First observation of the H2 quadrupole in saturation (or any quadrupole at all)

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VU

Why hydrogen?
Why a rovibrational transition?

- Simplest neutral molecule
  - 4 body problem
  - Rovibrational transitions are well calculable
- Excellent test for molecular QED theory[1]
- Put bounds on a ‘fifth’ force[2]
- Extract fundamen

$F(\alpha, R_\infty, r_p, \mu_p$

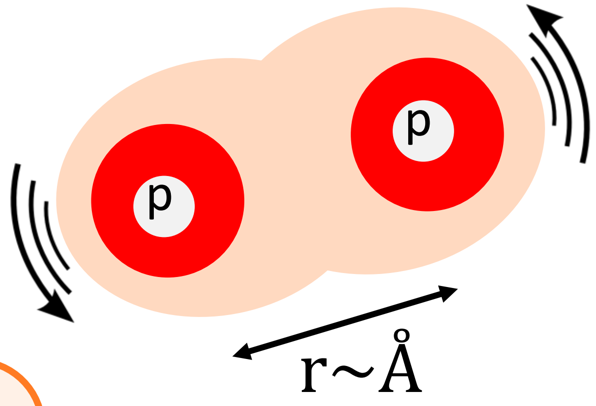
$\downarrow$

$F^{-1}(\alpha, R_\infty, r_p, \nu,$

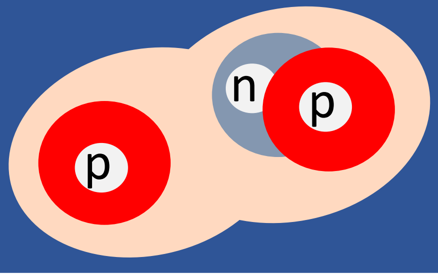
- Lifetimes exceed
  - 20 digit accuracy possible

$\leq$ kHz level accuracy required to determine μ_p and compete with other experiments

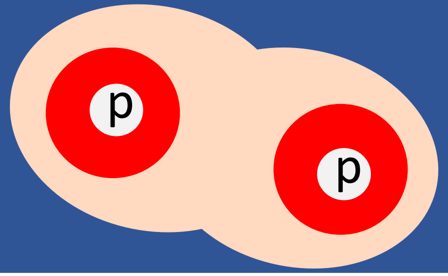
$$\frac{\delta\nu}{\nu} \leq 5 \times 10^{-12}$$



[1] Czachorowski *et. al.*, Phys. Rev. A **98**, 052506 (2018)
[2] Salumbides *et al.*, Phys. Rev. D **87**, 112008 (2013)

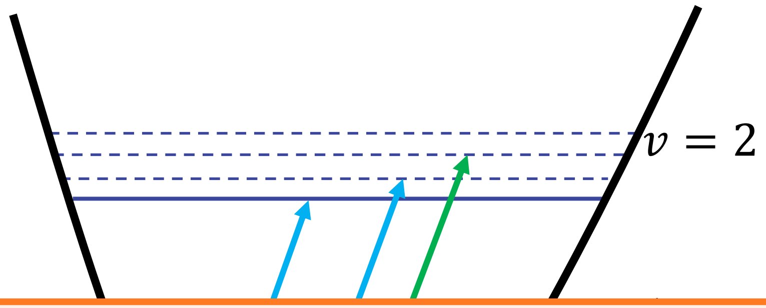


HD vs H_2



Dipole transitions

- $\Delta j = \pm 1$
- Weak dipole
  - First saturation signal in 2017
- Linestrength:
  3.5×10^{-25} cm mol^{-1}
- Einstein A-coeffi
  2.1
- Underlying l
- Complex sig
  - No easy
  - Limits accuracy to ~ 50 kHz



100 times weaker in detection

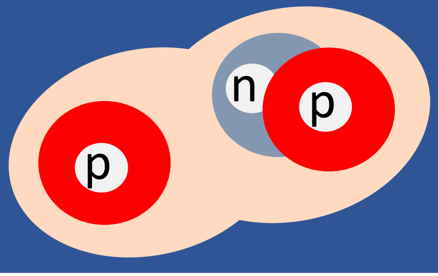
Probably 100 times more difficult to saturate

Quadrupole transitions

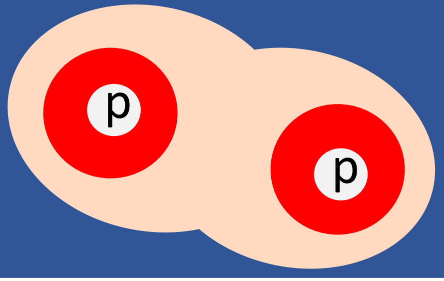
- $\Delta j = 0, \pm 2$
- Extremely weak
  - Transition moment over 100 times smaller
- Linestrength:
  10^{-27} cm mol^{-1}
- efficient:
  10^{-07} s^{-1}
- en has no hyperfine
  the most interesting target
- Best determination at 310 kHz[1]
  - Q(1) (ortho)

Cozijn *et al*., Phys. Rev. Lett. **120**, 153002 (2018)
Diouf *et al*., Opt. Lett. **44**, 4733-4736 (2019)
Diouf *et al*., Phys. Rev. Research **2**, 023209 (2020)
Cozijn *et. al*., PRA **105**, 062823 (2022)
Cozijn *et. al*., Eur. Phys. J. D **76, 220** (2022)

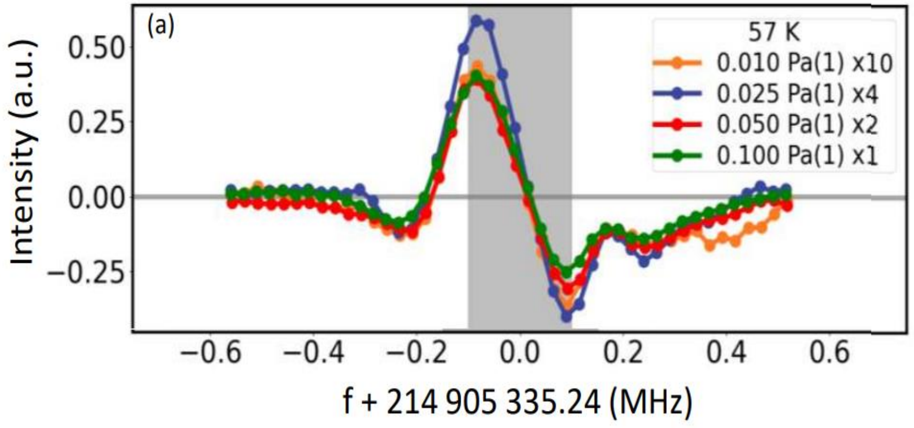
[1] M. Lamperti *et al*., Nat. Comm. **6**, 67 (2023)

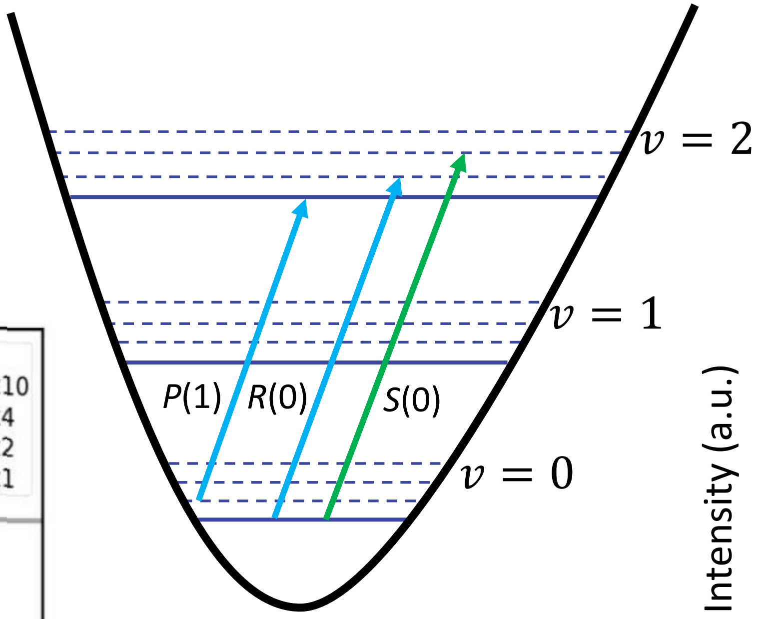


HD vs H2

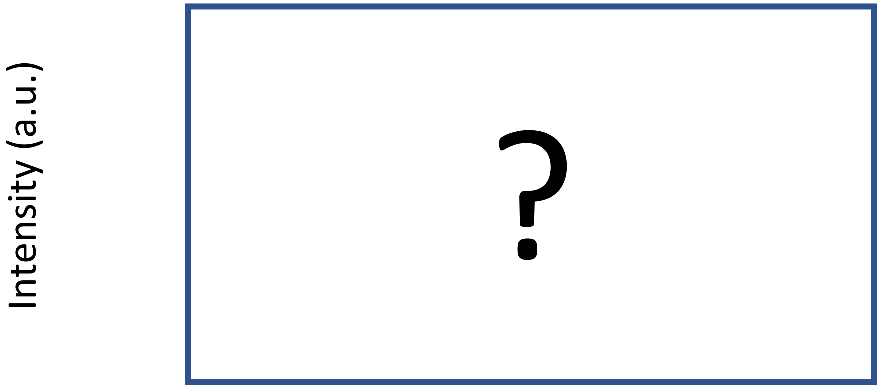


Previous R(0) result:





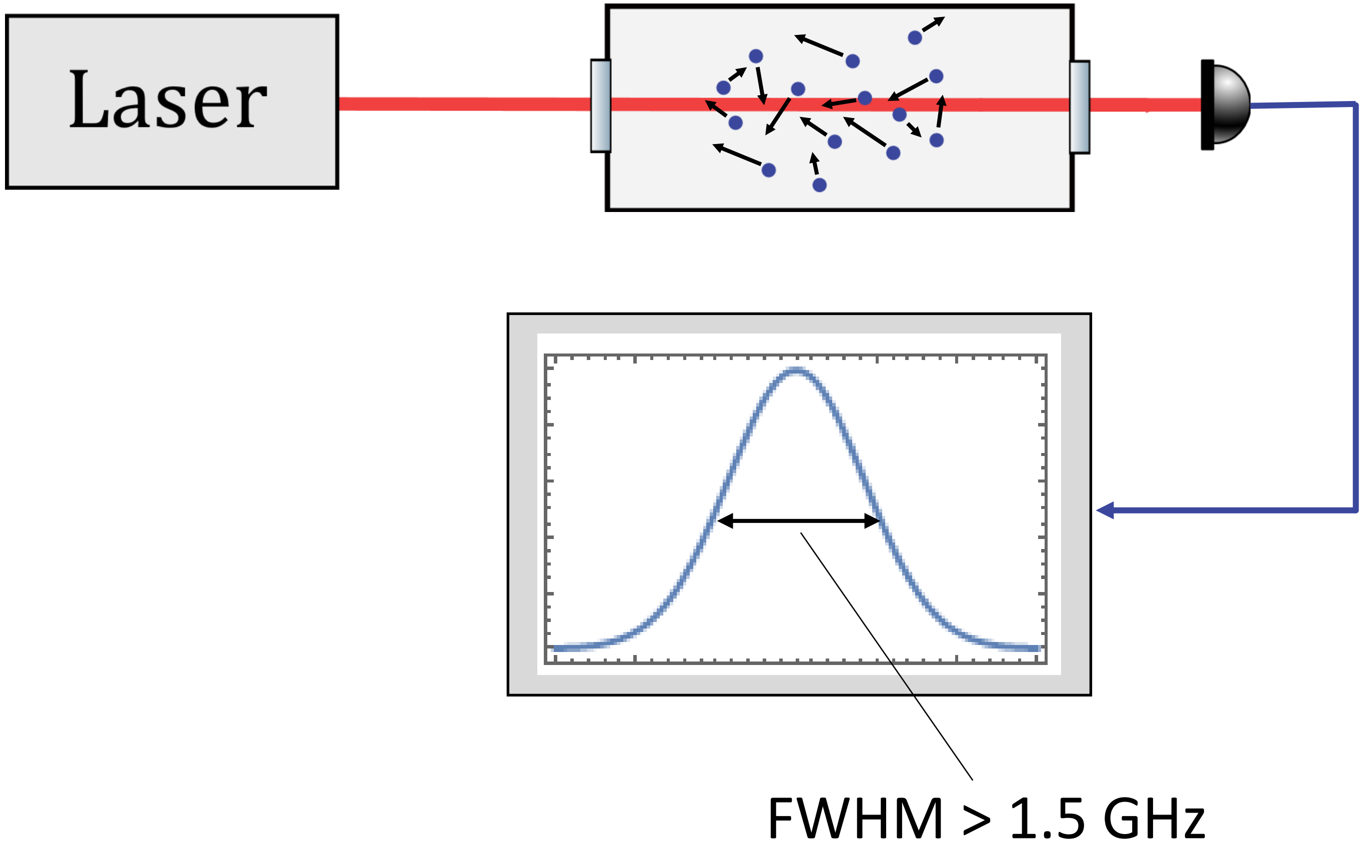
H2 S(0)



Rovibrational transitions of hydrogen: Absorption spectroscopy

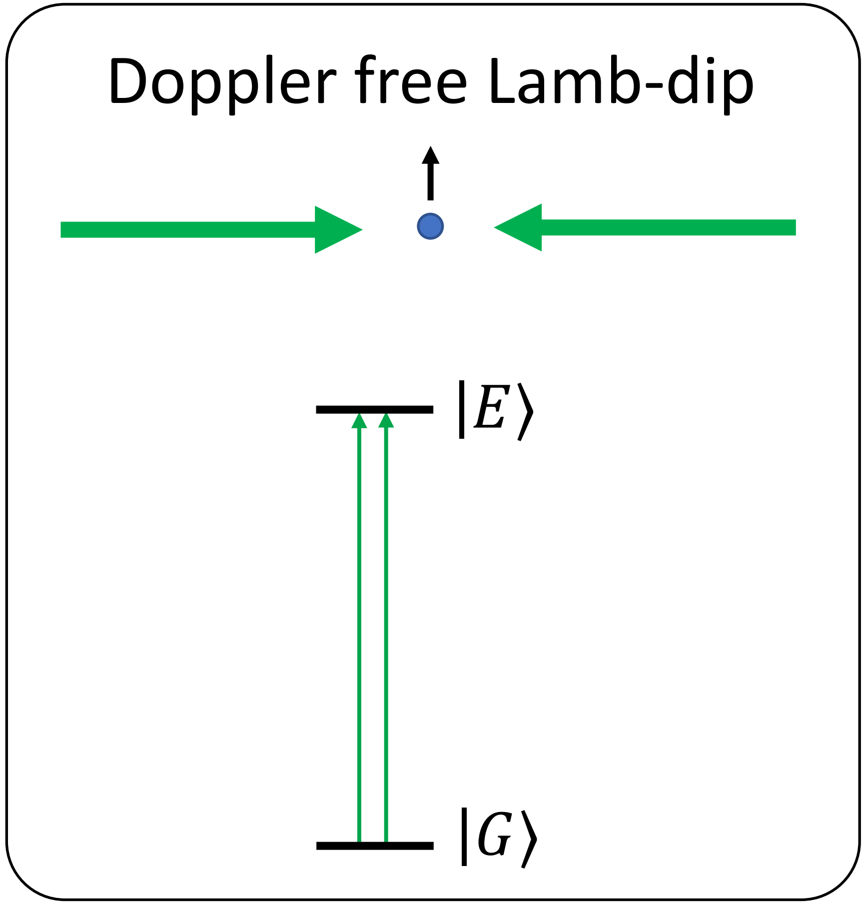
- Doppler broadened absorption spectroscopy
  - Most recent measurement of S(0) in 1982 at 60 MHz accuracy[1]
  - Other lines at around 30 MHz accuracy[2]

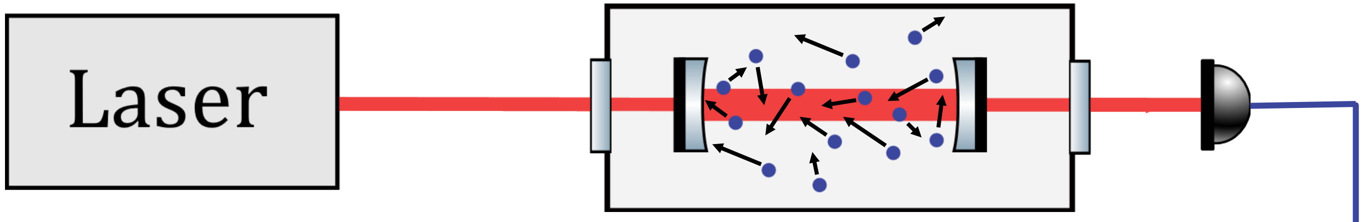
- 33 kHz accuracy best result on HD R(1) transition by a Doppler broadened measurement [3]



[1] S.L. Bragg, W.H. Smith, J. W. Brault, Astroph. J. **263**, 999 (1982)
[2] S. Kassi, A. Campargue, J. Mol. Spectr. **300**, 55 (2014)
[3] Kassi *et. al.* Phys. Chem. Chem. Phys., **24**, 23164-23172 (2022)

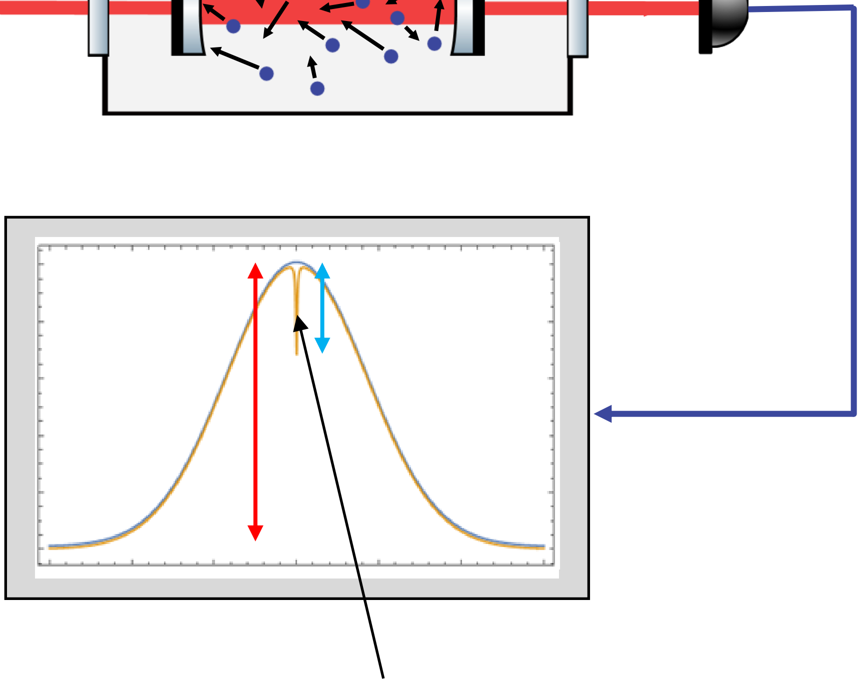
Saturation spectroscopy in H_2





- Lamb-dip in Doppler broadened profile
- Factor 1000 reduction in linewidth

- Sensitivity to detect the profile
- Power to burn the hole

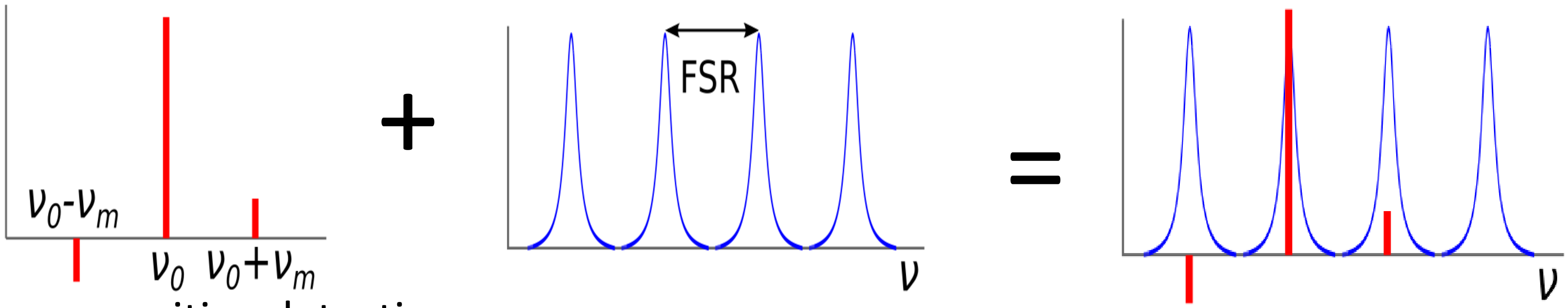


Only ~10 ppm absorption for HD

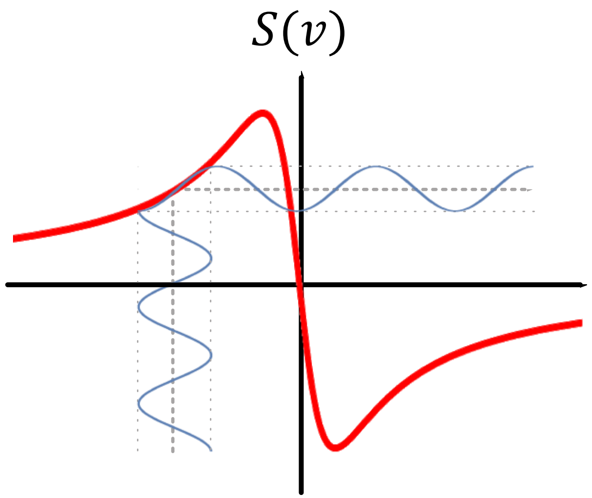
NICE-OHMS:
Noise-Immune Cavity-Enhanced Optical Heterodyne Molecular Spectroscopy

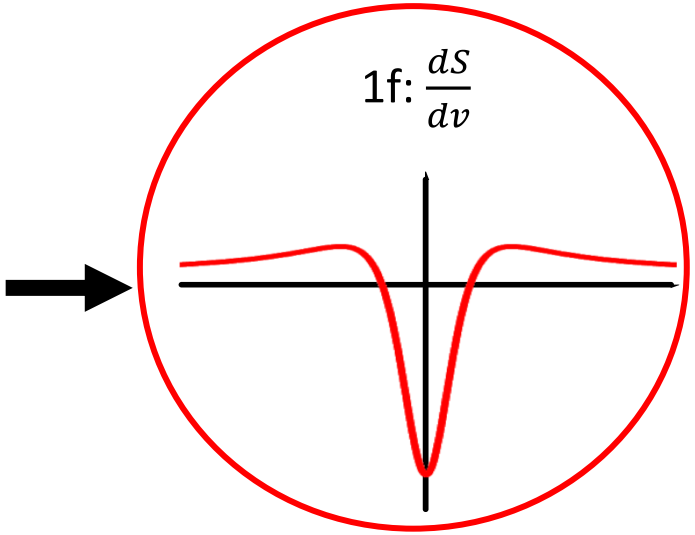
Frequency Modulation Spectroscopy in a cavity

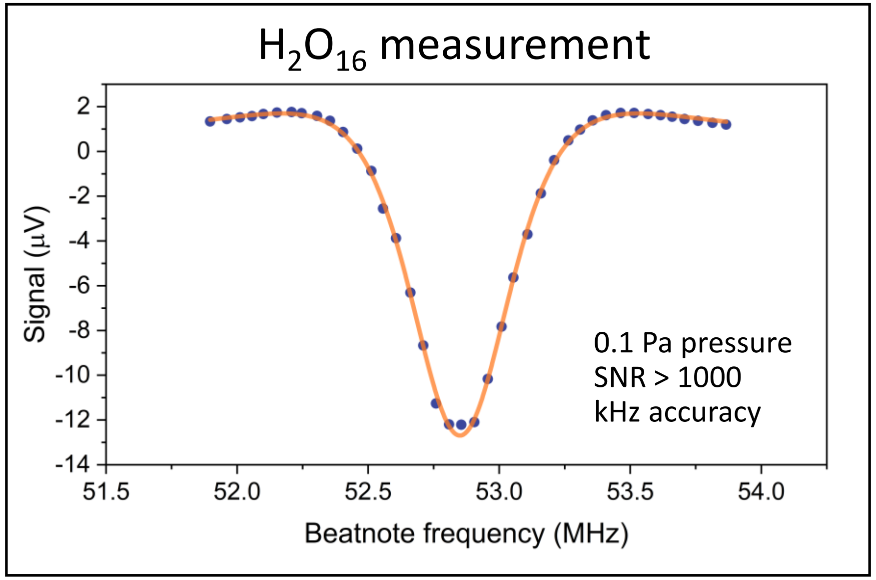
J. Ye, L.S. Ma, and J. Hall, Opt. Lett. **21**, 1000 (1996)



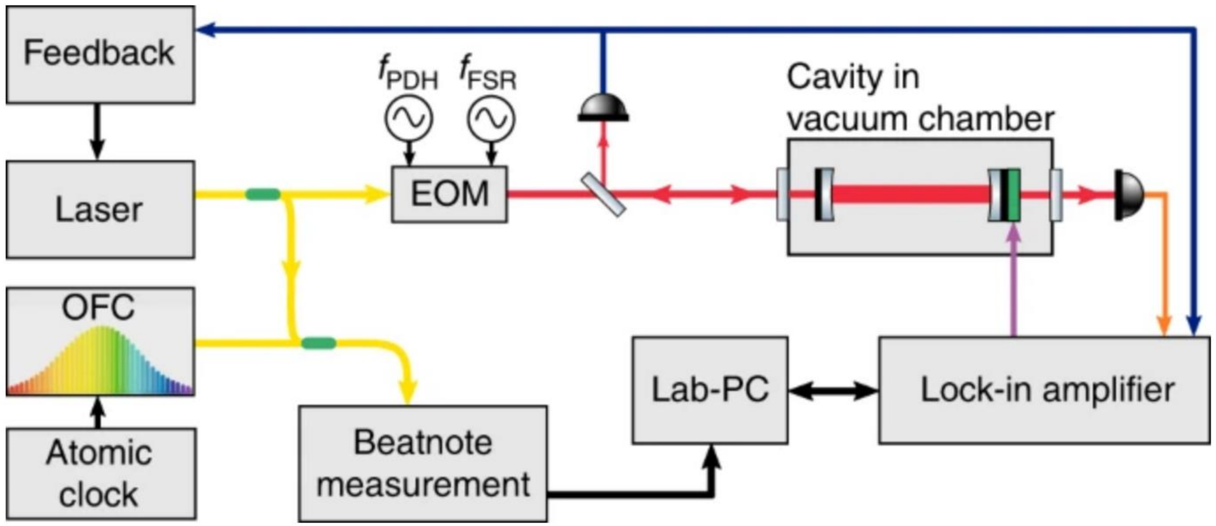
- Phase sensitive detection

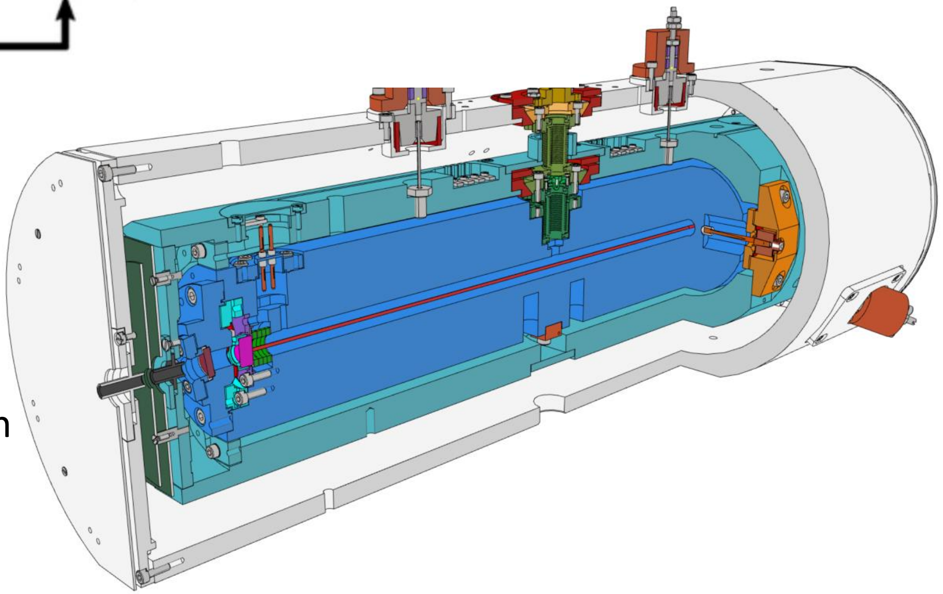


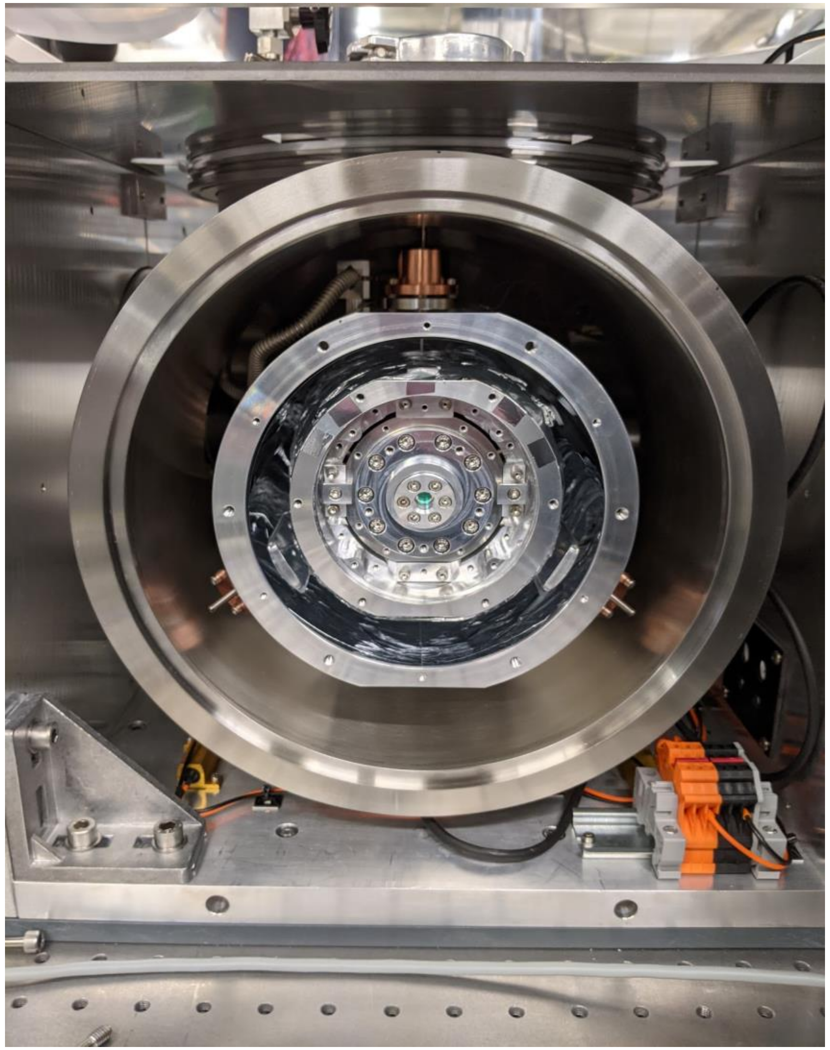




Cryogenic NICE-OHMS setup at VU



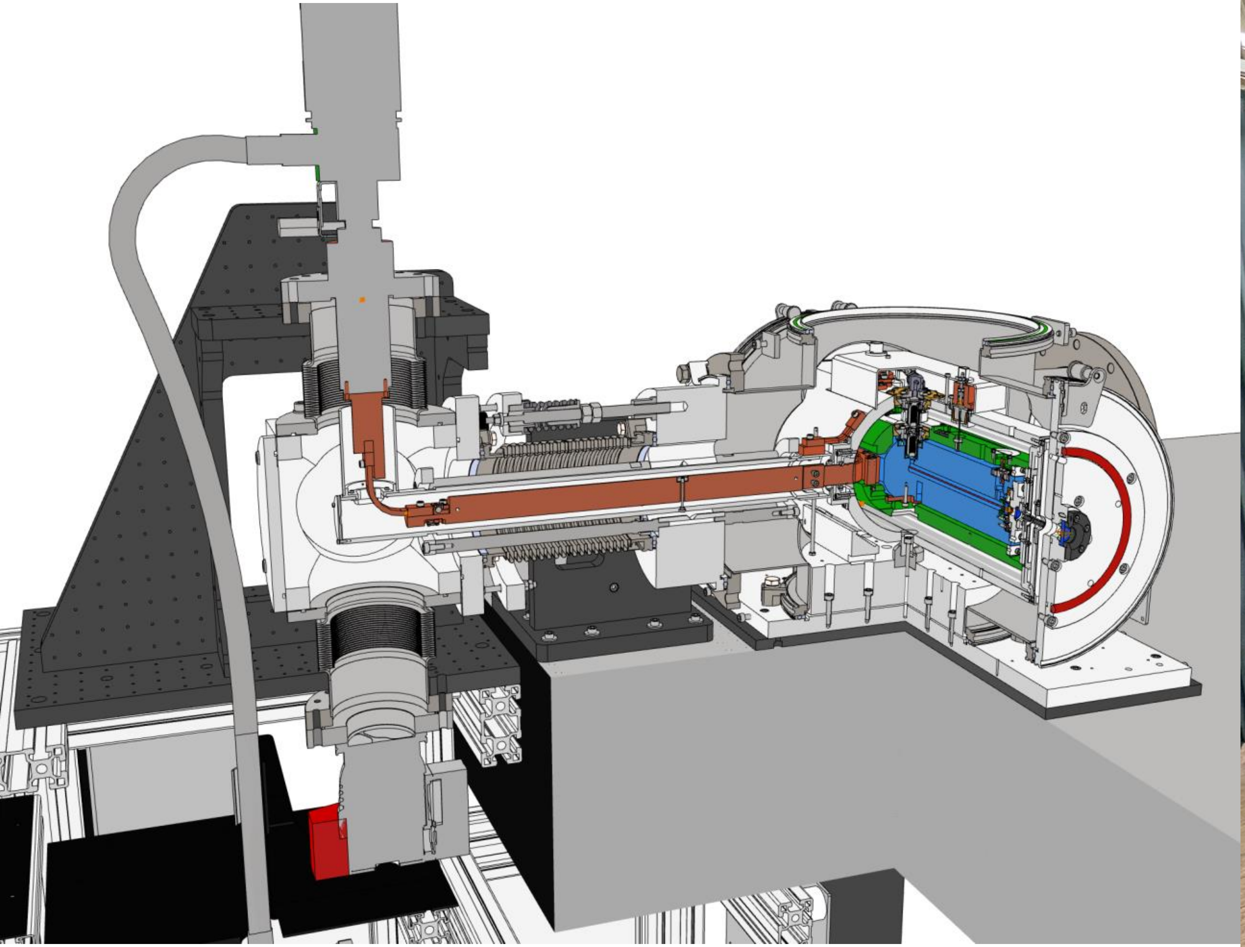


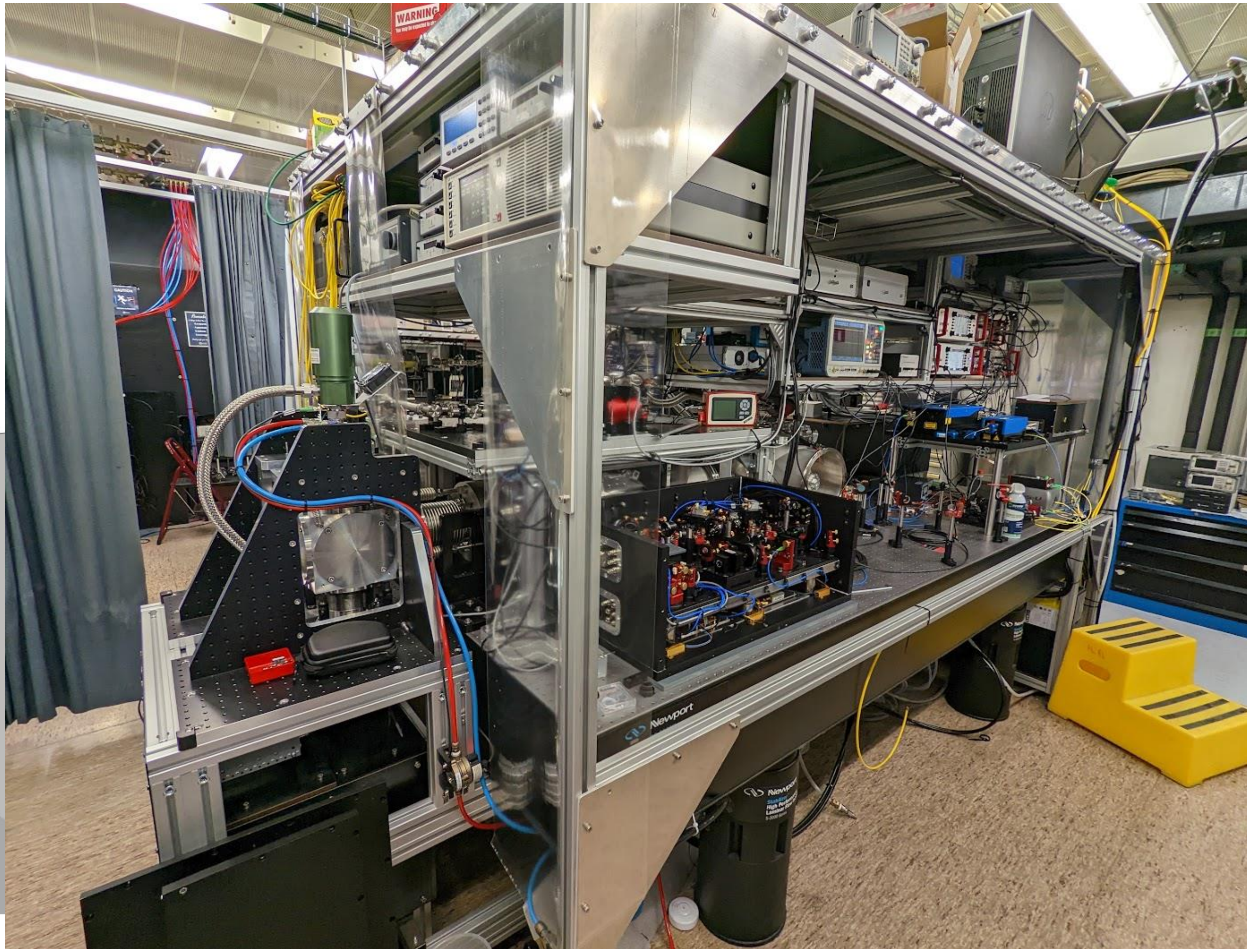


- Finesse of 350 000
- 10 kW roundtrip power
- 55 K limit

- High stability
  - Cryo-cooler noise suppression
  - Short-term linewidth well below sub-kHz

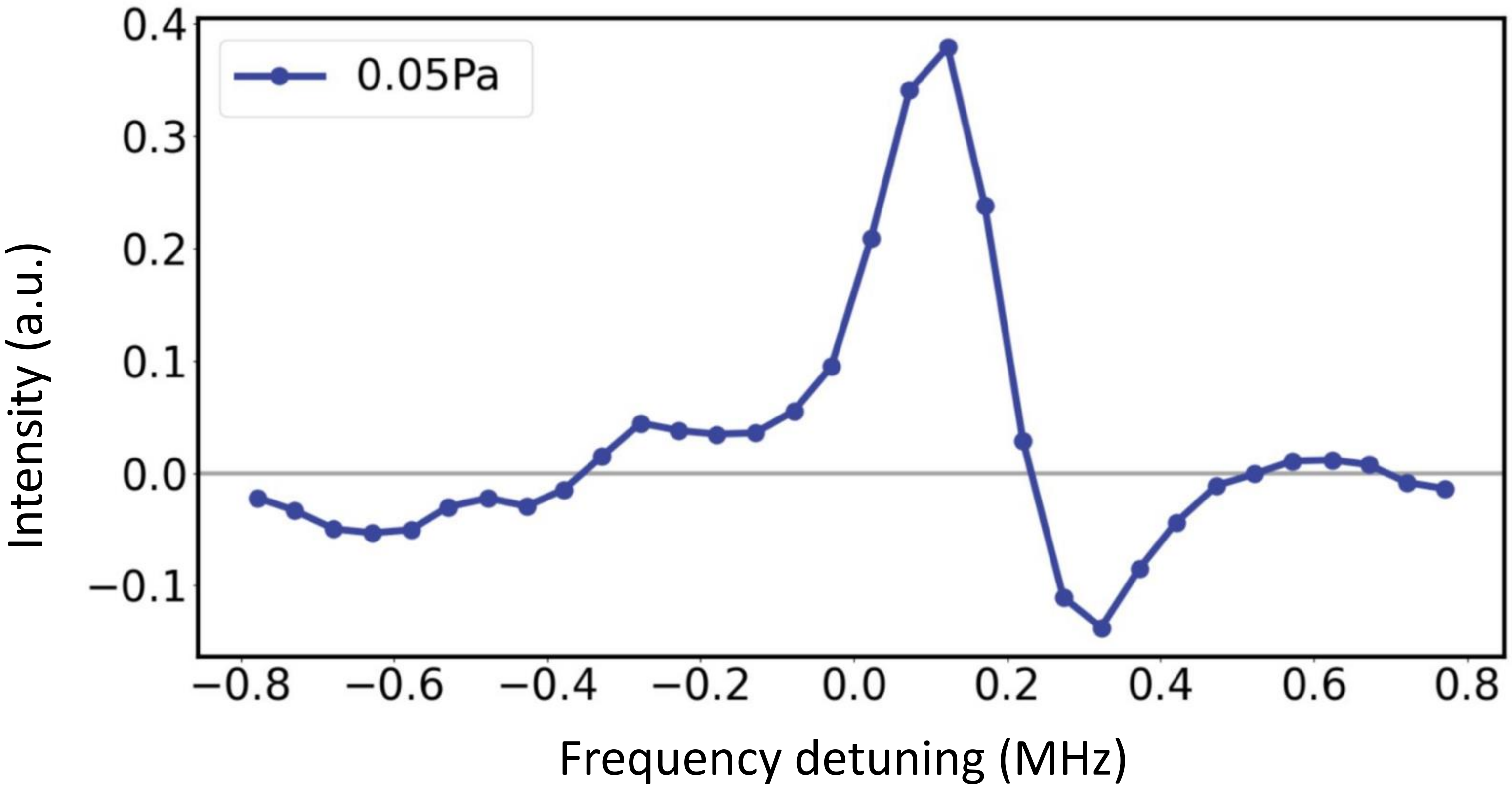
Cryogenic cavity design



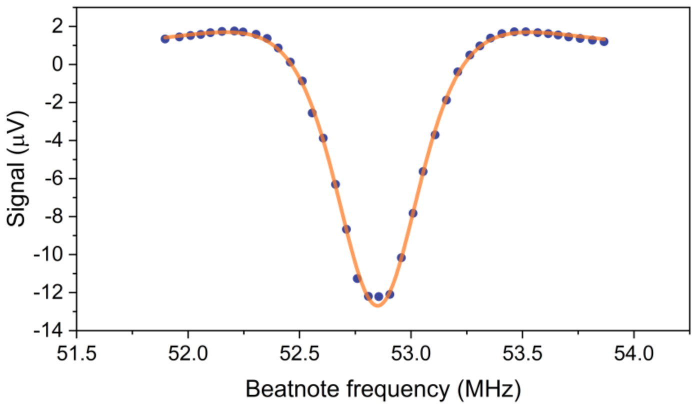


First Results in H2:

S(0) at 8KW



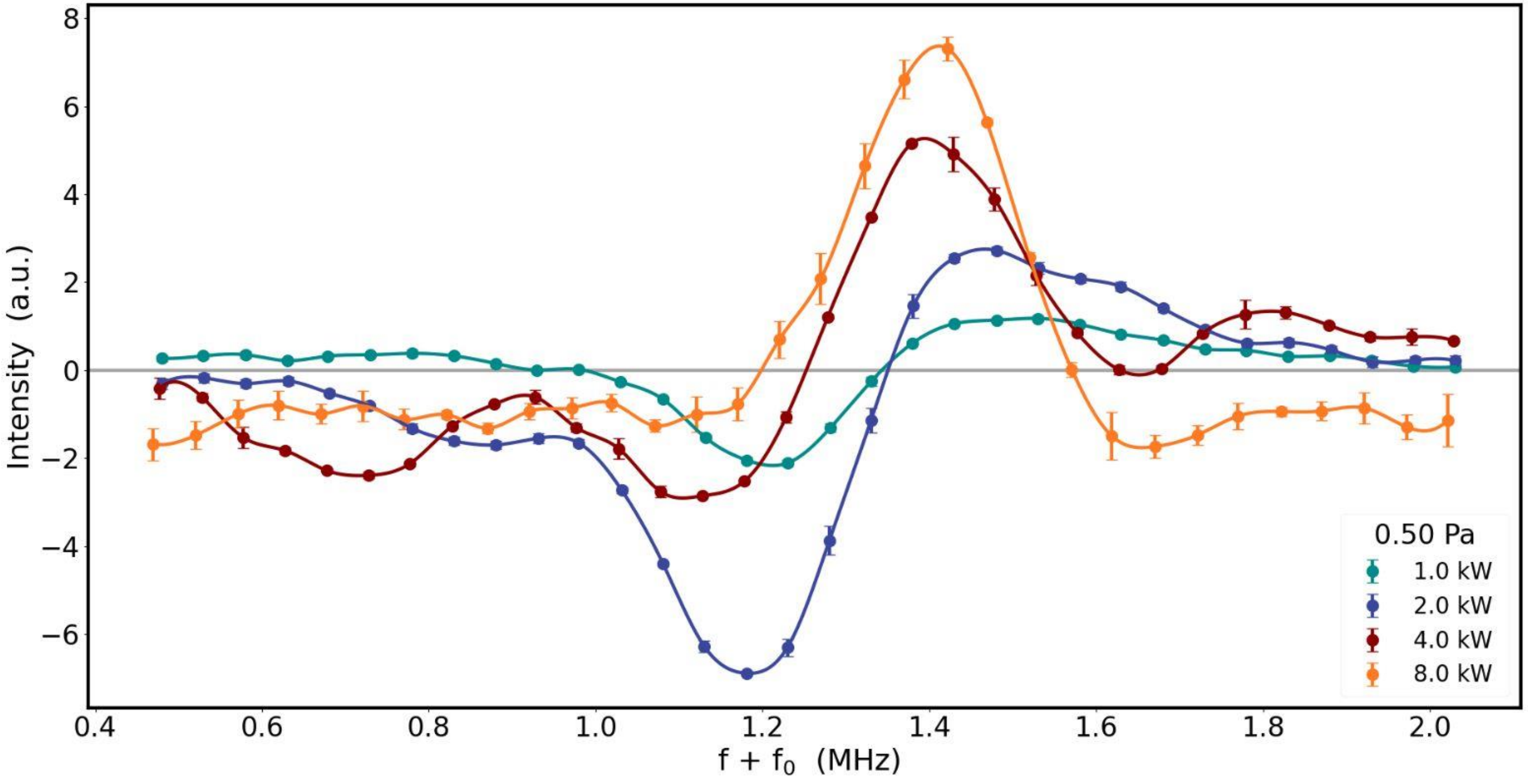
Saturation template



Lineshape puzzling:
Strong dependance with power and pressure

Results in H2:
What if we lower the power?

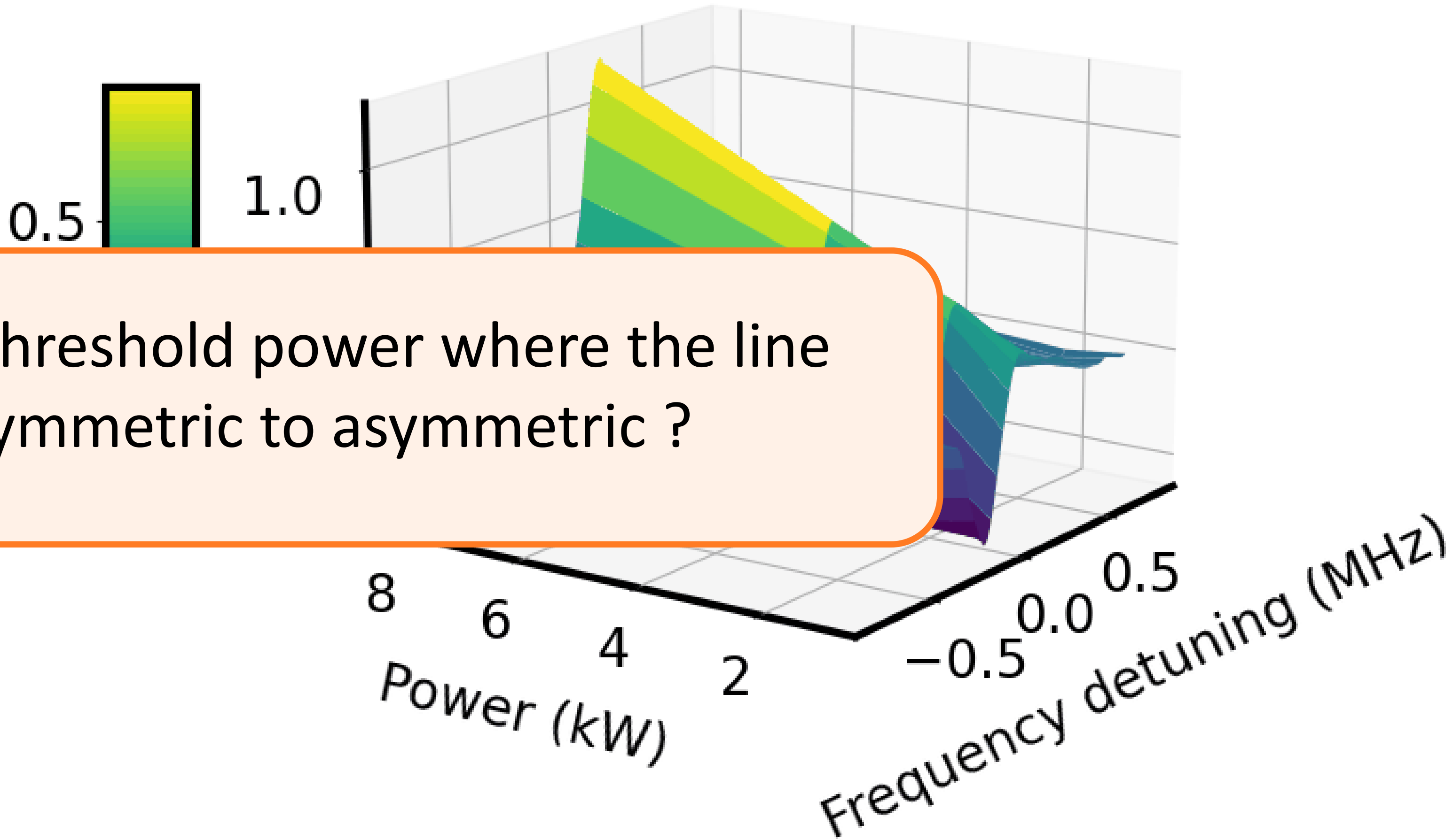
- Varying from 8 kW to 1 kW of power
- Signal transforms from peak to dip!
- Still plenty of signal



Frequency detuning (MHz)

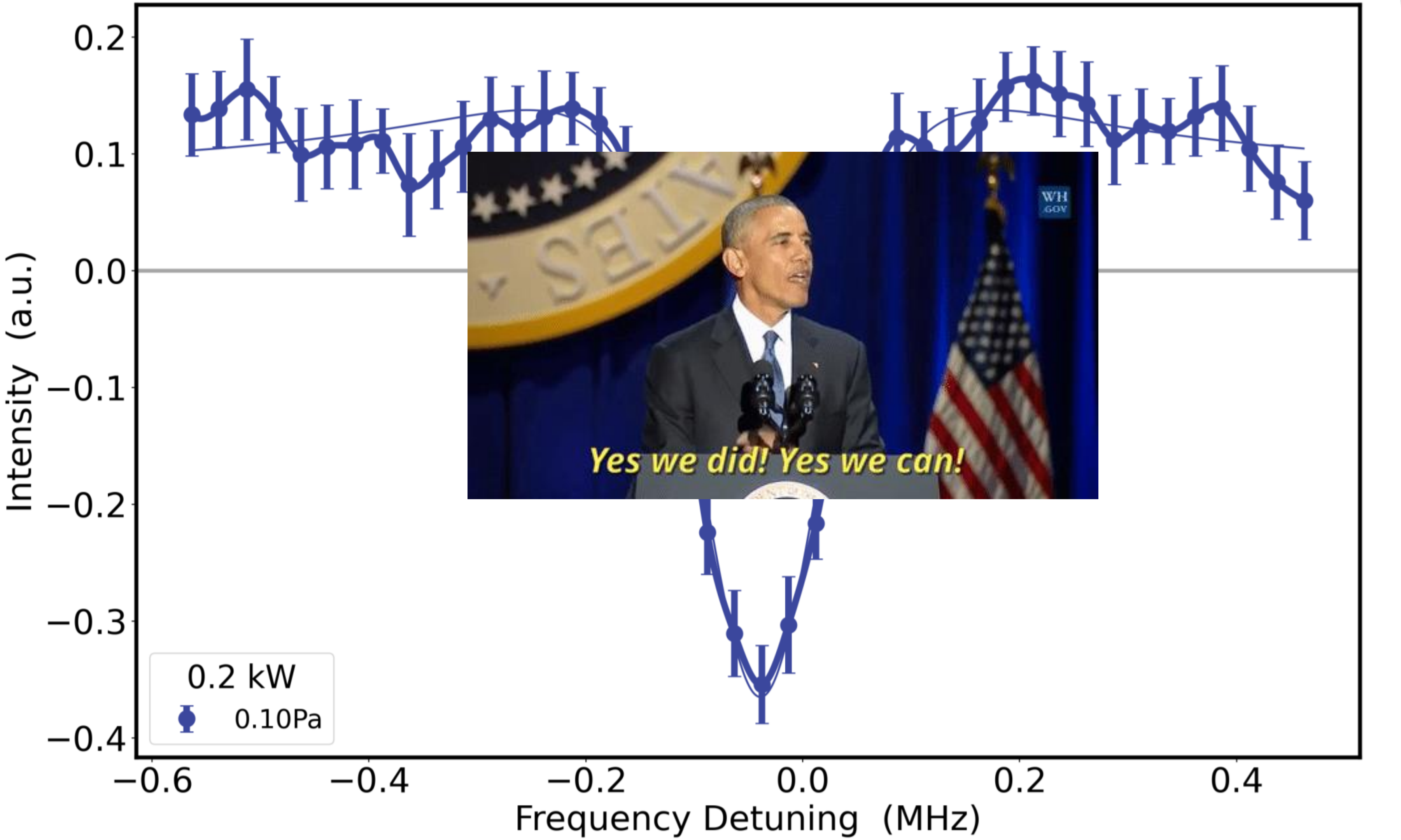
Results in H2: What if we lower the power?

- Do we form a Lamb-dip?
- If so, the trend is clear, we need to lower the power



Can we find the threshold power where the line goes from symmetric to asymmetric ?

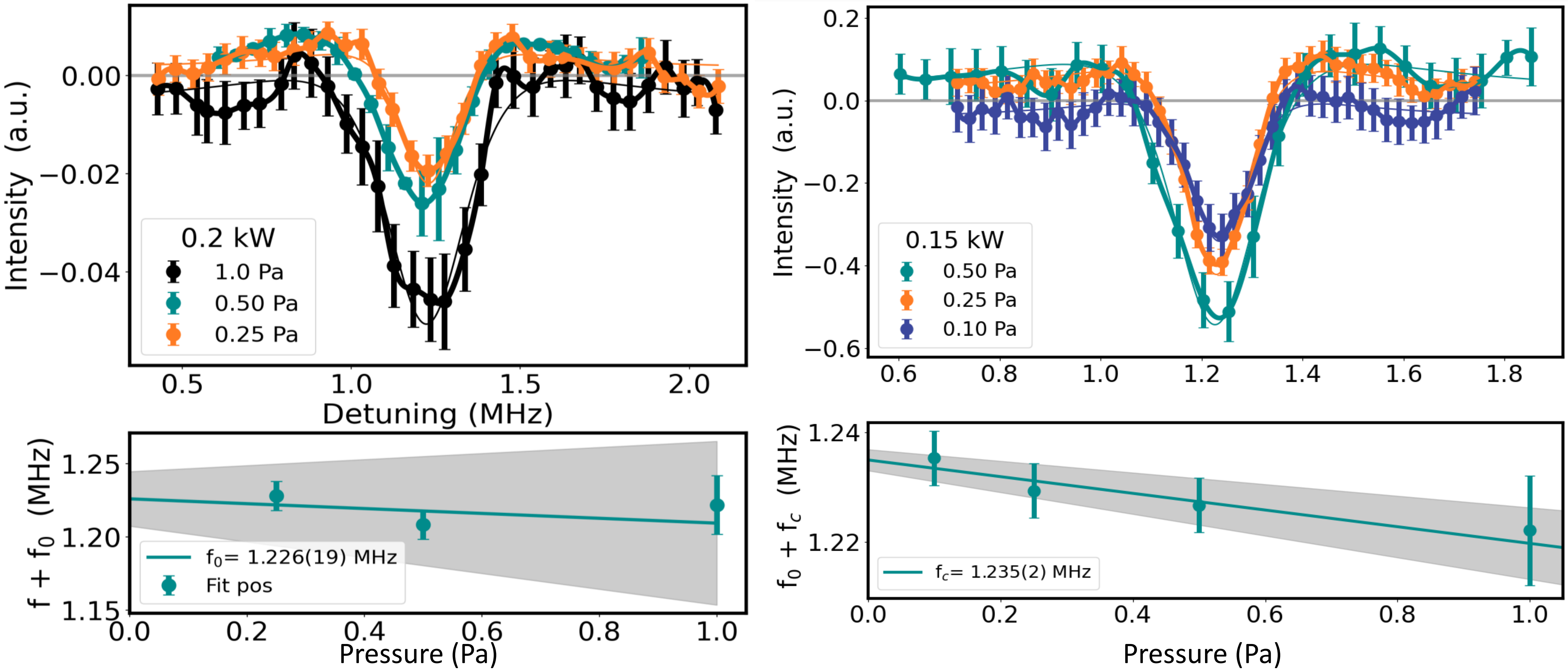
H_2 S(0)
Low power



0.1 Pa @ 200 W

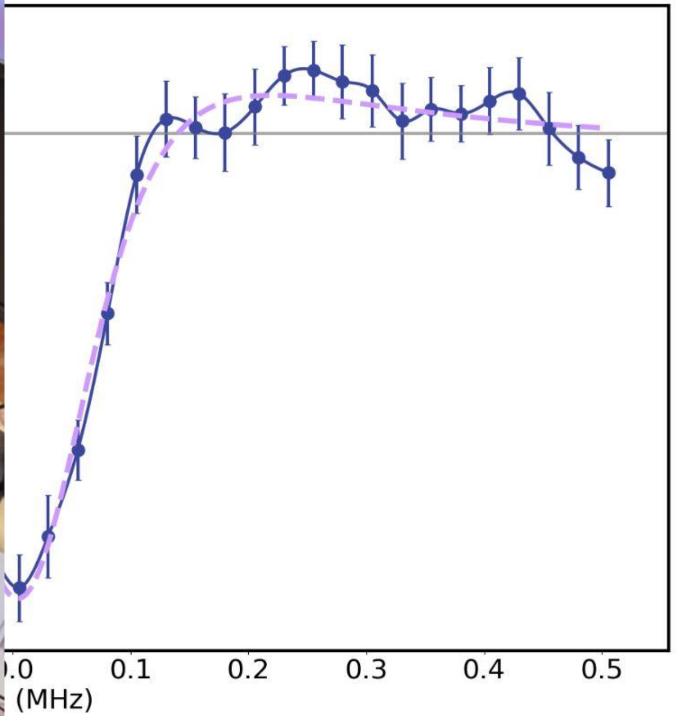
Symmetric lineshape

H_2 S(0) position extraction



Results in H_2 :
Low power measu

- <300 W
- Transformation to perfe
  - Fitting accuracy of a few
- Twice narrower than tra predicts
  - Optical selection of cold
  - 13 K effective temperatu
- Zero pressure, zero pow 252 016 361 234.4 (7.3)
  - Blue line marker in f



Extracting the absolute transition frequency

$$E_{\text{photon}} = h\nu_0 \pm \frac{h\vec{k}\cdot\vec{v}}{2\pi} \pm \frac{(h\nu_0)^2}{2mc^2} - \frac{(h\nu_0)v^2}{2c^2}$$

Recoil shift:
70 kHz

What about the sign?

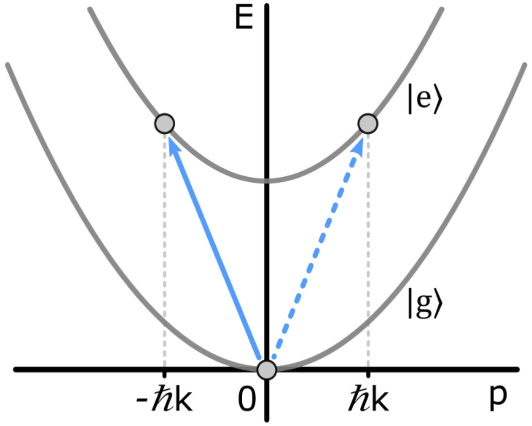
Aborption is +

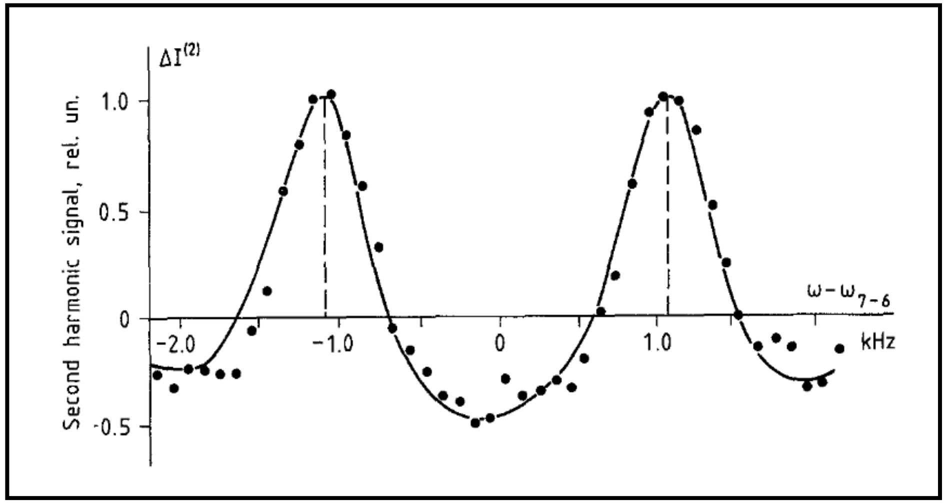
Emission is -

Relativistic Doppler shift:
< 200 Hz
(13 K effective temperature)

Recoil doublet when saturating

Absorption





Doublet unresolvable in most experiments

- kHz splitting
- 100's kHz profile widths
- No shift applied

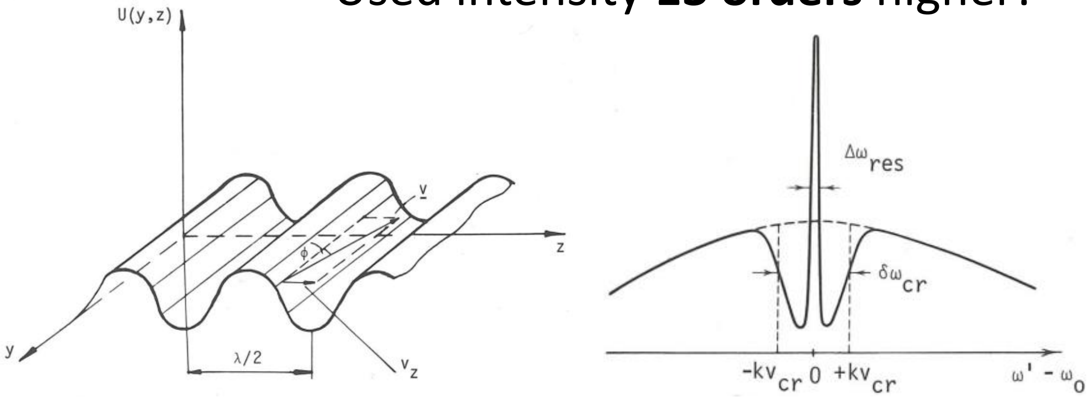
Recoil doublet resolved

- Only 3 experiments so-far
- All in methane

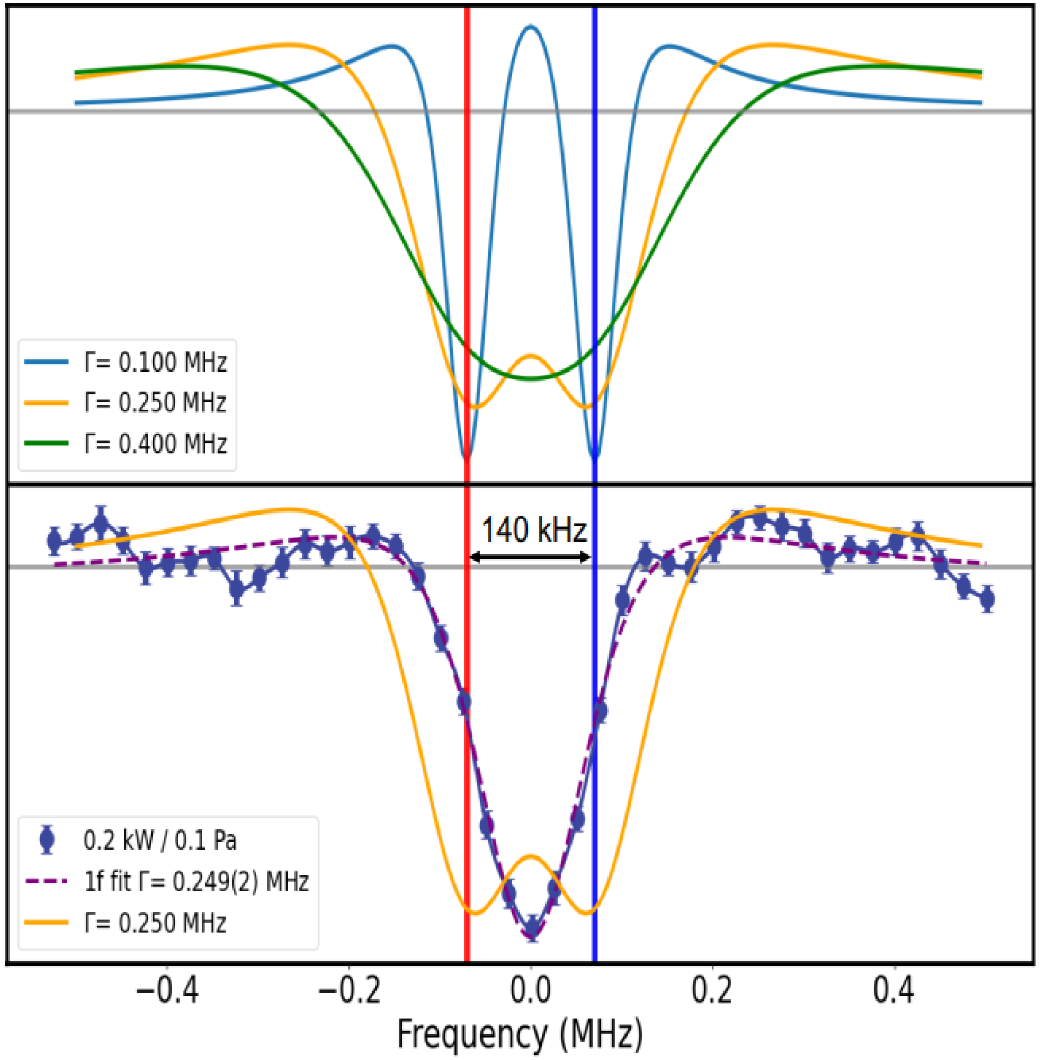
A.P. Kol'chenko, S.G. Rautian, R.I. Sokolovskii, Zh. Eksp. Teor. Fiz. **55**, 1864-1873 (1968)
S.N. Bagayev, V.P. Chebotayev, A.K. Dmitriyev et. al., Appl. Phys. B **52**, 63–66 (1991)

Recoil in H_2

- H_2 is the lightest molecule
  - Recoil splitting of 140 kHz
  - Linewidth of 250 kHz
- Single Lorentzian fits perfectly
  - Model is significantly broader
  - One recoil component is suppressed
- Mechanisms of suppression
  - Pressure → collisions
  - Power → standing wave forces
    - Used intensity **13 orders** higher!

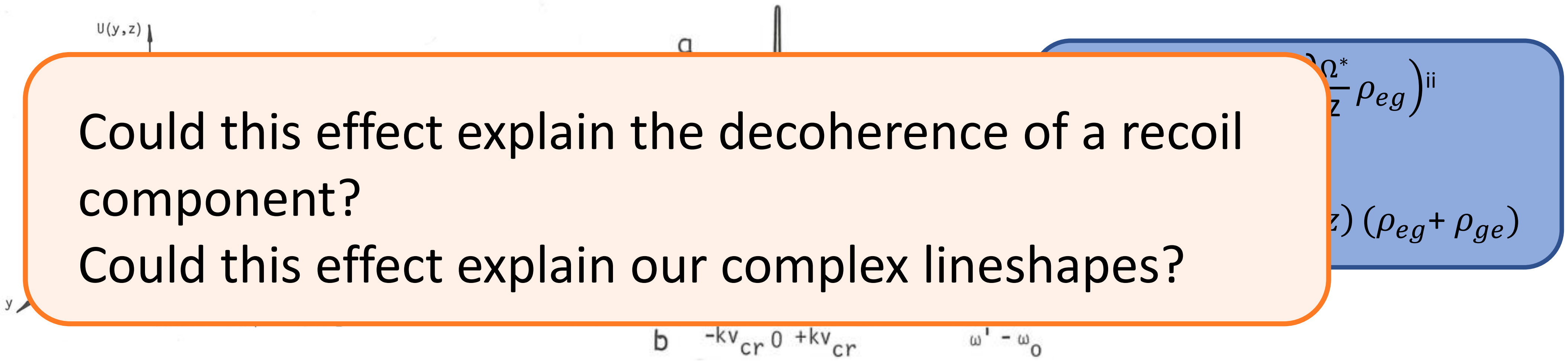


"Nonlinear Laser Spectroscopy", V.S. Letokhov, V.P. Chebotayev (1977)



Possible 1D-lattice effects?

- Standing wave gives strong axial field gradient
  - Striction force on locally enhanced polarizability is significant
- Effect leads to altered velocity distribution



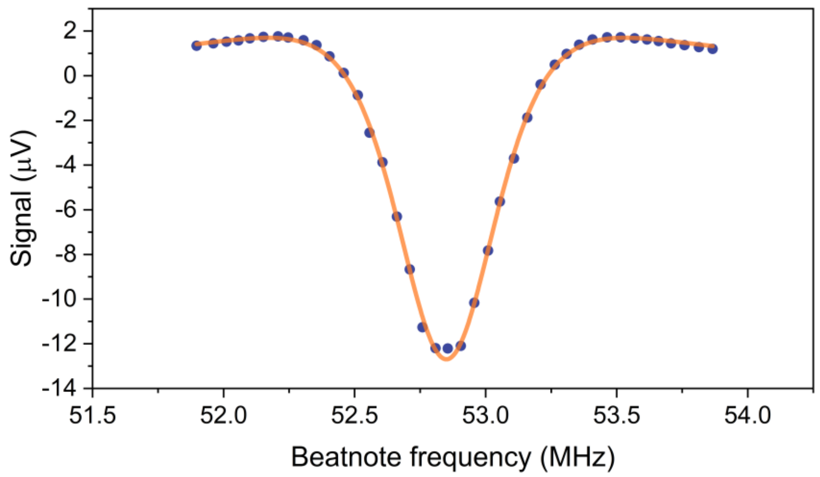
Could this effect explain the decoherence of a recoil component?
Could this effect explain our complex lineshapes?

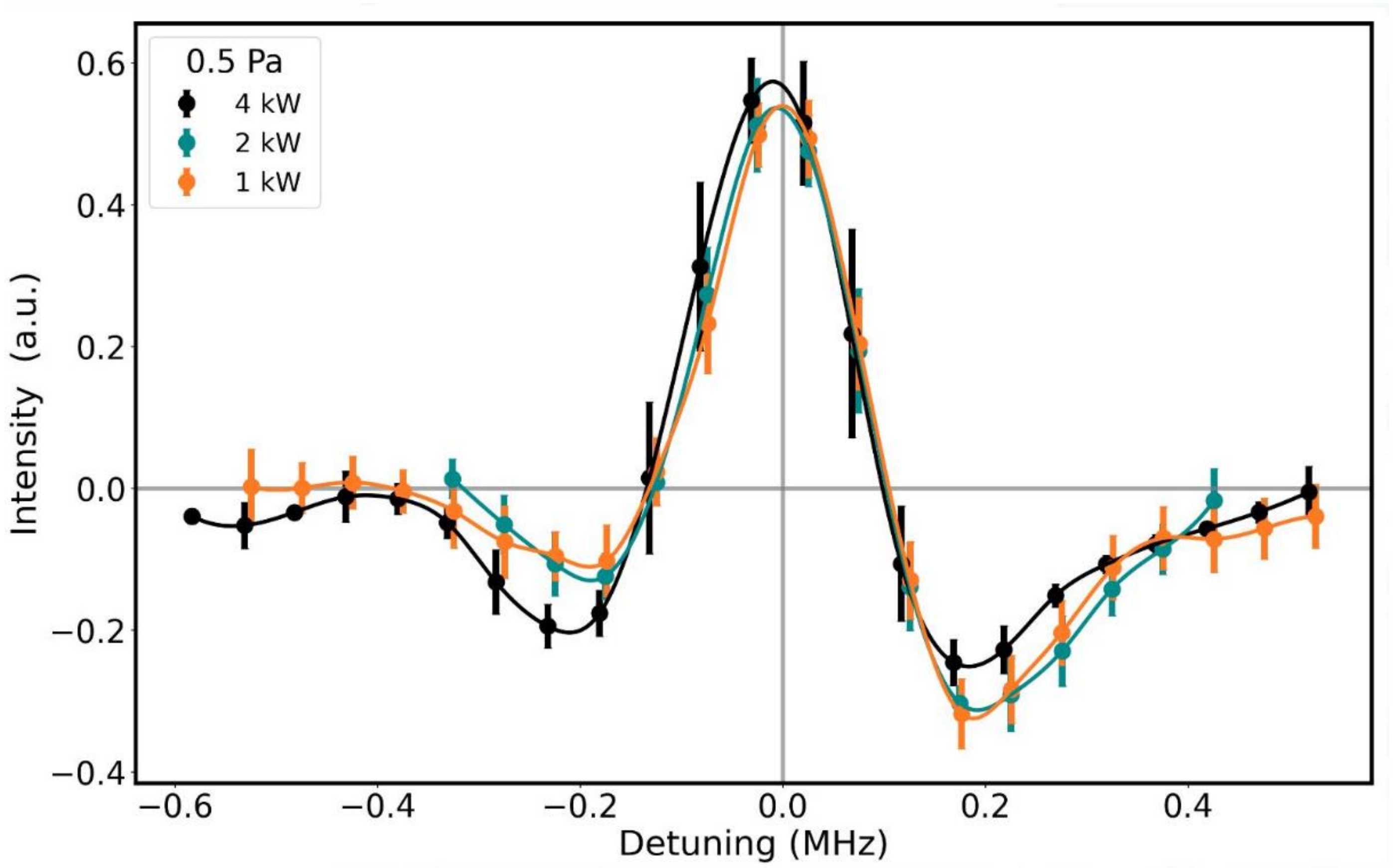
"Nonlinear Laser Spectroscopy", V.S. Letokhov, V.P. Chebotayev (1977)

"Laser Cooling and Trapping", H.J. Metcalf, P. v. d. Strafen (1999)

Peaked lineshape in CO_2

- Fully peaked signal for extremely weak CO2 transition
  - Einstein-A: 1.56E-07 s^{-1}
  - Linestrength of 1.25E-28 cm mol^{-1}
  - R24e at 8247.420872 cm-1

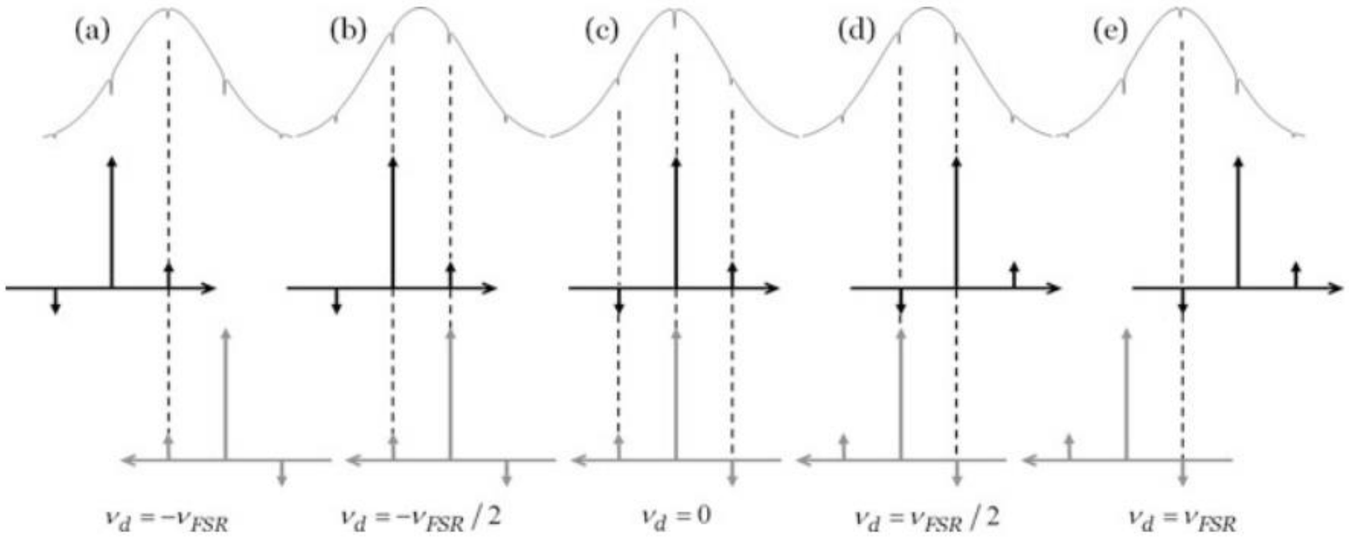




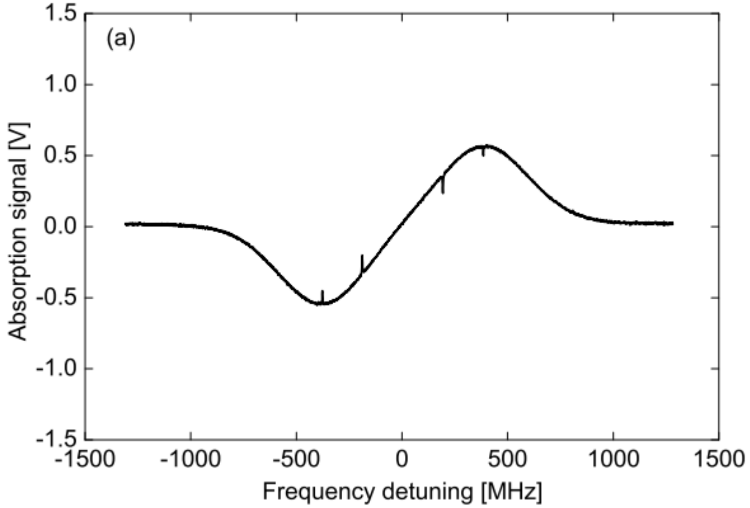
INTERMEZZO:
Sub-Doppler signals in NICE-OHMS

- 3 distinctive saturation schemes
  - 5 signal positions

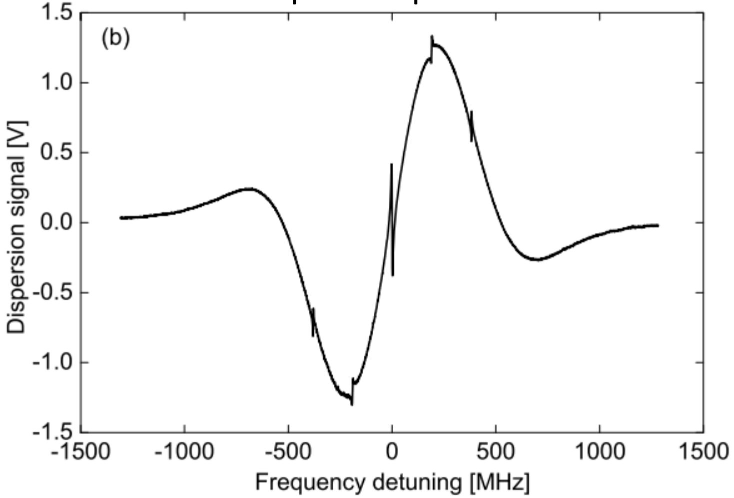
- Central position: Carrier-Carrier
- FSR/2 detuning: Carrier-Sideband
- FSR detuning: Sideband-Sideband



Absorption phase

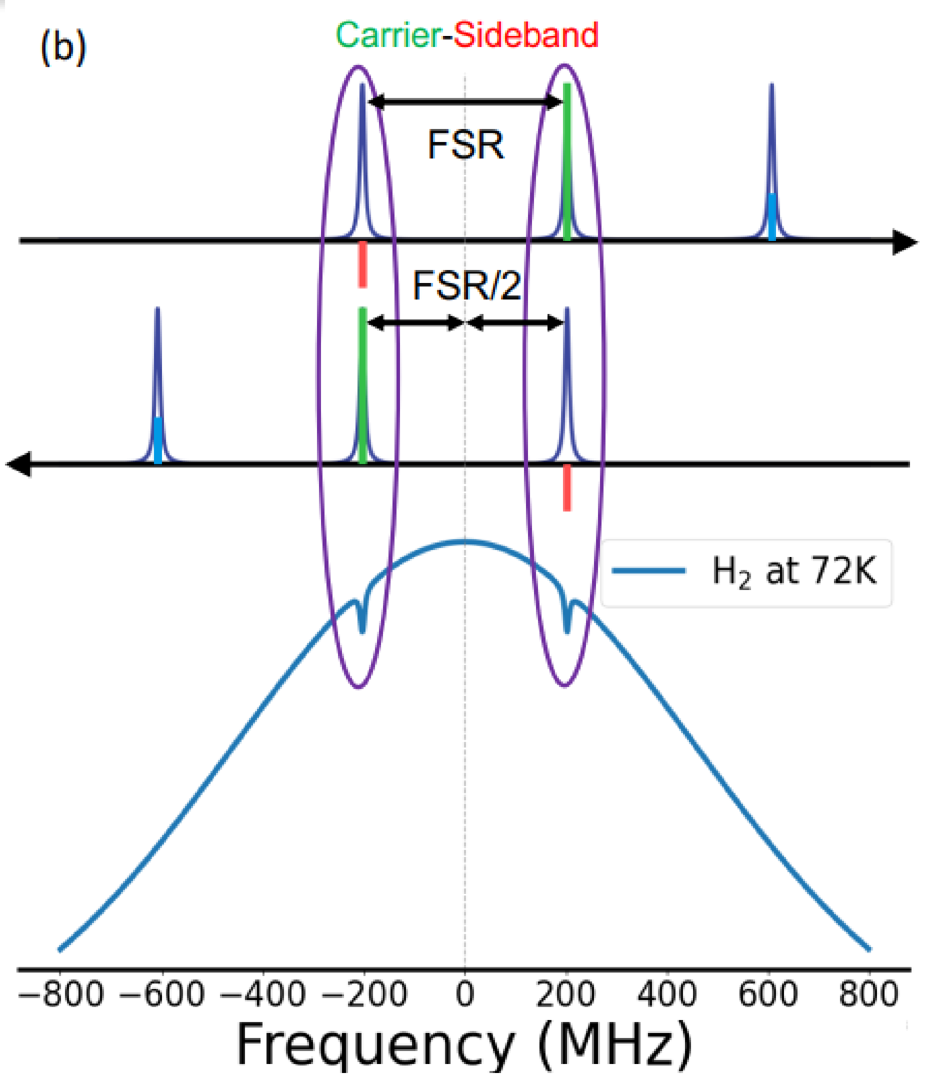


Dispersion phase



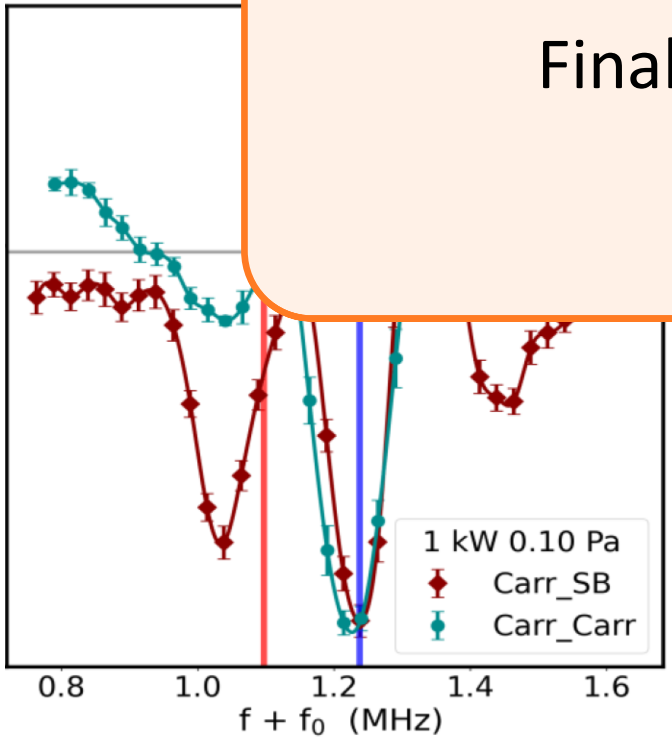
Carrier-Sideband saturation: (pump probe) Doppler-detuned saturation spectroscopy

- Detuned by $\pm$FSR/2
  - Doppler shift cancels detuning
- Molecules fly along the beam
  - Part of velocity is projected radially
  - Reduced radial speed
  - Reduced 2nd order Doppler
  - Increased transit time
- On-resonance traveling wave
- Off-resonance standing wave



Doppler-detuned saturation spectroscopy (pump probe) Results on H2

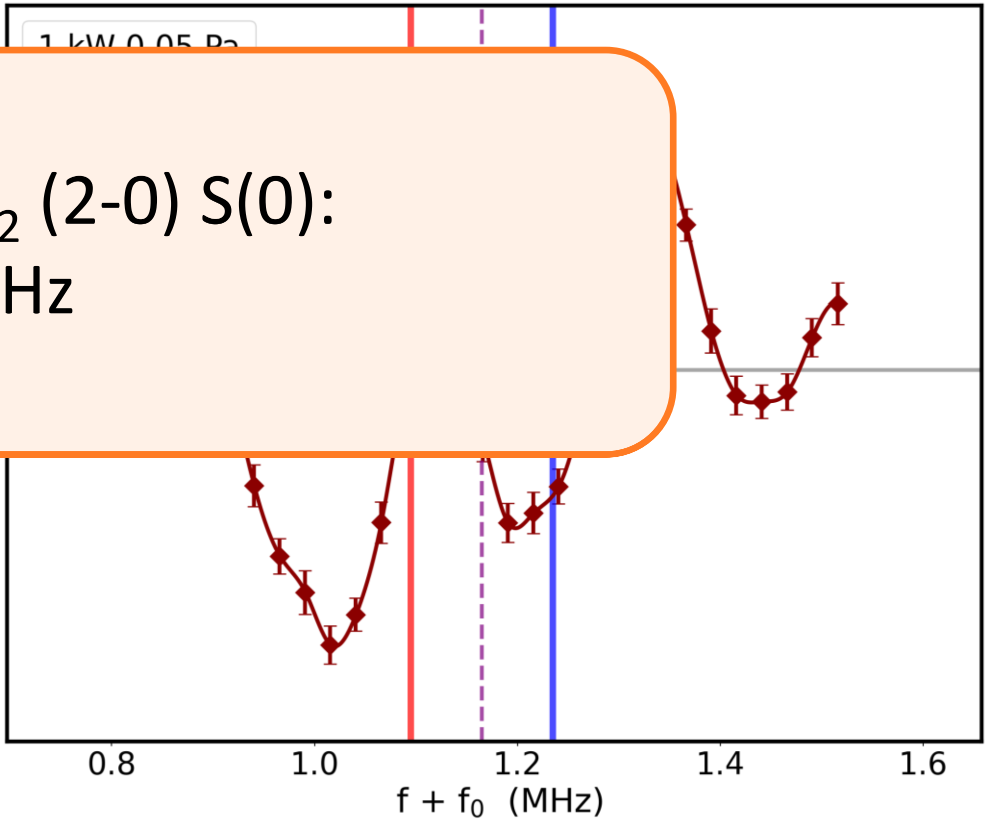
- Saturating molecules flying along the beam
- Doppler shift equal to detuning
- On-resonance standing wave → travelling wave
  - Si



Final transition frequency of H_2 (2-0) S(0):
252 016 361 164 (8) kHz

Recoil doublet recovered

- Asymmetric lineshapes
- Red recoil is suppressed
- Recoil shift must be applied



Conclusion of H_2

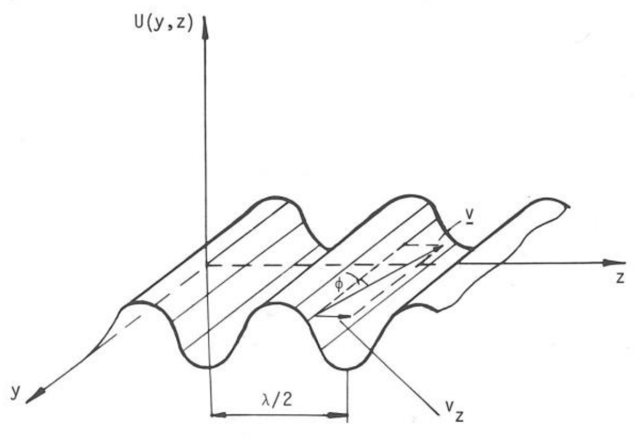
- Molecular quadrupole transition saturated for the first time
  - New-era in rovibrational saturation spectroscopy
- H_2 transition frequency of 252 016 361 164 (8) kHz [i]
  - 3 x 10^{-11} relative accuracy
  - Most accurate determination of any vibrational splitting within all isotopes of molecular hydrogen
  - Recent Doppler Broadened measurement is -65(60)kHz off [ii]
  - Theory is 2.6 MHz (1.6 σ) off ($E^{(5)}$)
    - 1.6 MHz uncertainty
- Recoil splitting observed
  - 2nd molecule ever
  - Absence of red recoil under certain conditions

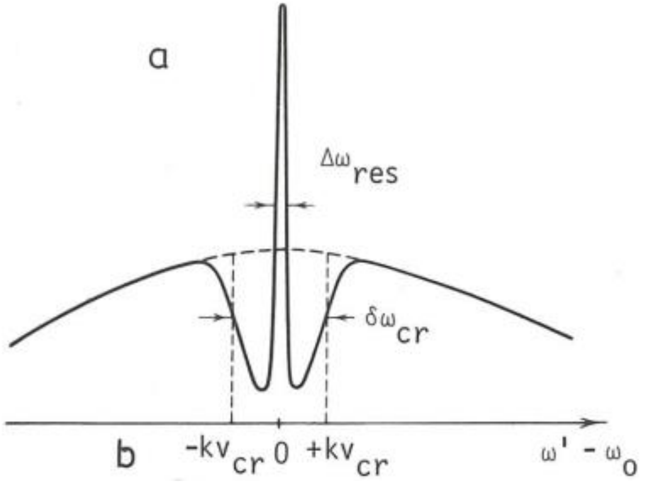
[i] http://arxiv.org/abs/2303.17818

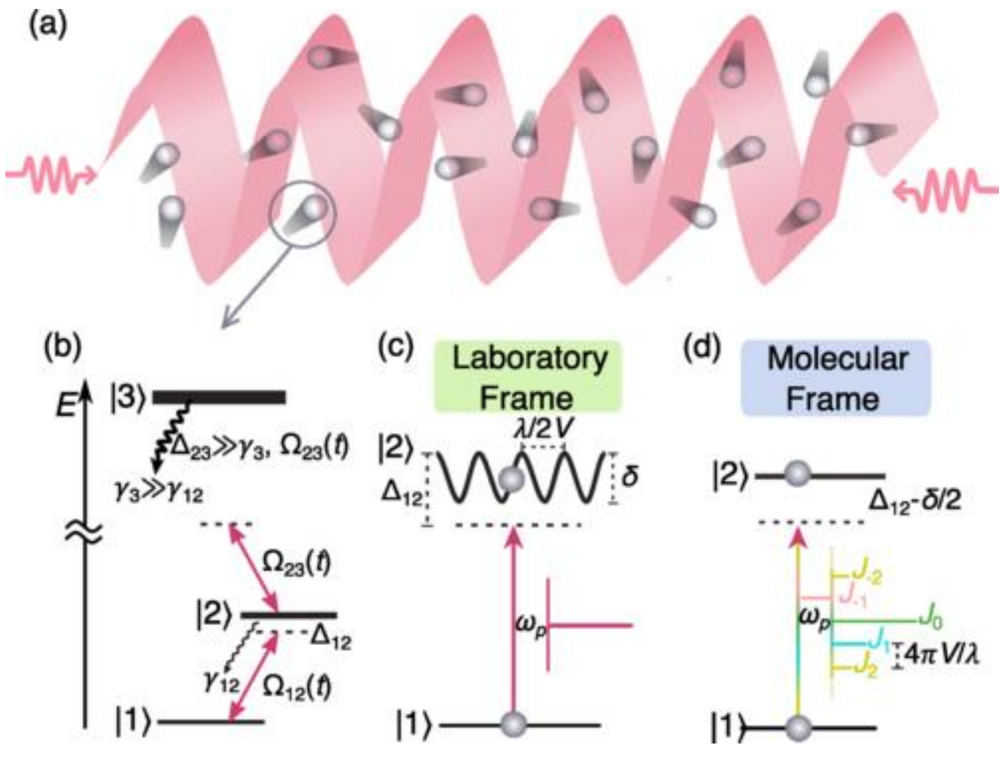
[ii] H. Fleurbaey *et al,* PCCP accepted

Outlook

- HT (radioactive) measurements
- Last targets before the end
  - H_2 Q(1)
  - Understanding lineshapes finally ?
    - $\rightarrow$ Transitions below $10^{-4}s^{-1}$
    - → Standing wave effects







Y.N. Lv *et. al.*, Phys. Rev. Lett. **129**, 163201 (2022)

Acknowledgements & questions

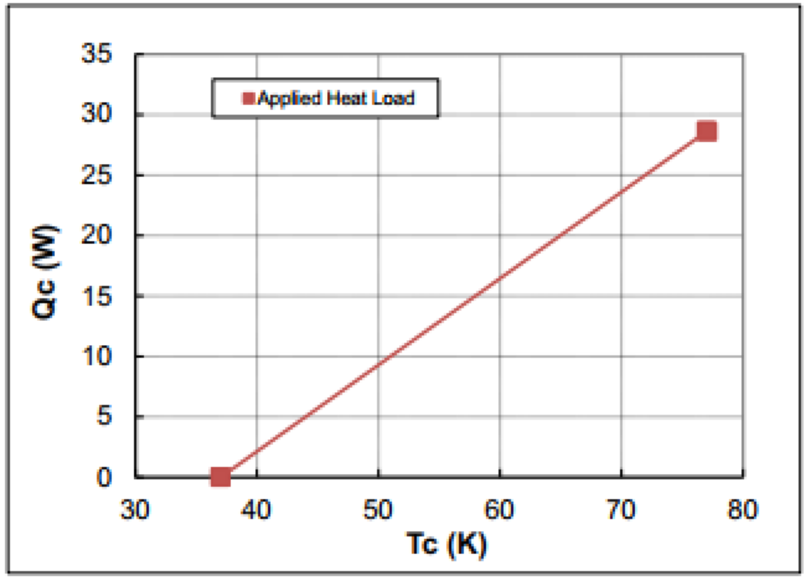
NICE-OHMS team:
Meissa Diouf
Frank Cozijn
Wim Ubachs

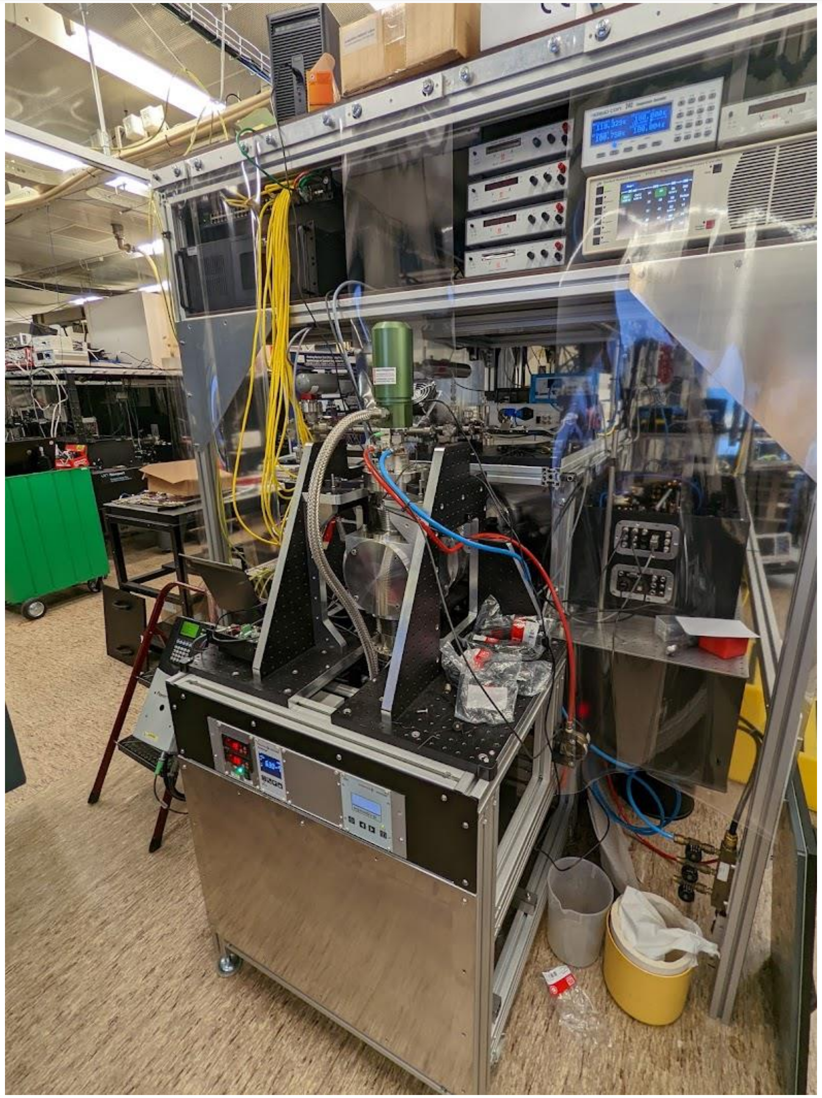
Cryo-cooler

- Thermo acoustic cryo-cooler
  - 60 Hz oscillation with low noise

- Vibration damping works excellent
  - 60 Hz vibration invisible in noise analysis

LOAD CURVE:

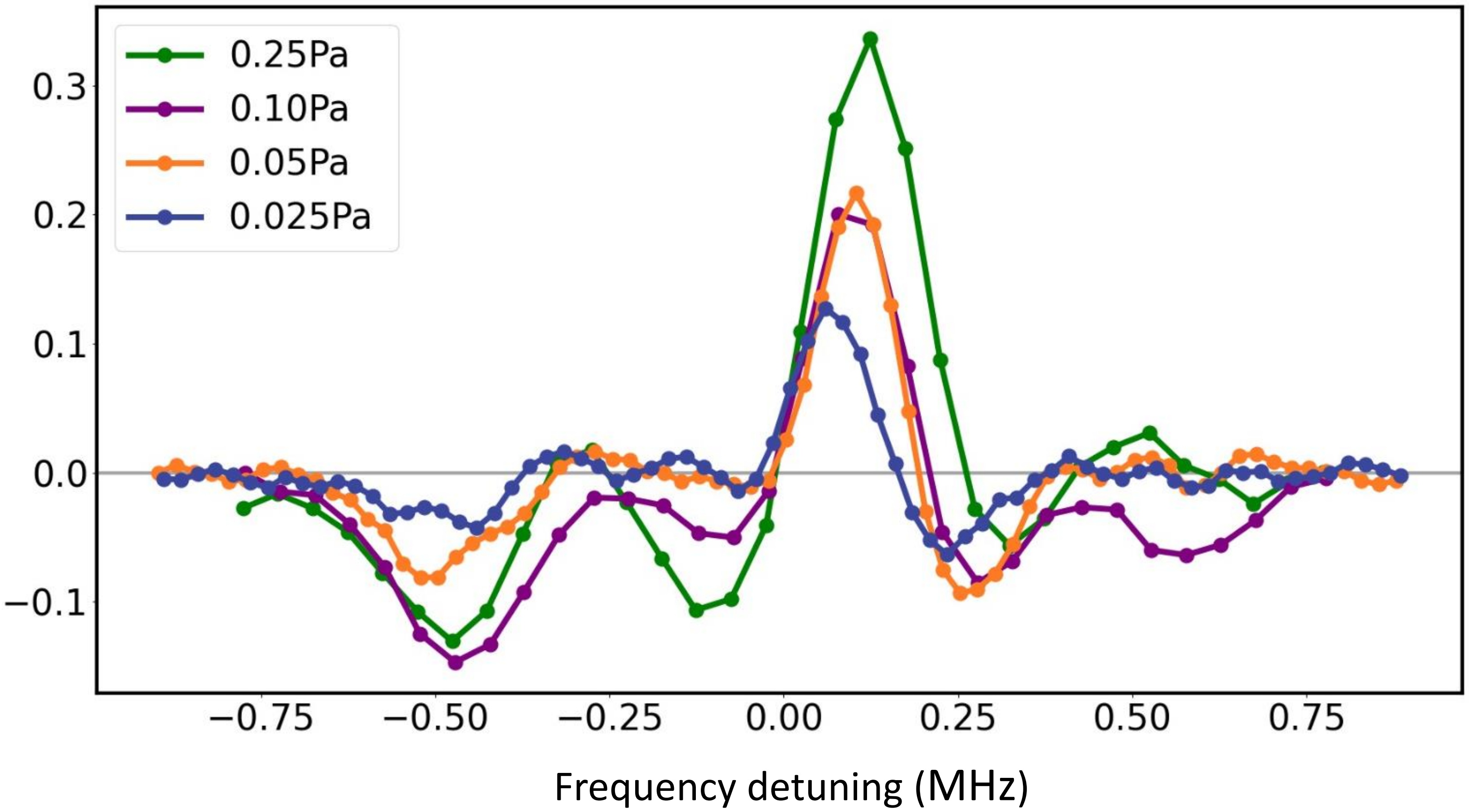




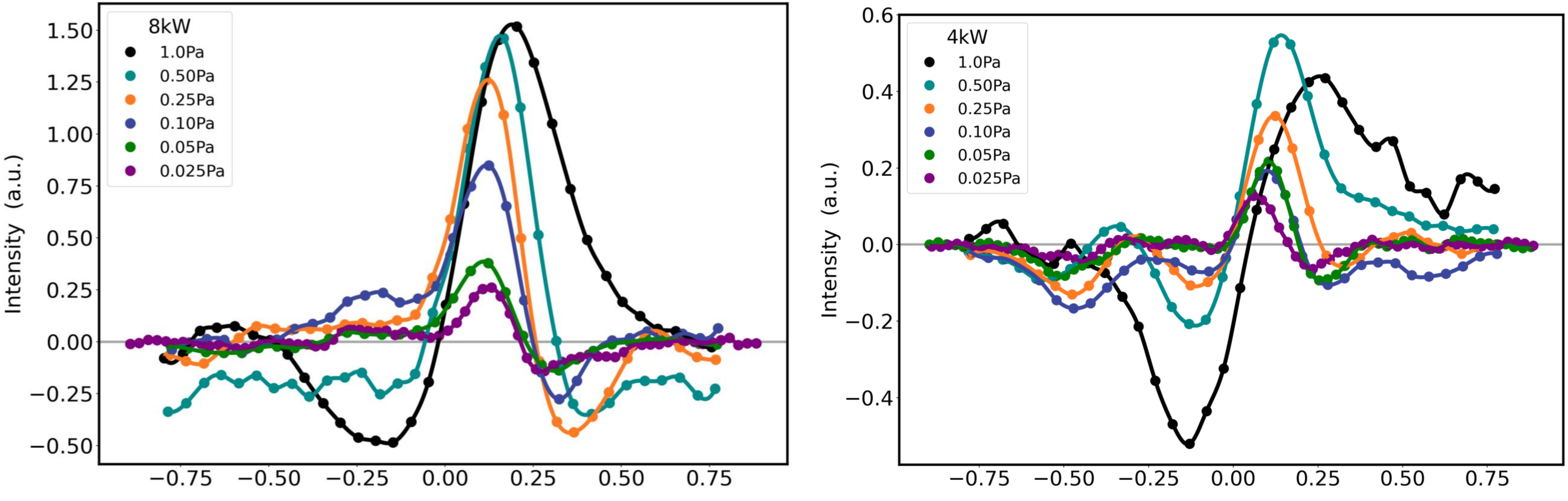
Results in H2: Pressure variation

- 4 kW of power
- 55 K temperature

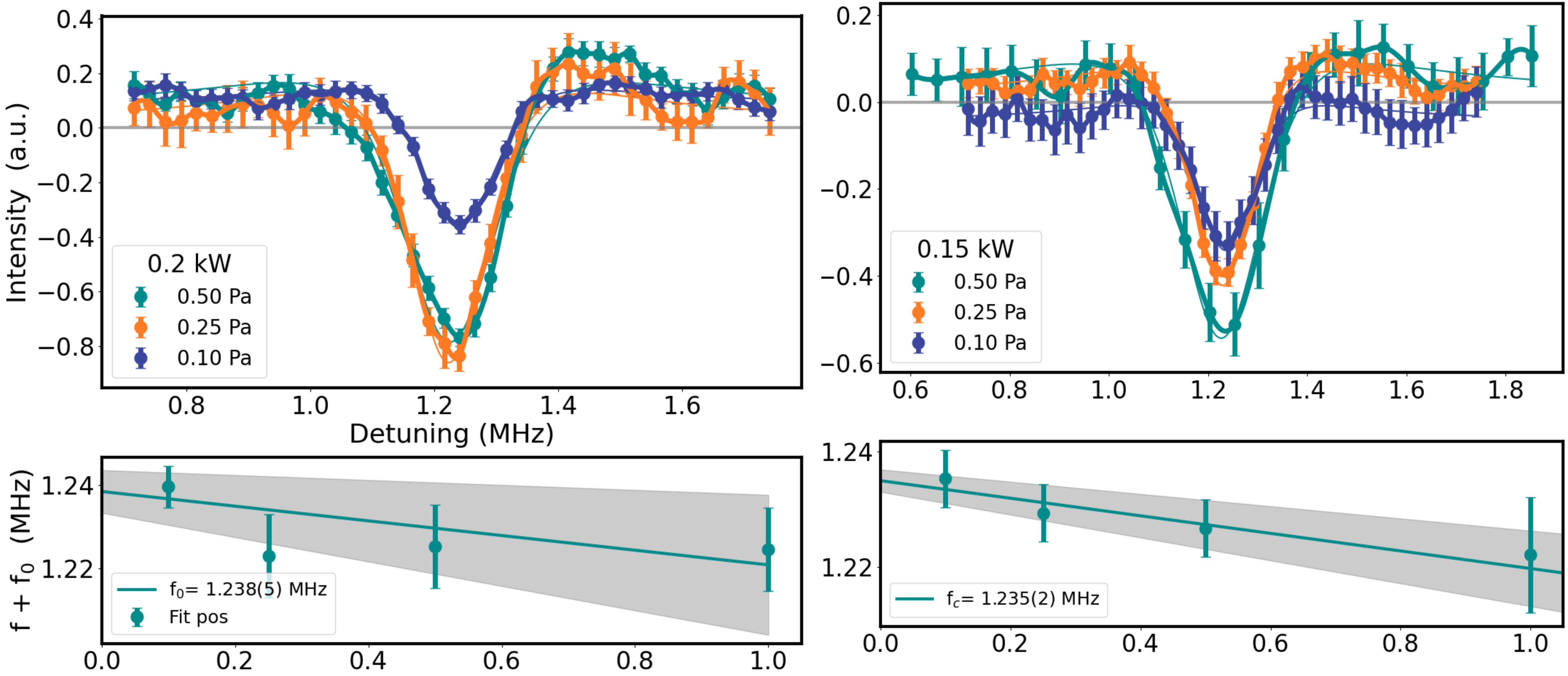
- Signal doesn’t drop as expected



Niceohms = random lineshape simulator



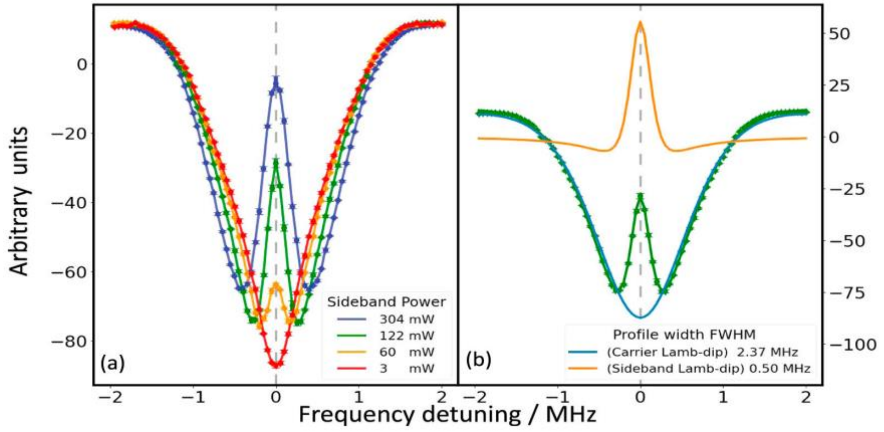
H_2 S(0) position extraction

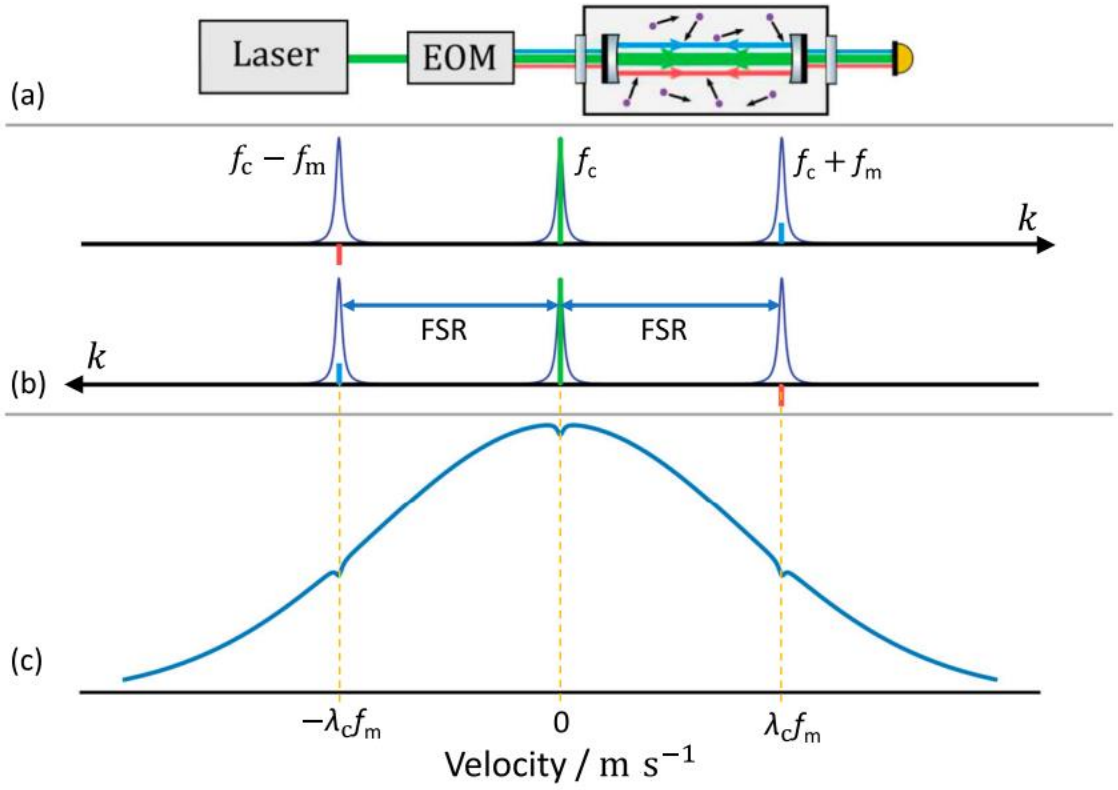


Carrier-Carrier saturation

- ‘standard’ scheme
- Orthogonal flying molecules
- Strong on-resonance standing wave
- Only signal in dispersion phase

- Actually composition of two signals
  - Carrier-Carrier
  - Doppler detuned sidebands
- Typically invisible as summation of opposite sign
- Visible for strong transitions such as water

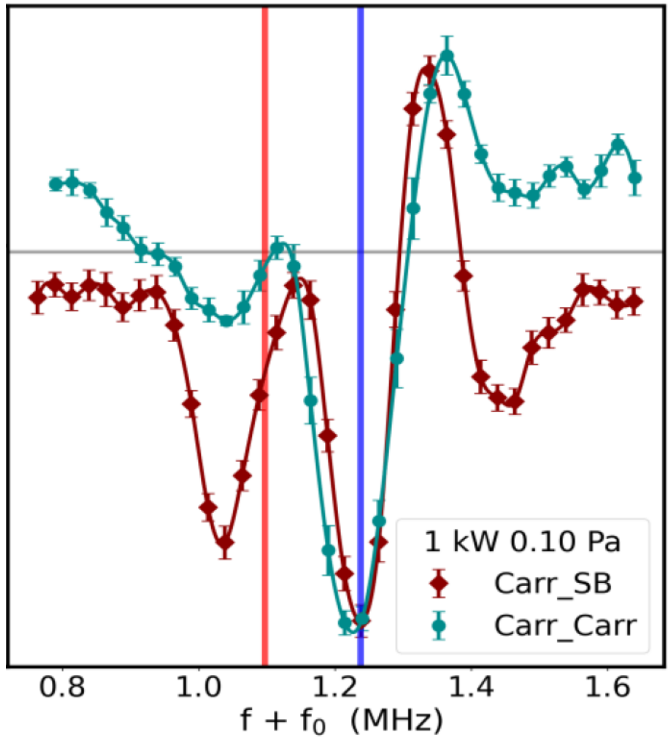




Doppler-detuned saturation spectroscopy (pump probe) Results on H2

- Saturating molecules flying along the beam
- Doppler shift equal to detuning
- On-resonance standing wave → travelling wave
  - Sideband only a few watt

Both dispersion and absorption channel



Recoil doublet recovered

- Asymmetric lineshapes
- Red recoil is suppressed
- Recoil shift must be applied