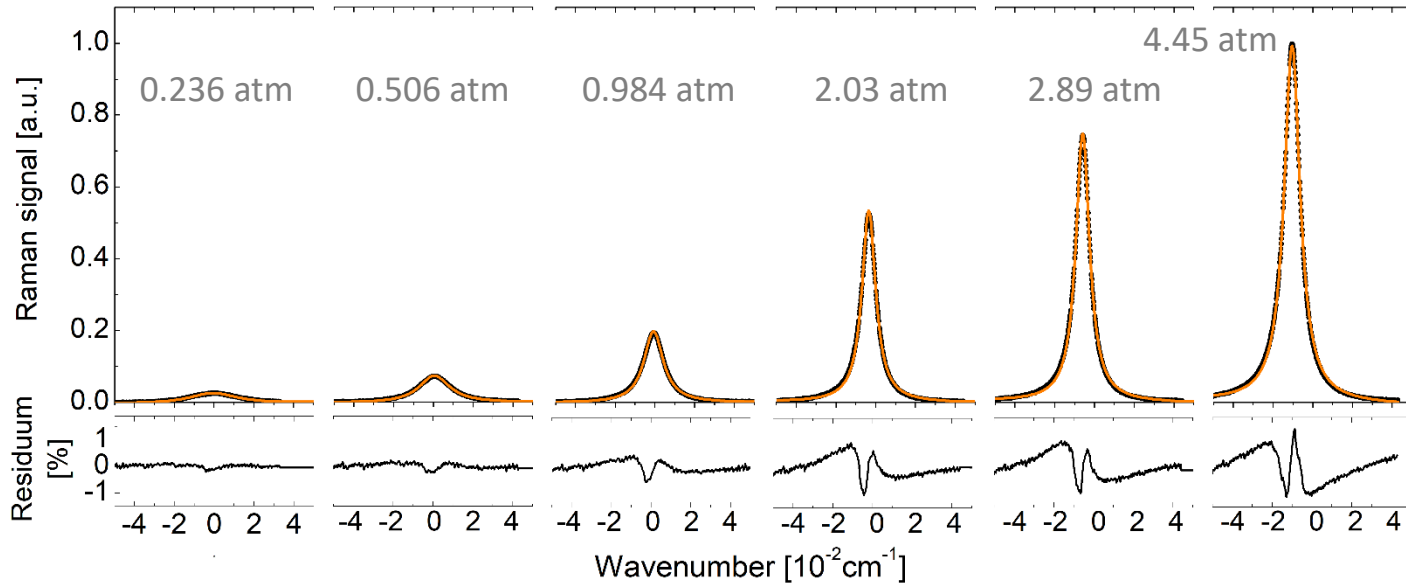
Literature:

$-3.4(1) \cdot 10^{-3}$ cm^{-1}/atm [1]

$-3.14(15) \cdot 10^{-3}$ cm^{-1}/atm [2]

[1] P. Wcislo et al., JQSRT **177**, 75 (2016)
[2] Rahn *et al.*, PRA **43** (11), 6075 (1991);

MULTISPECTRUM FIT

Ngo *et al.*, JQSRT **129**, 89 (2013)

	γ_0 [10^{-4} cm^{-1}/atm]	δ_0 [10^{-3} cm^{-1}/atm]	γ_2 [cm^{-1}/atm]	δ_2 [cm^{-1}/atm]	ν_{VC} [cm^{-1}/atm]	η	γ_D [10^{-2} cm^{-1}]
Multi-line	7.3(5)	-2.83(2)	0(0.0004)	0.0018(5)	*0.04499 [3]*	*0*	*1.82*
Lit.	8.7(2) [1]	-3.14(15) [2]	3.3(1)10^{-4} [3]	0.0023(1) [3]			
Single line	80(90)	-	0.014(13)	-0.0015(5)	0.021(12)		

[1] May *et al.*, Can. J. Phys. **39**, 1769 (1961); [2] Rahn *et al.*, PRA **43** (11), 6075 (1991); [3] Wcisło *et al.* JQSRT **177**, 75 (2016)

CONCLUSIONS & OUTLOOK

- SRS + comb is a powerful approach for H_2 spectroscopy

- A better lineshape model is under preparation

- Fixing the laser intensity noise with active stabilization

  < 100 kHz frequency accuracy, SNR > 10^3 between 0.5 and 5 atm

- Target pure rotational lines

THANK YOU!

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