Hyperfine cross-over resonances producing Lamb-dips and Lamb-peaks in the saturation spectrum of HD

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Motivation

Hydrogen (and isotopes) as a benchmark molecules

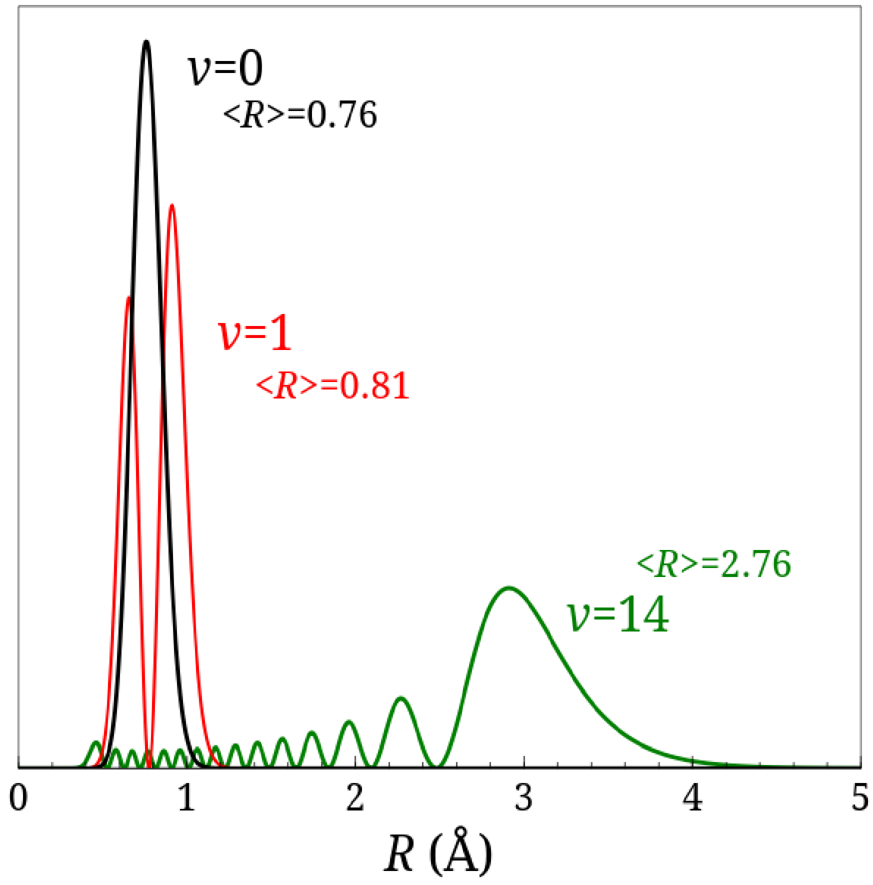
- Best calculated neutral molecule
  - ‘simple’4-body system
- Excellent test for molecular QED theory
- Extract fundamental constants from theory

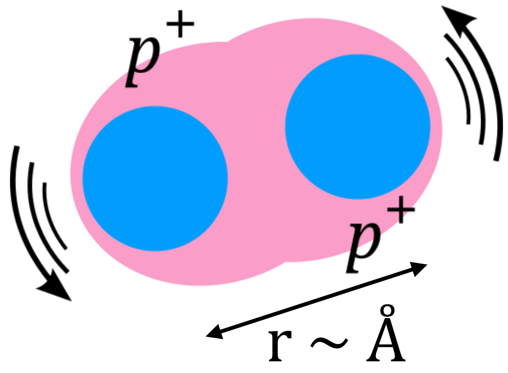
$$F\left(\alpha, R_{\infty}, r_{\mathrm{p}}, \mu_{\mathrm{p}}, \mu_{\mathrm{d}}\right) = \nu$$

$$\downarrow$$

$$F^{-1}\left(\alpha, R_{\infty}, r_{\mathrm{p}}, \mu_{\mathrm{d}}, \nu\right) = \mu_{\mathrm{p}}$$
$$F^{-1}\left(\alpha, R_{\infty}, \mu_{\mathrm{p}}, \mu_{\mathrm{d}}, \nu\right) = r_{\mathrm{p}}$$

Vibrations in hydrogen as probe for new physics





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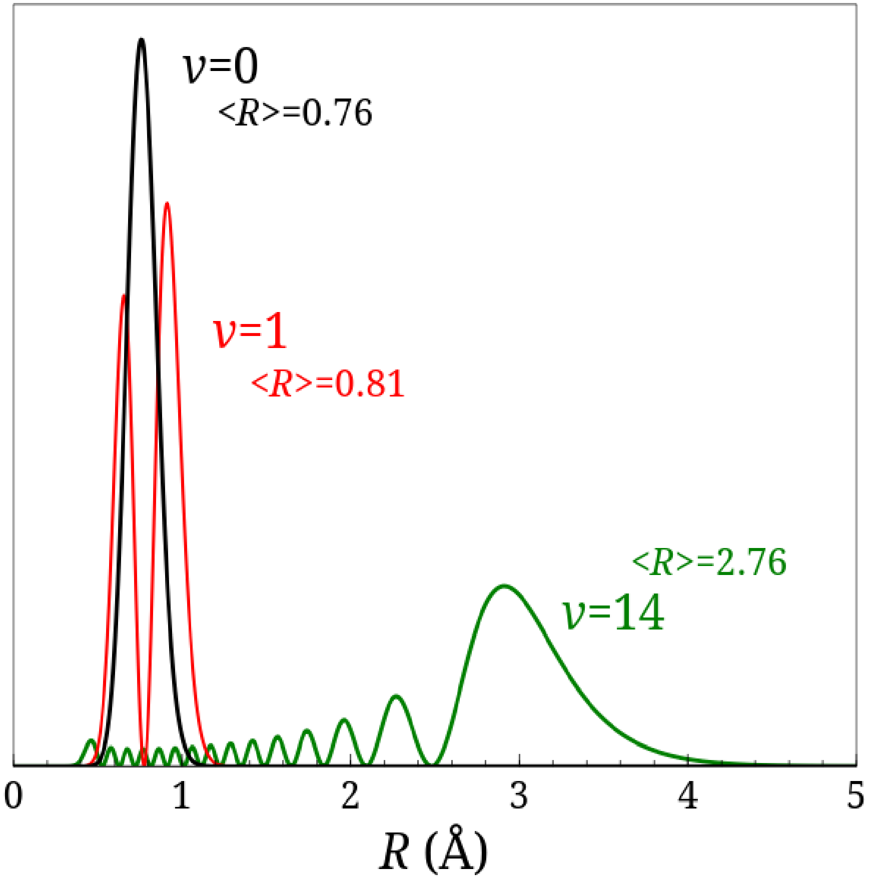
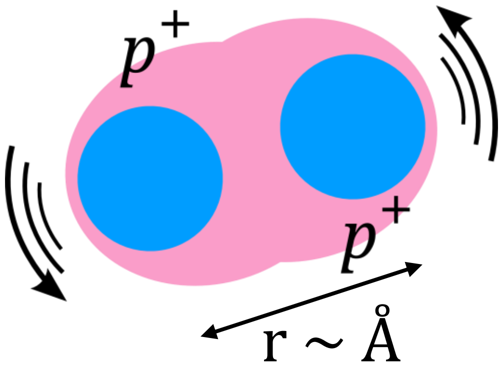
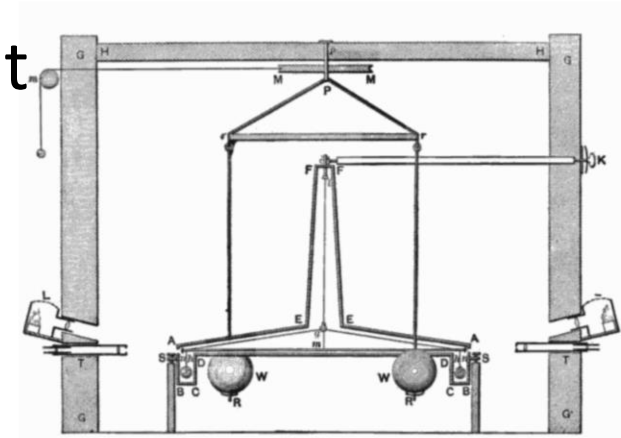
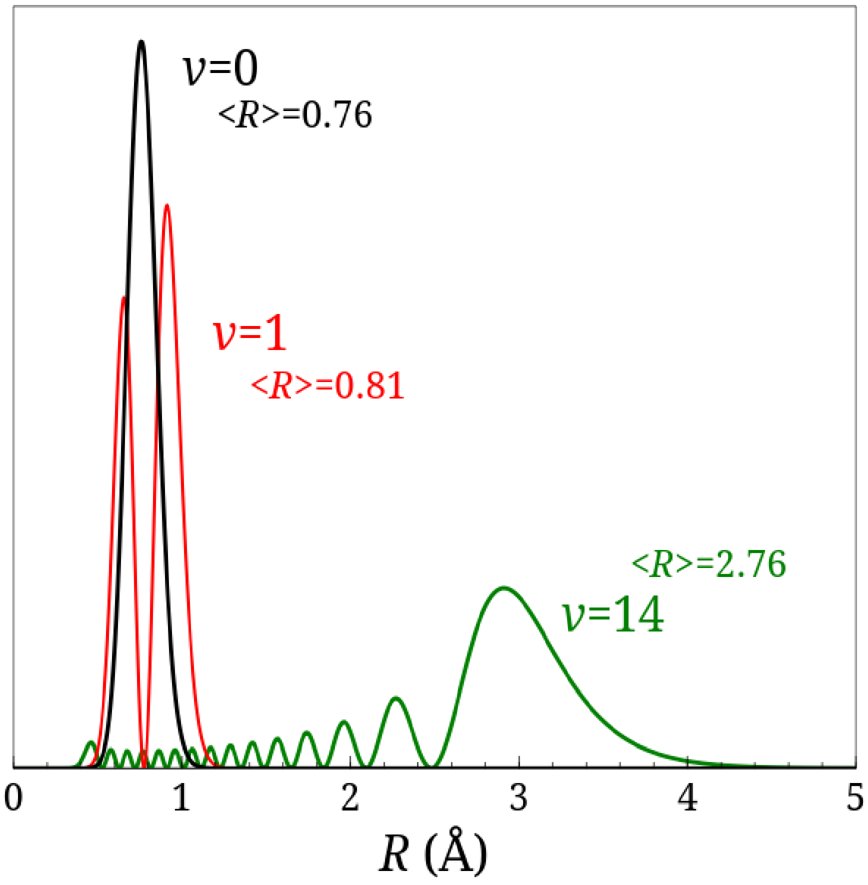


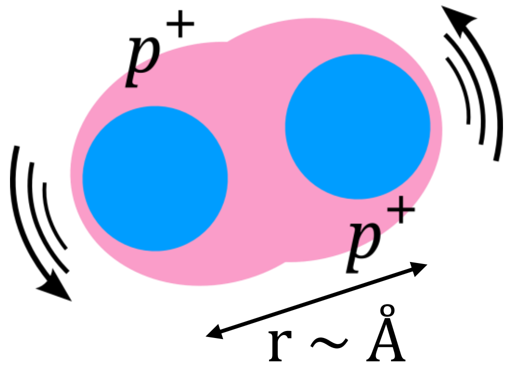


- Cavendish experiment



Vibrations in hydrogen as probe for new physics





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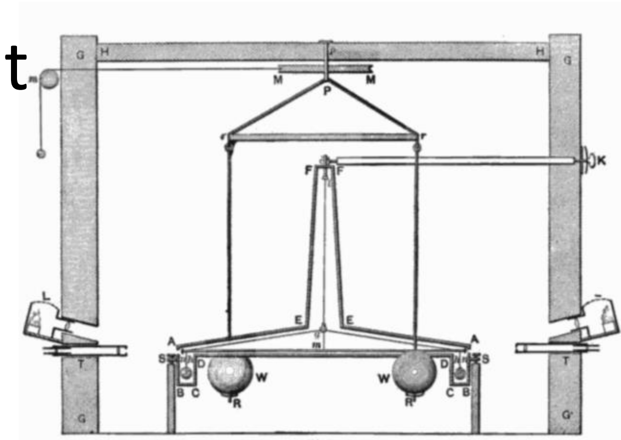

- Bounds on fifth forces

E.J.Salumbides, J. Komasa, K. Pachucki, *et al.*,
Phys. Rev. D **87**, 112008 (2013)

History of H_2 and HD

- $\mathrm{H_2}$ quadrupole transitions → very low intensity
- Absorption path length $\sim 50\ \mathrm{km}$ for 1 atm

- HD weak dipole transition due to non-adiabatic effects
- Absorption path length $\sim 1\ \mathrm{km}$ for 1 atm

History of H_2 and HD

- H_2 quadrupole transitions $\rightarrow$ very low intensity
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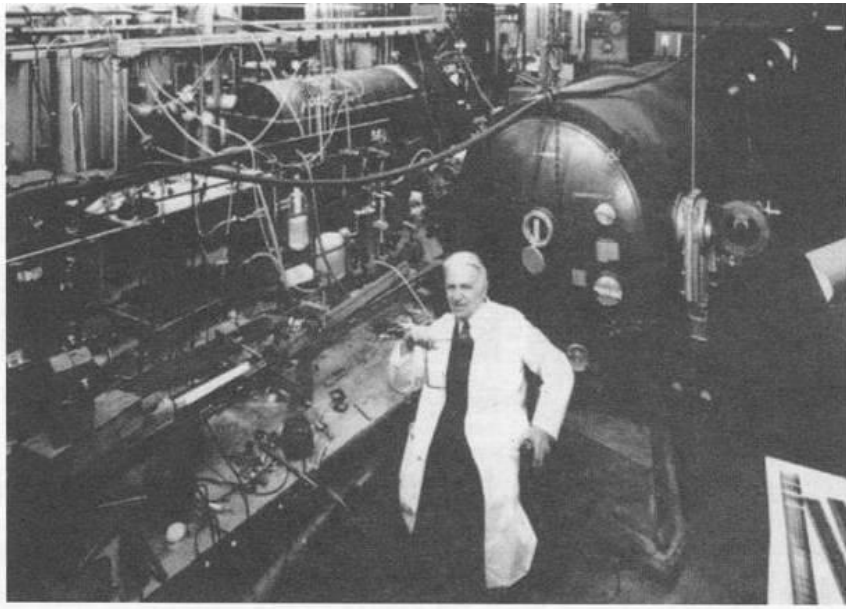
- HD weak dipole transition due to non-adiabatic effects
- Absorption path length ~ 1 km for 1 atm

- Herzberg in the 1950 : at 3 atm / path of 200m


Herzberg in his laboratory.

FORBIDDEN TRANSITIONS IN DIATOMIC MOLECULES

V. THE ROTATION–VIBRATION SPECTRUM OF THE HYDROGEN-DEUTERIDE (HD) MOLECULE[1]

R. A. DURIE[2] AND G. HERZBERG

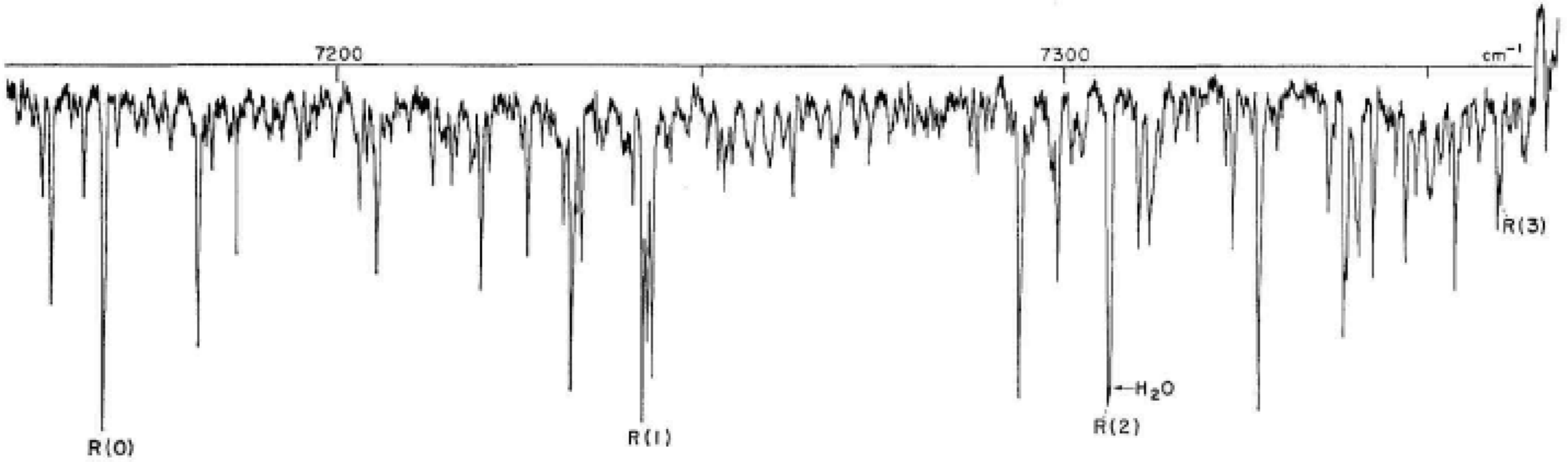
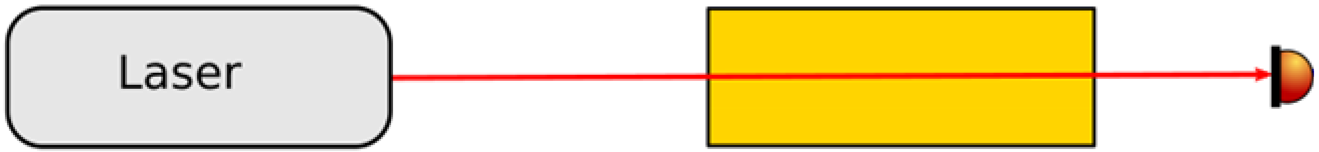


FIG. 2. *R* branch of 2–0 band of HD.
Absorbing path 200 m atm. Rapid scan in 2nd order of 7200 lines/inch grating. The unmarked absorption lines are due to H_2O.

Direct excitation of first overtone band in HD



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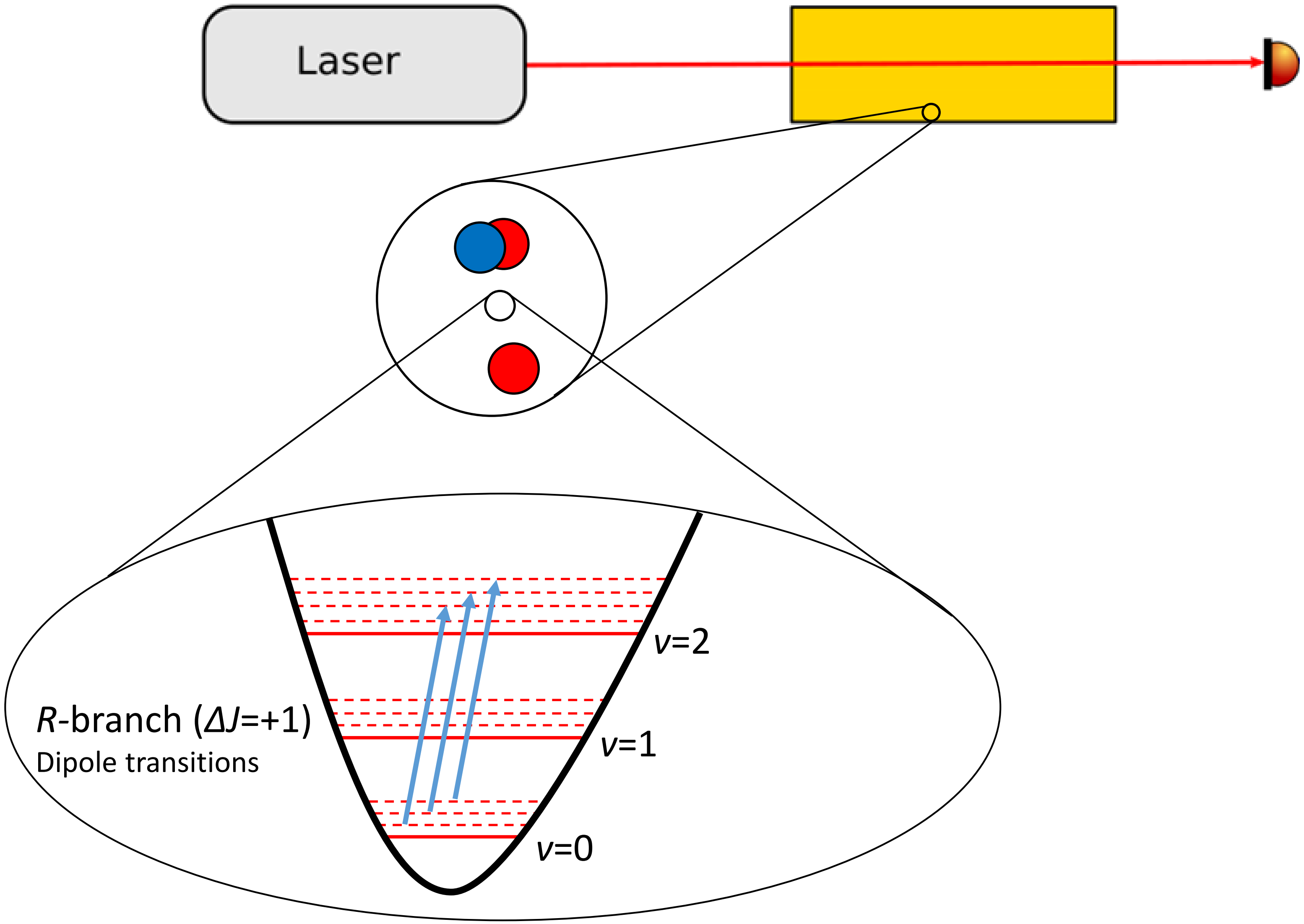

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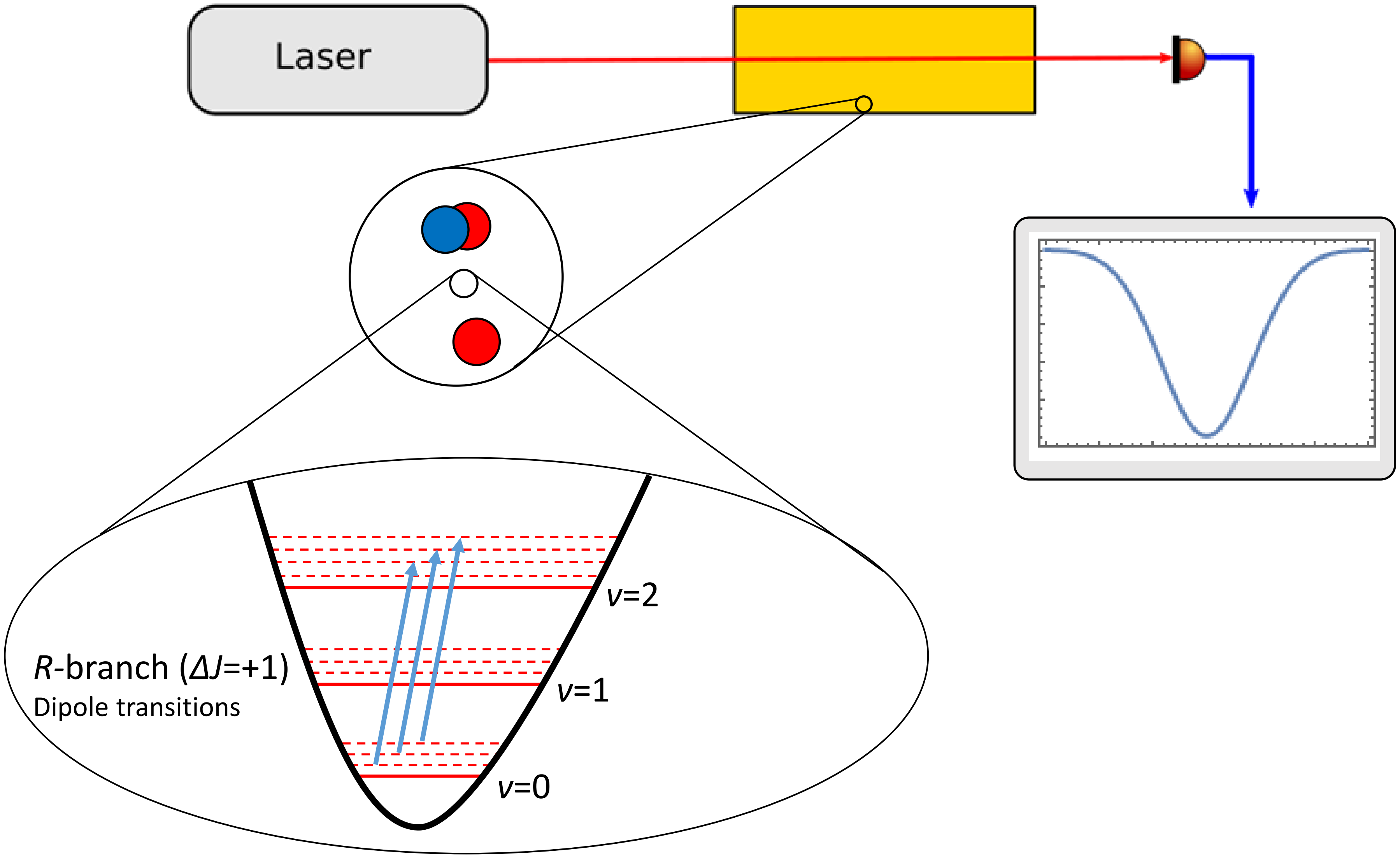

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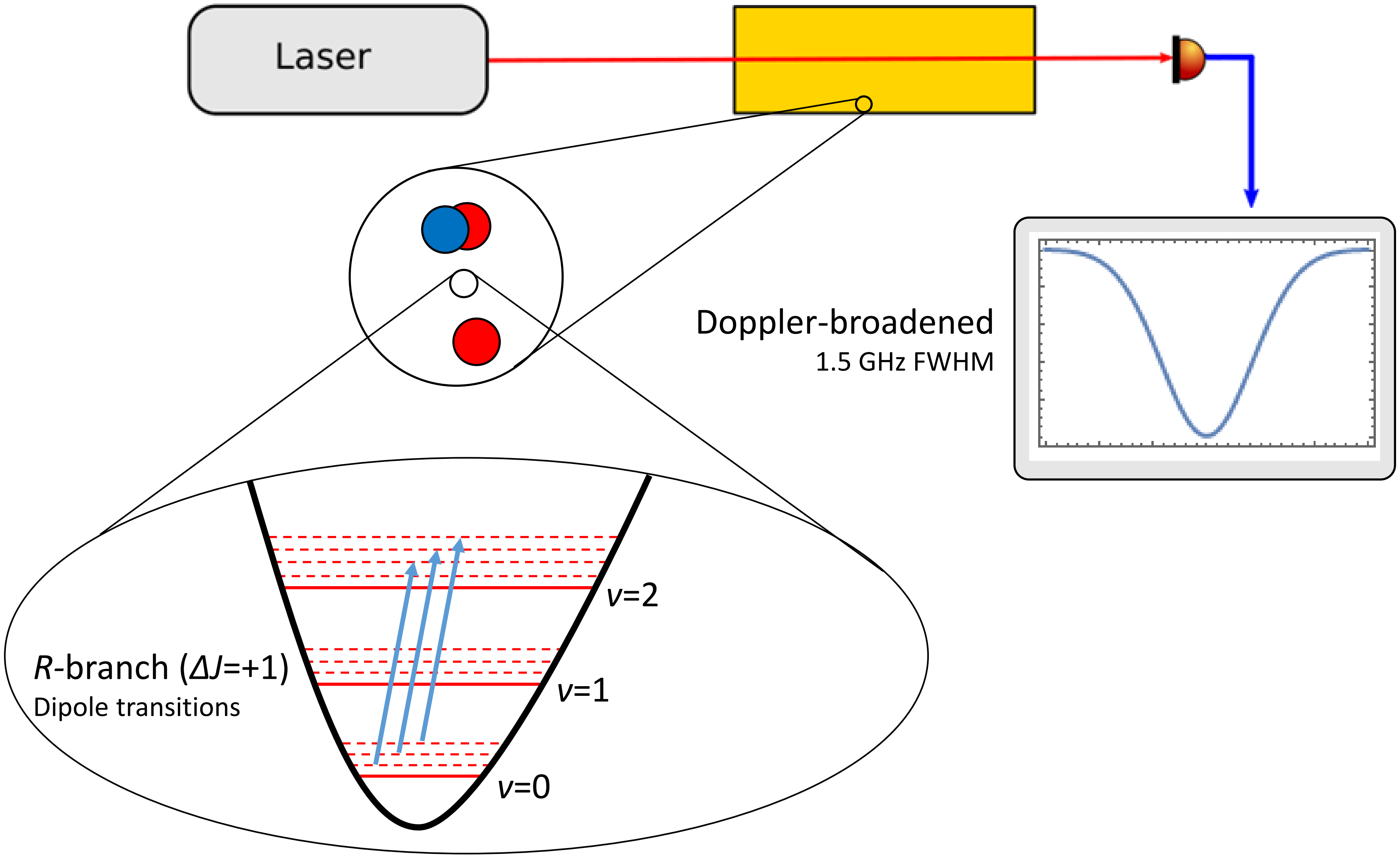

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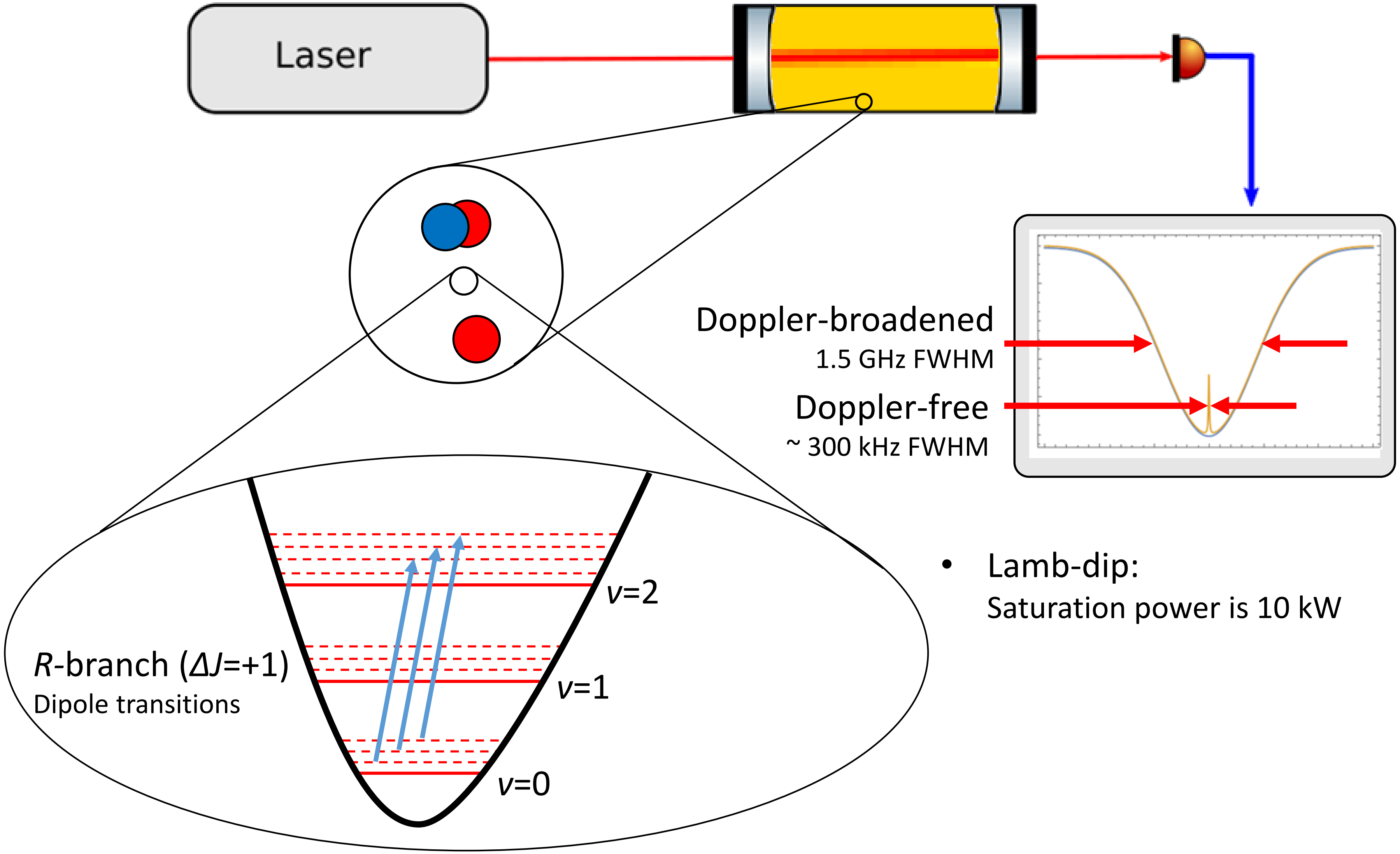
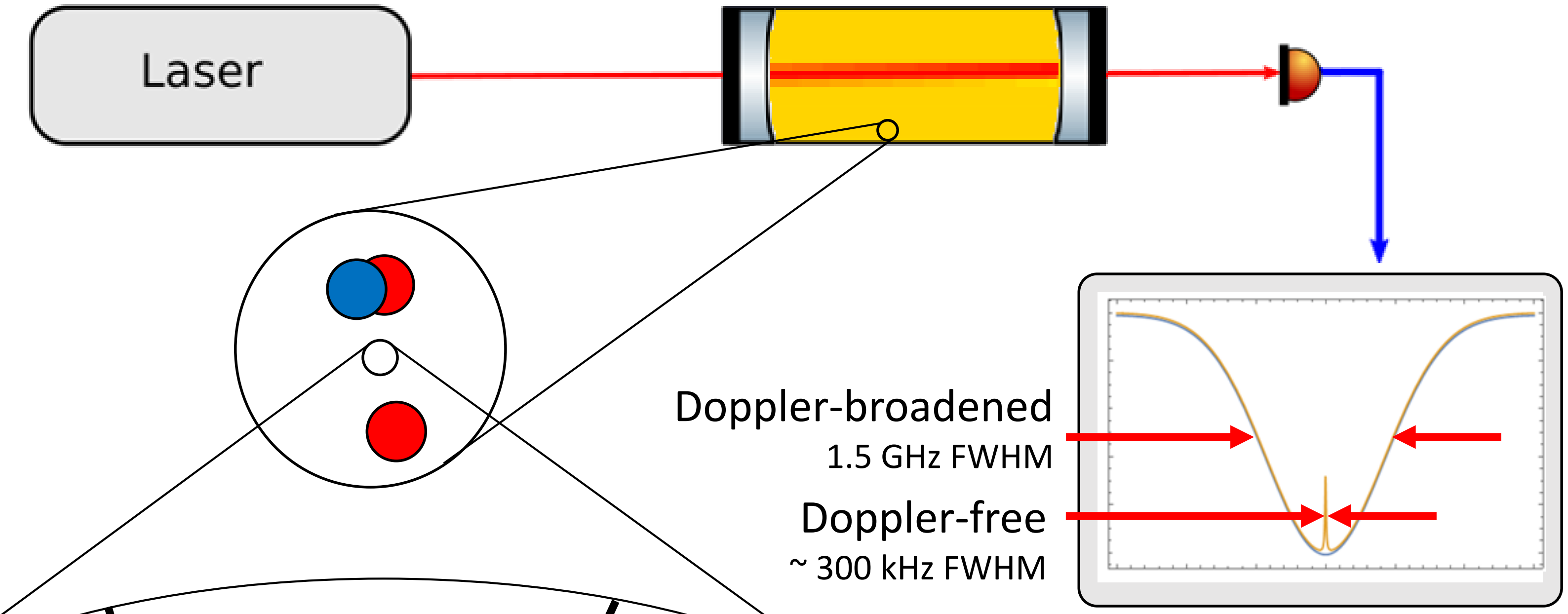
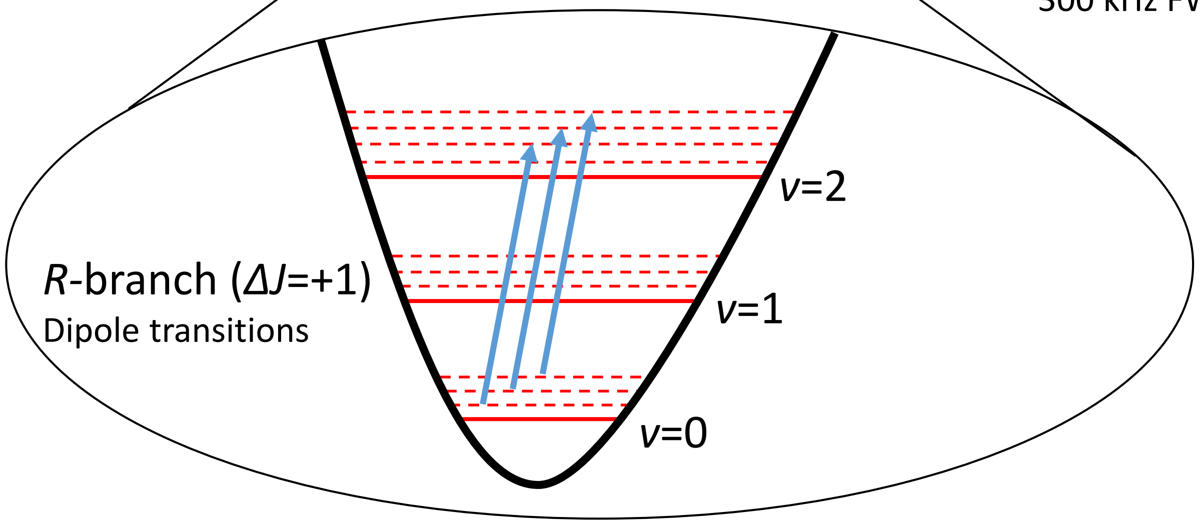


- Lamb-dip:
  Saturation power is 10 kW

Direct excitation of first overtone band in HD





- Last years result
  Cozijn *et al.*, *PRL* 120, 153002 (2018)
- Lamb-dip:
  Saturation power is 10 kW
  In our case : 100 W

NICE-OHMS

Noise-Immune Cavity-Enhanced Optical Heterodyne Molecular Spectroscopy

NICE-OHMS

Noise-Immune Cavity-Enhanced Optical Heterodyne Molecular Spectroscopy

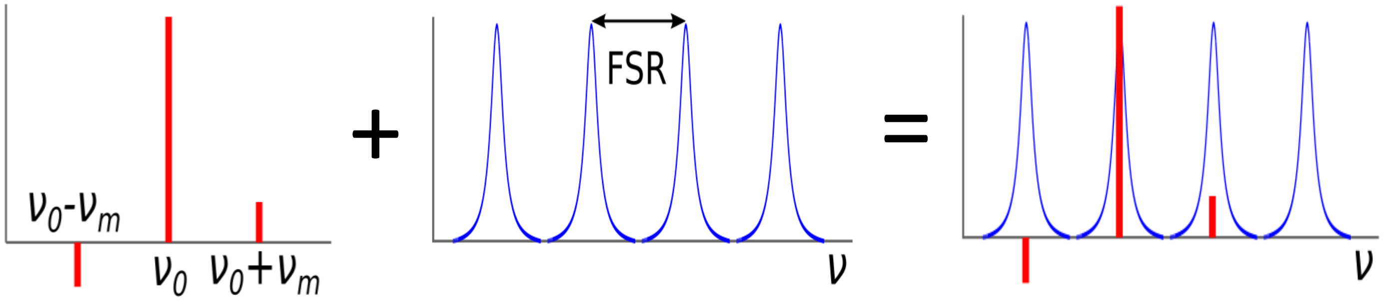
Cavity → POWER

Optical Heterodyne → SENSITIVITY

NICE-OHMS

Noise-Immune Cavity-Enhanced Optical Heterodyne Molecular Spectroscopy

- Frequency Modulation Spectroscopy (FMS) in a cavity



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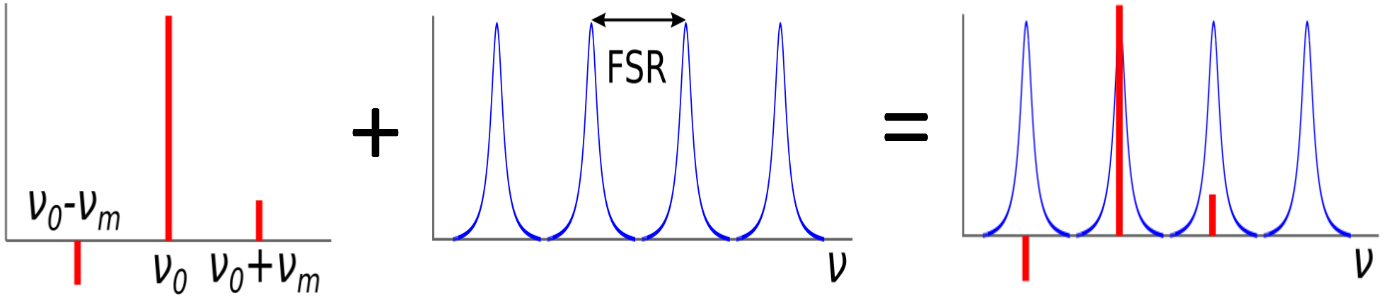
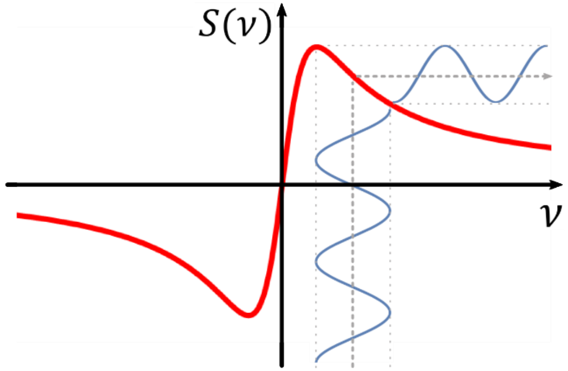


- Signals identical to FMS
  - Dispersion measurement



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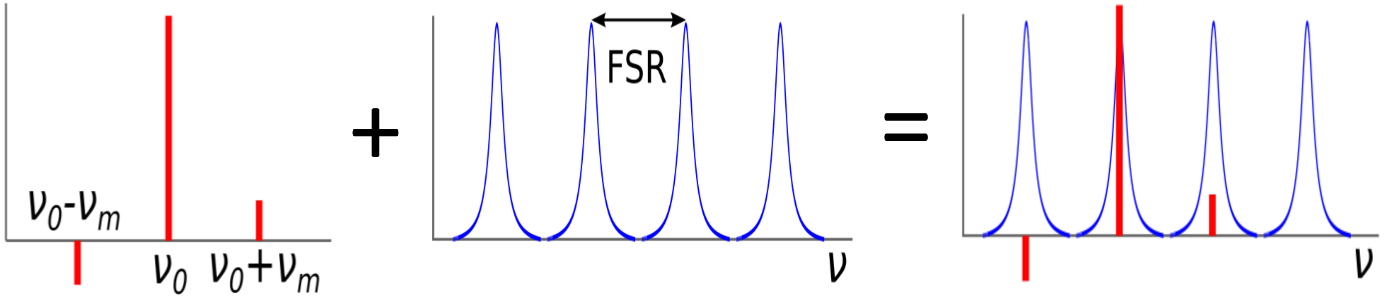
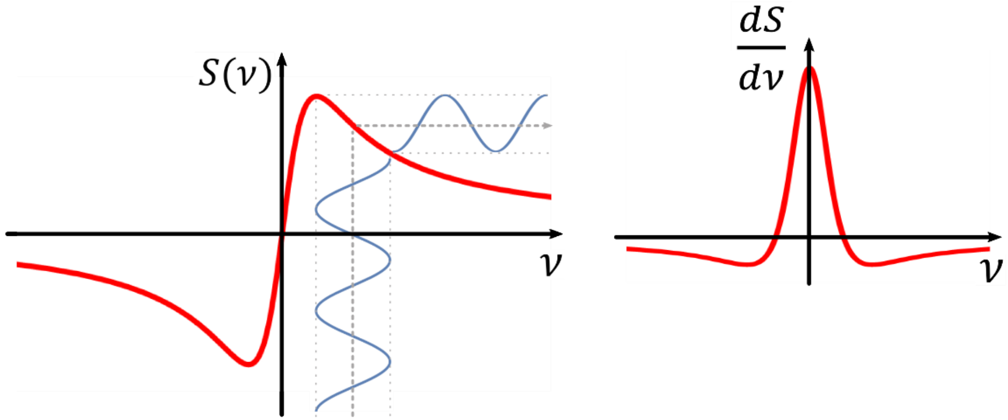
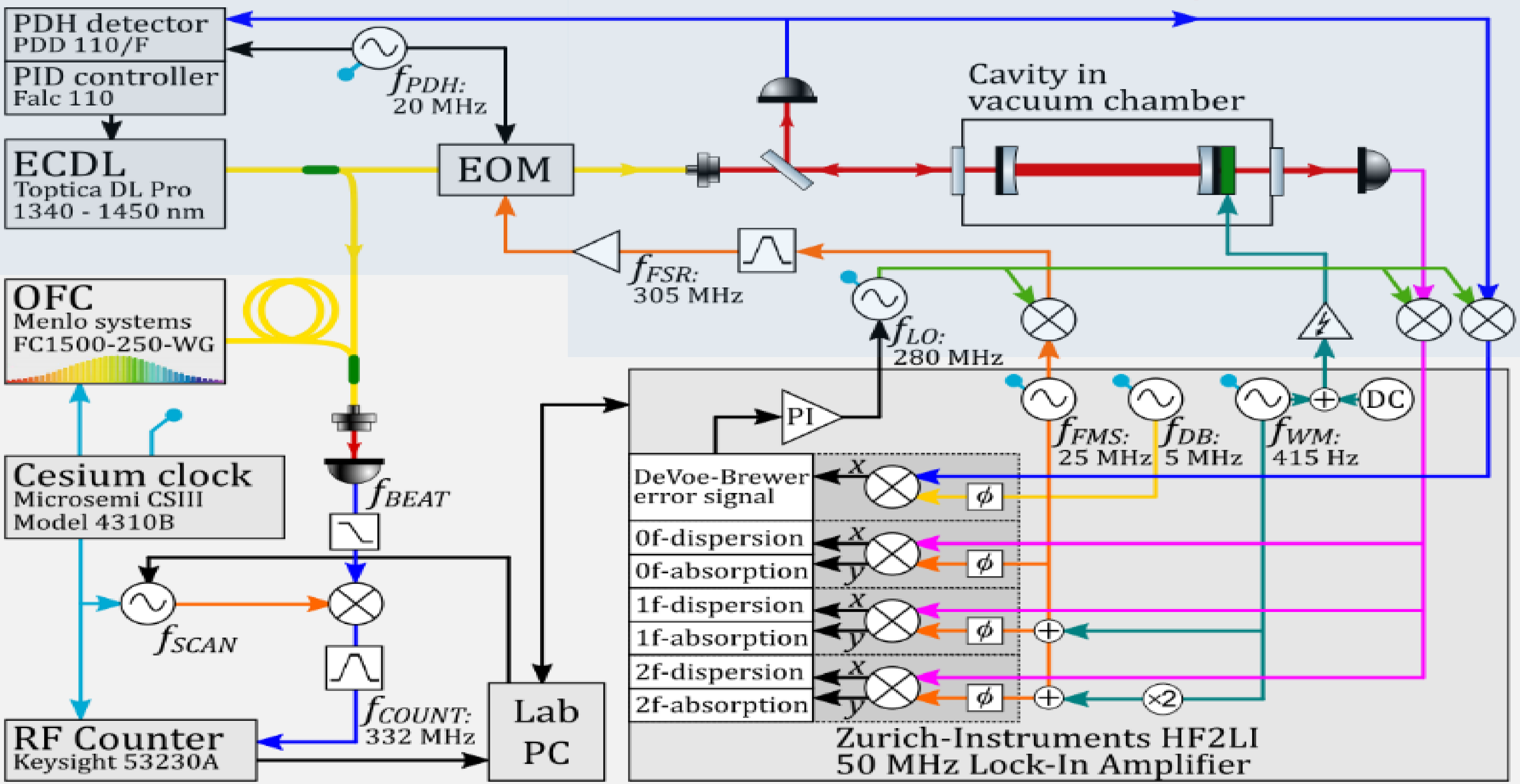


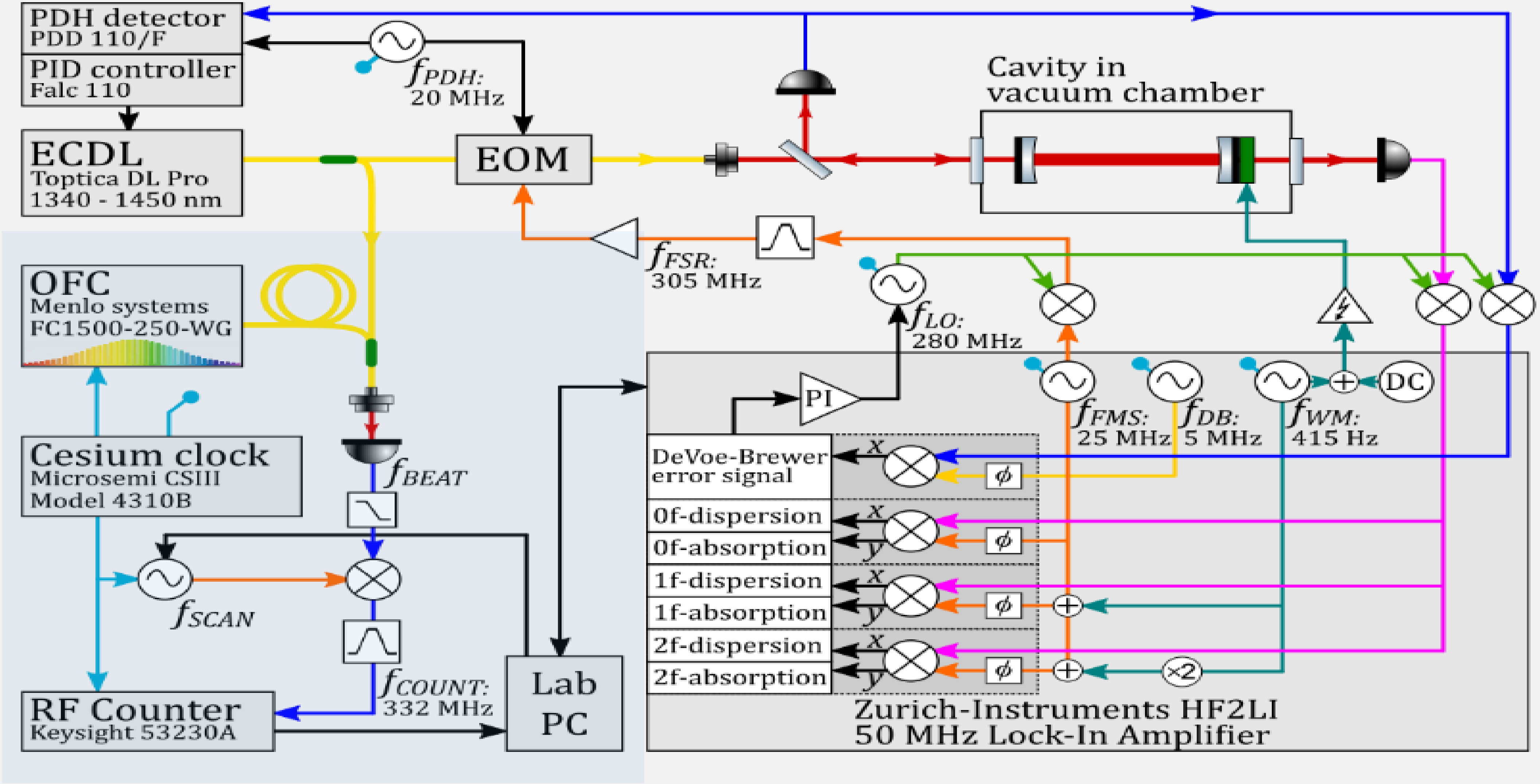
- Signals identical to FMS
  - Dispersion measurement
  - Additional wm-dither



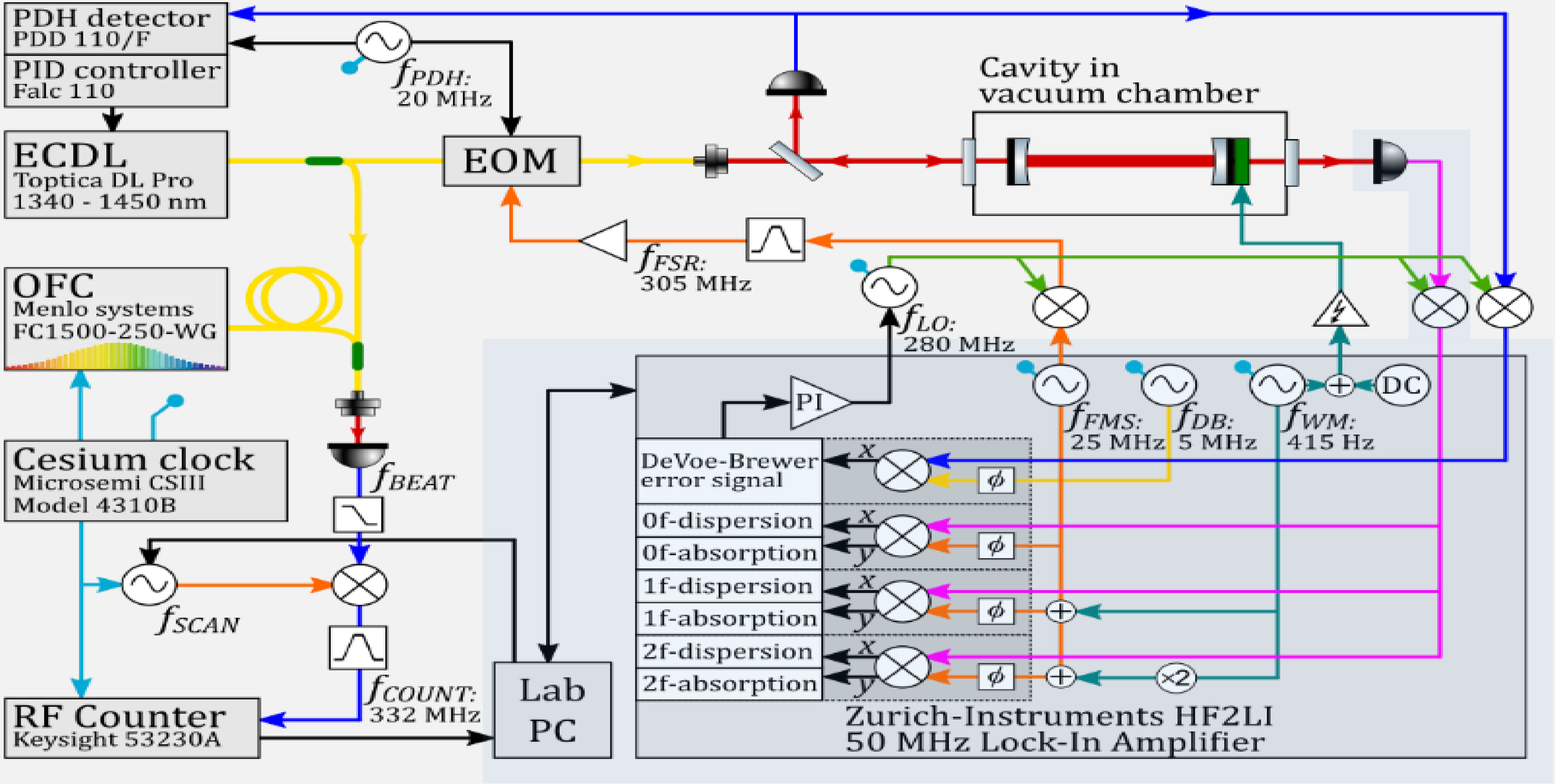
Setup

Finesse=150 000, $L_{eq} = 46$ km





PDH detector
PDD 110/F
PID controller
Falc 110
f_{PDH}: 20 MHz
ECDL
Toptica DL Pro
1340 - 1450 nm
EOM
Cavity in vacuum chamber
f_{FSR}: 305 MHz
f_{LO}: 280 MHz
OFC
Menlo systems
FC1500-250-WG
f_{BEAT}
Cesium clock
Microsemi CSIII
Model 4310B
f_{SCAN}
f_{COUNT}: 332 MHz
RF Counter
Keysight 53230A
Lab PC
PI
DC
f_{FMS}: 25 MHz
f_{DB}: 5 MHz
f_{WM}: 415 Hz
DeVoe-Brewer error signal
0f-dispersion
0f-absorption
1f-dispersion
1f-absorption
2f-dispersion
2f-absorption
x
y
ϕ
×2
Zurich-Instruments HF2LI
50 MHz Lock-In Amplifier



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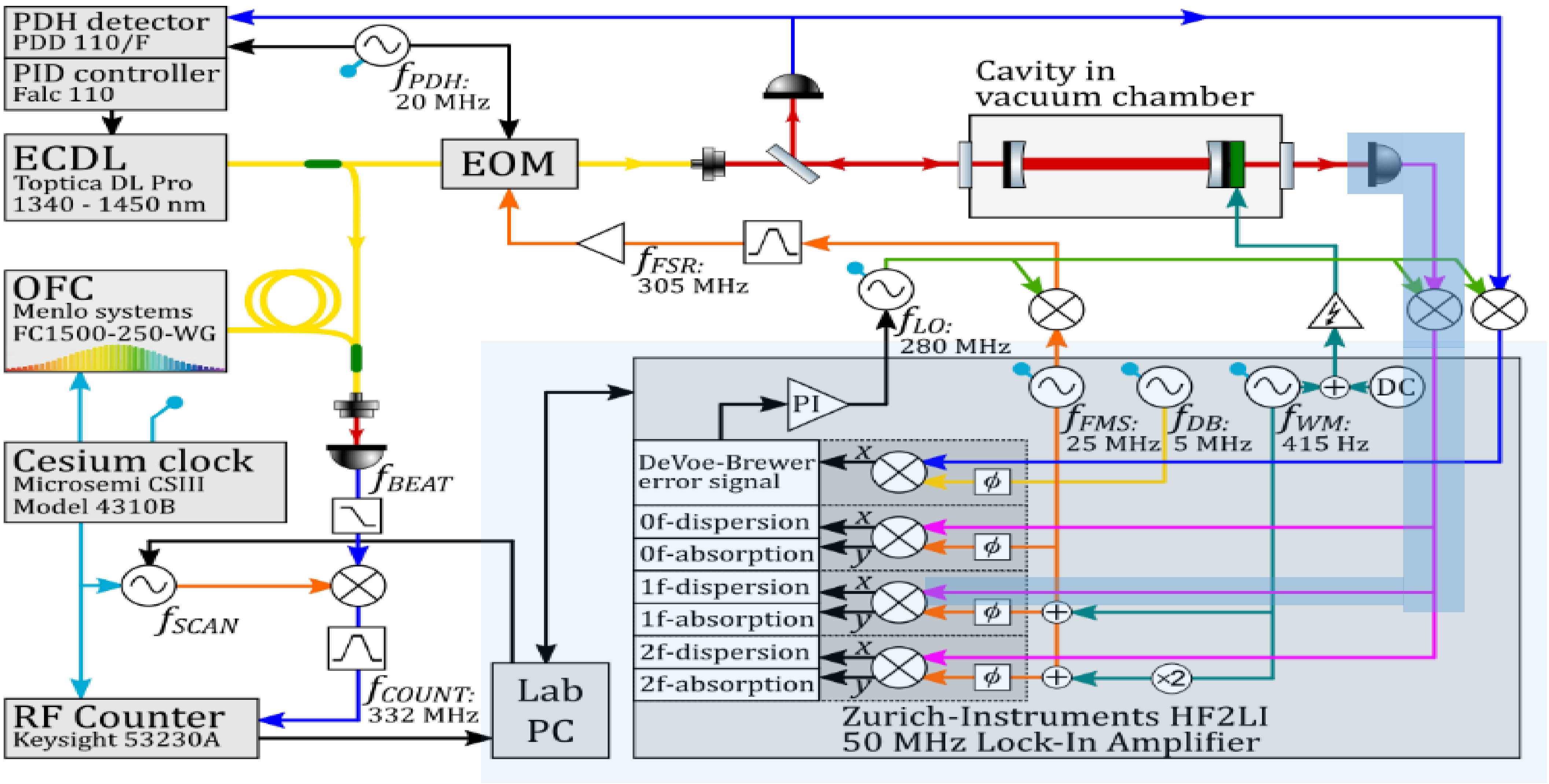

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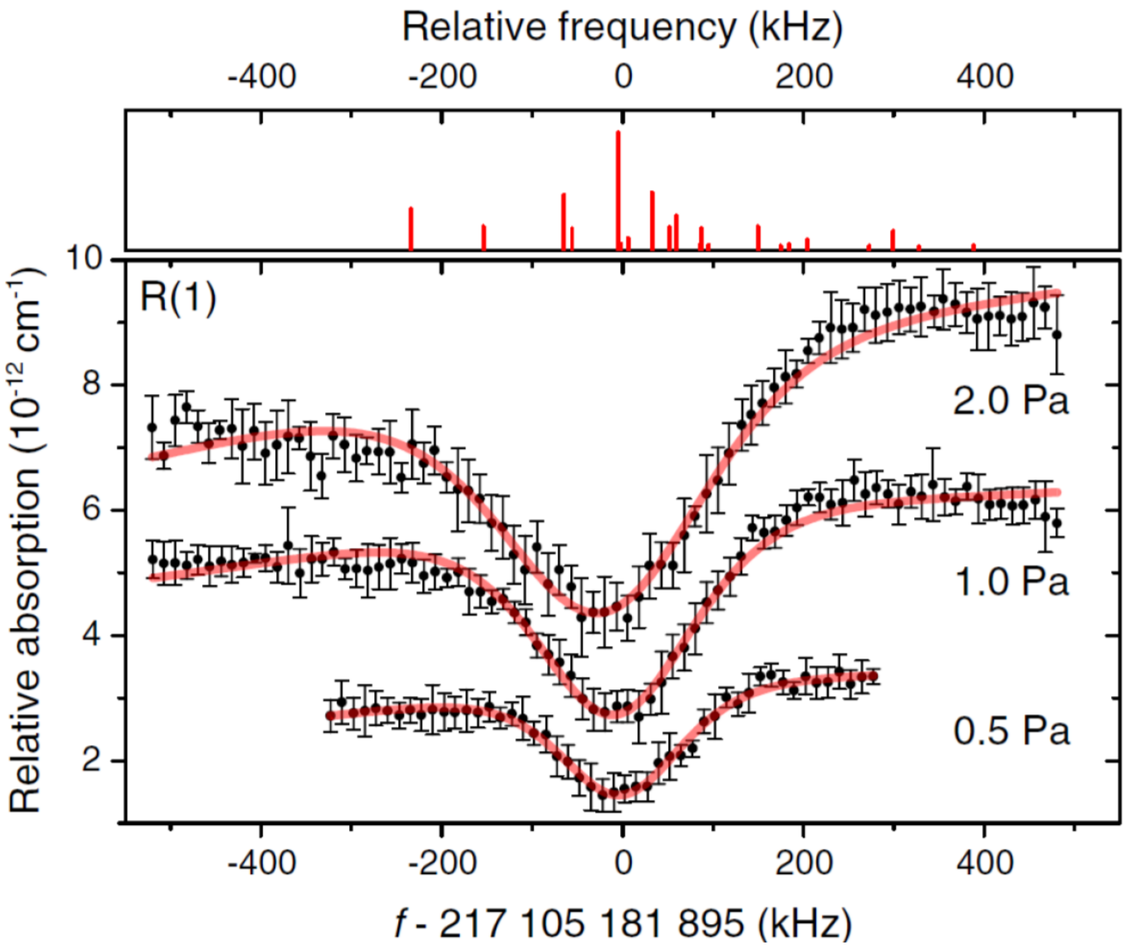
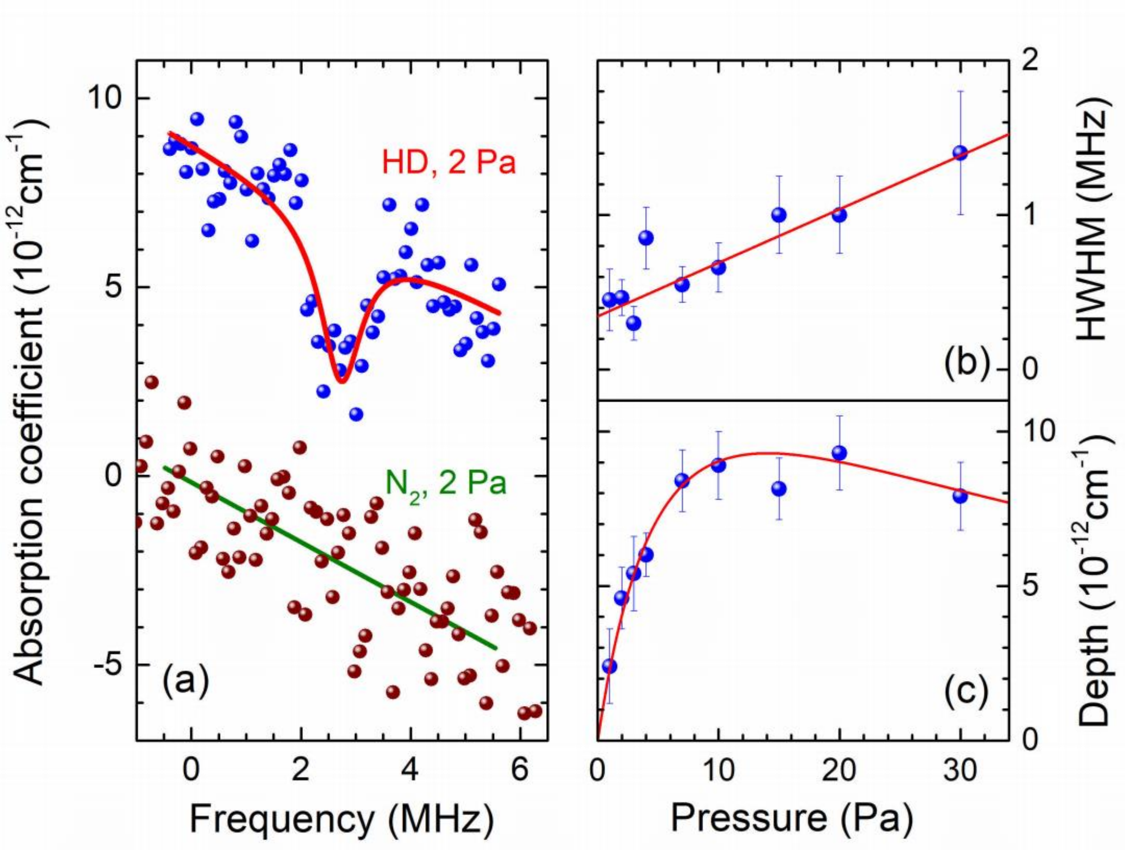
Experiments (2018) Amsterdam and HEFEI



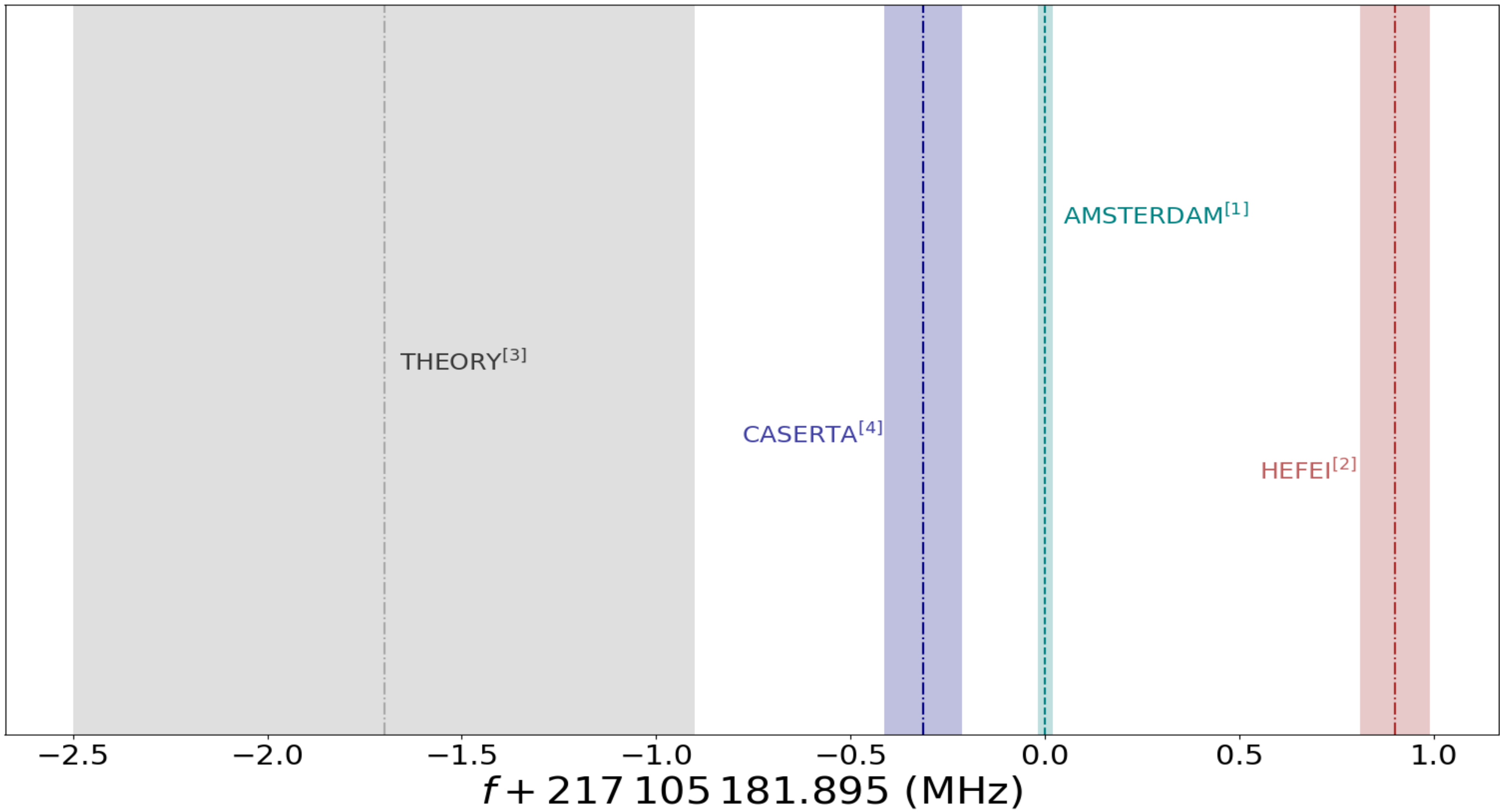


$f_0 = 217\ 105\ 182.79$ MHz

Cozijn et al., *PRL* **120**, 153002 (2018)
Amsterdam

Tao et al., *PRL* **120**, 153001 (2018)
HEFEI

HD R(1) discrepancy



[1] Cozijn et al., *PRL* **120**, 153002 (2018)

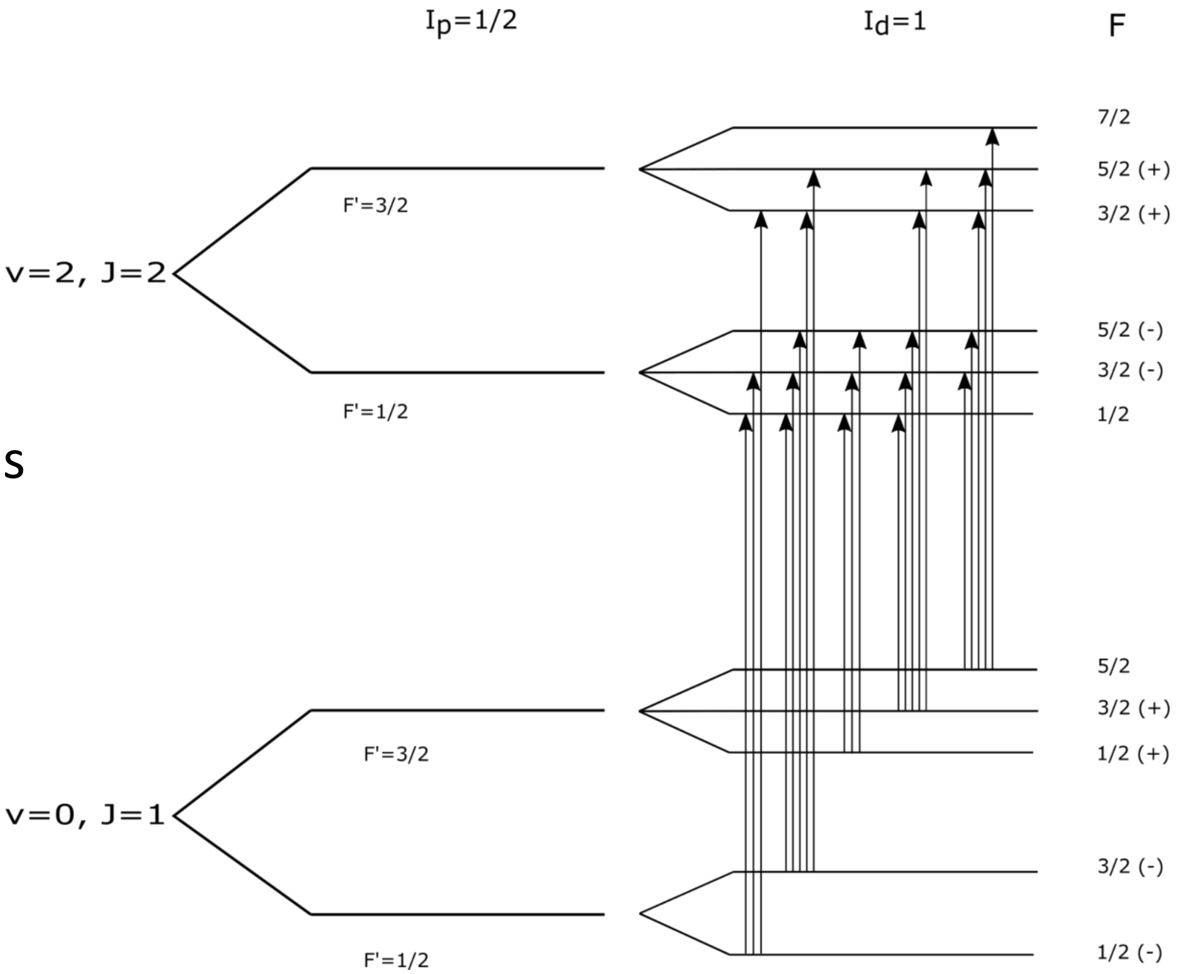
[2]Tao et al., *PRL* **120**, 153001 (2018)

[3]Czachorowski et al., PRA **98**, 052506 (2018)

[4] Fasci et al., PRA 98, 022516 (2018)

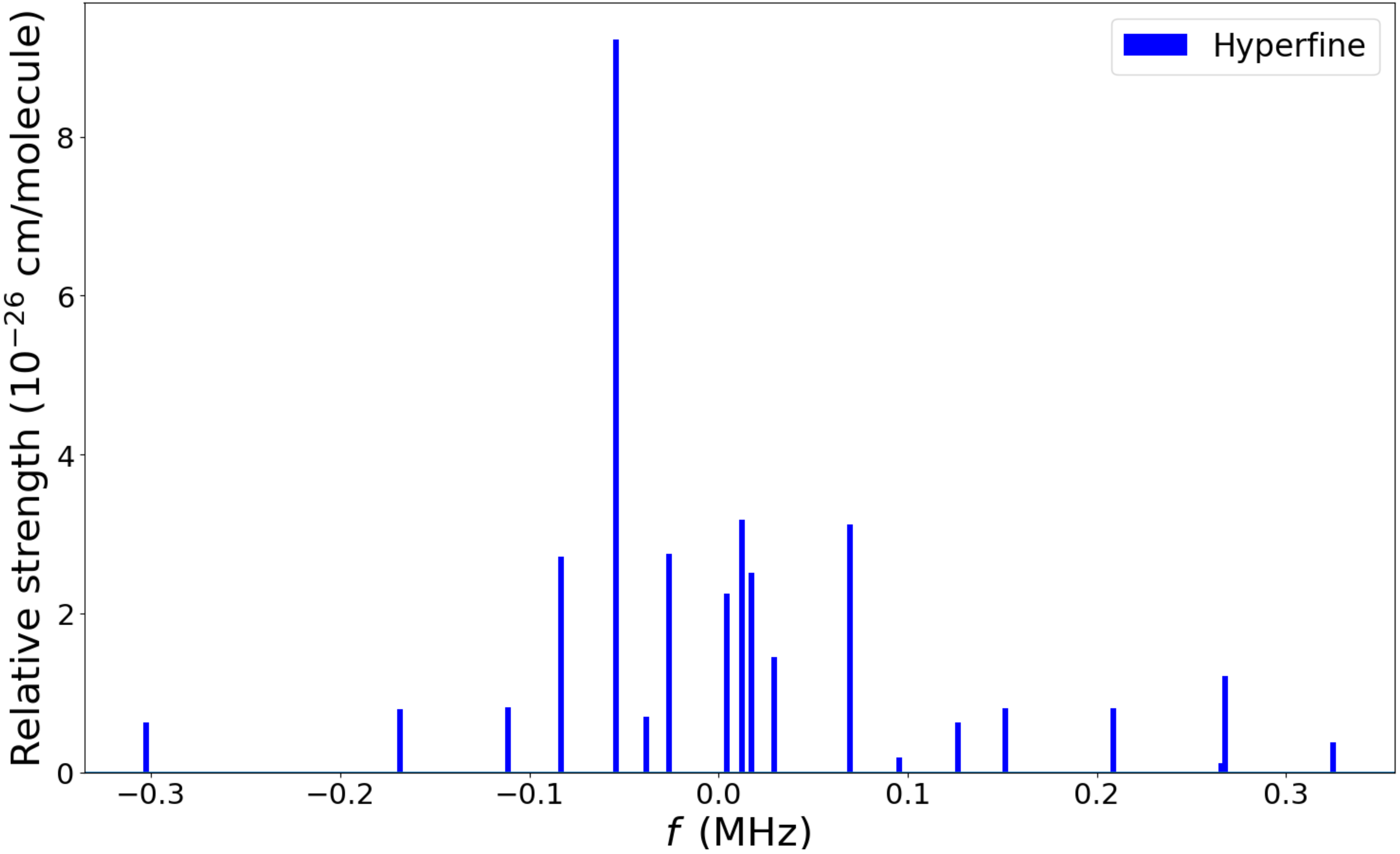
HD hyperfine levels

- Interaction nuclear spins(d-p): $I_d = 1, I_p = {}^1\!/_2$ and rotational quantum number J
  - v=0, J=1 → 5 levels
  - v=2, J=2 → 6 levels
- Allowed transitions $\Delta F = 0, {}^{+}/_{-}1$ → 21 transitions



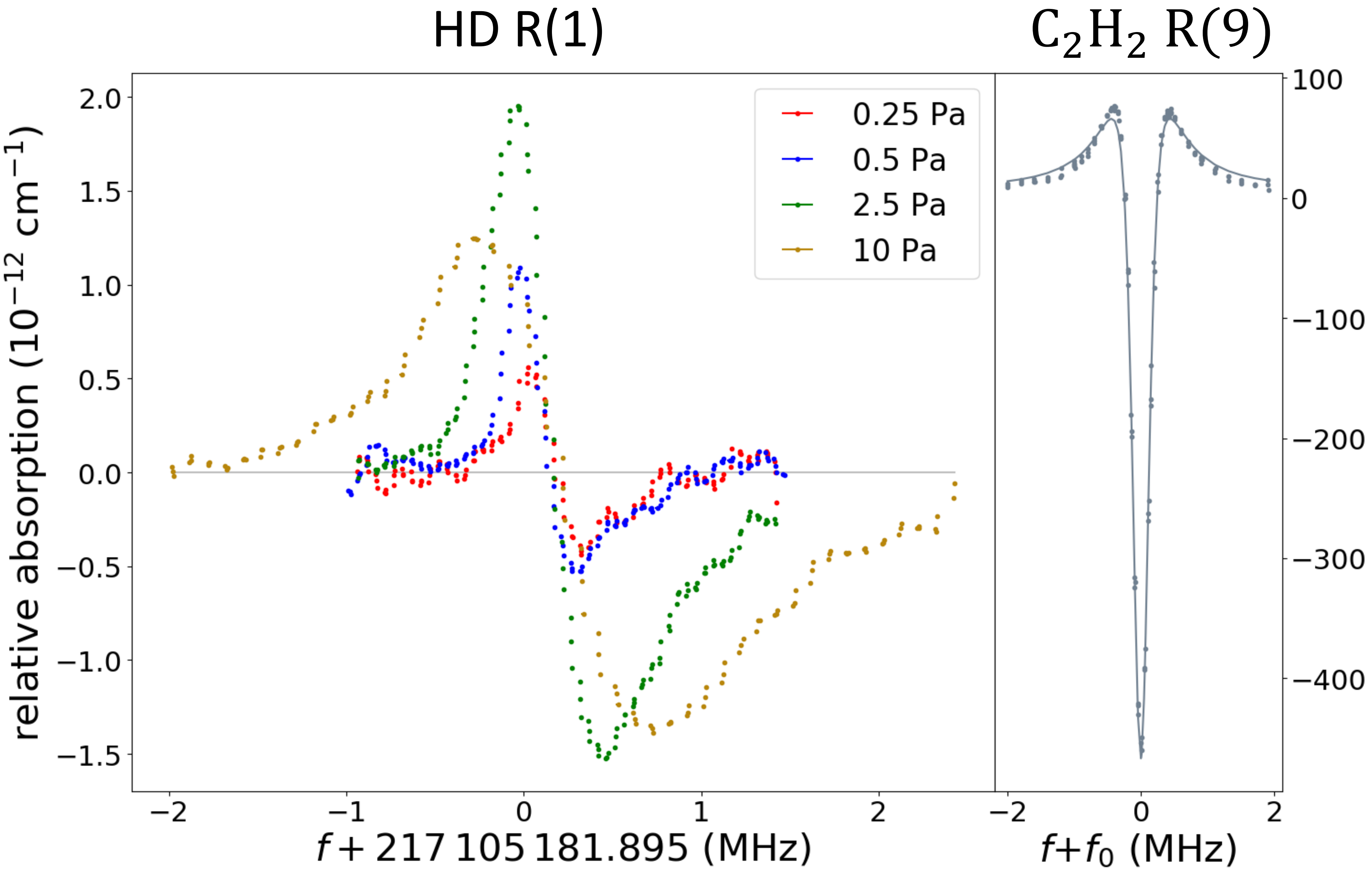
N. F. Ramsey and H. R. Lewis, Phys. Rev. **108**, 1246 (1957)

HD R(1) Hyperfine structure

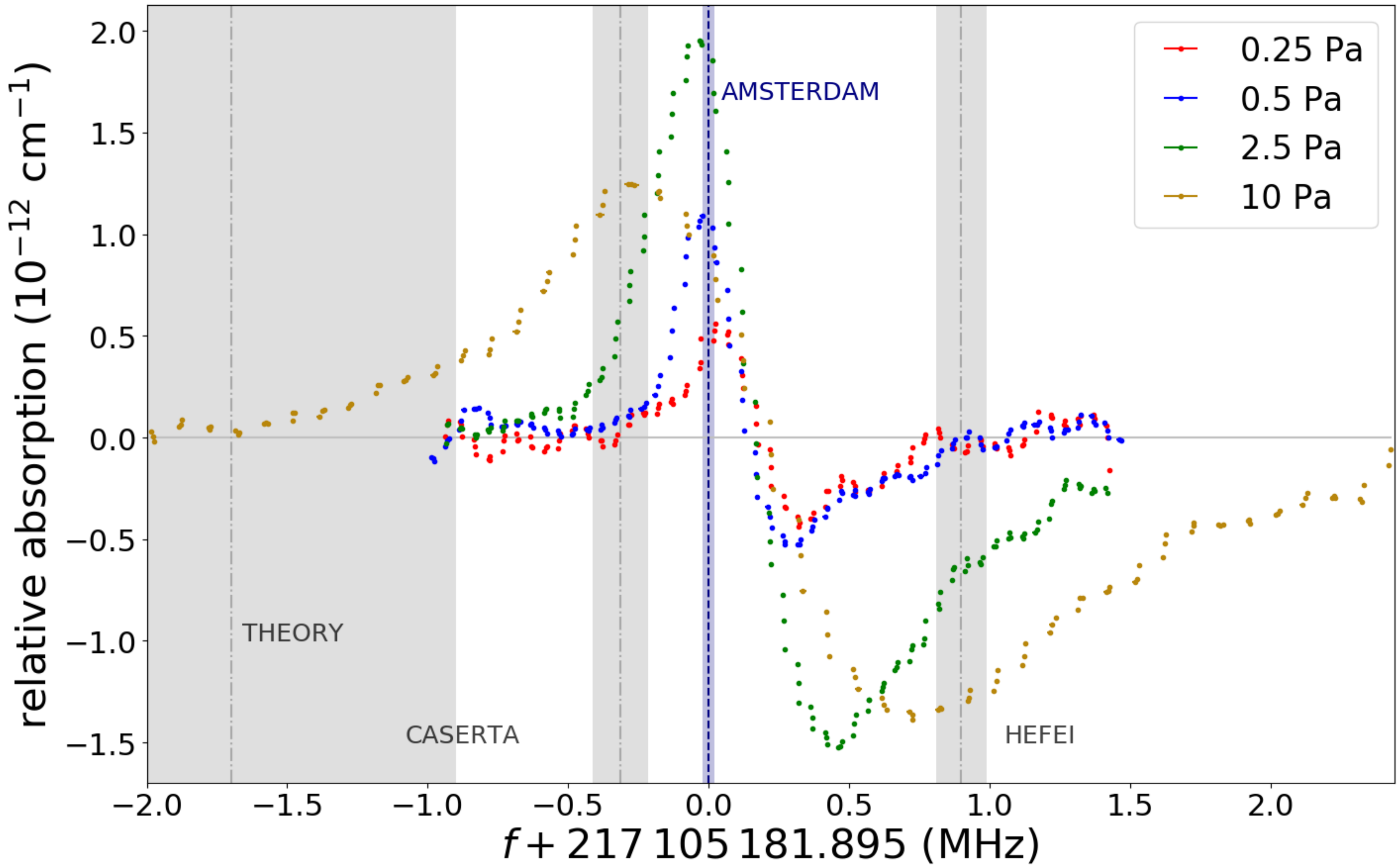


P. Dupré and J. Gauss

Results



HD R(1)
C_2H_2 R(9)
relative absorption (10^{-12} cm^{-1})
0.25 Pa
0.5 Pa
2.5 Pa
10 Pa
2.0
1.5
1.0
0.5
0.0
−0.5
−1.0
−1.5
−2
−1
0
1
2
f + 217 105 181.895 (MHz)
100
0
−100
−200
−300
−400
f+f_0 (MHz)

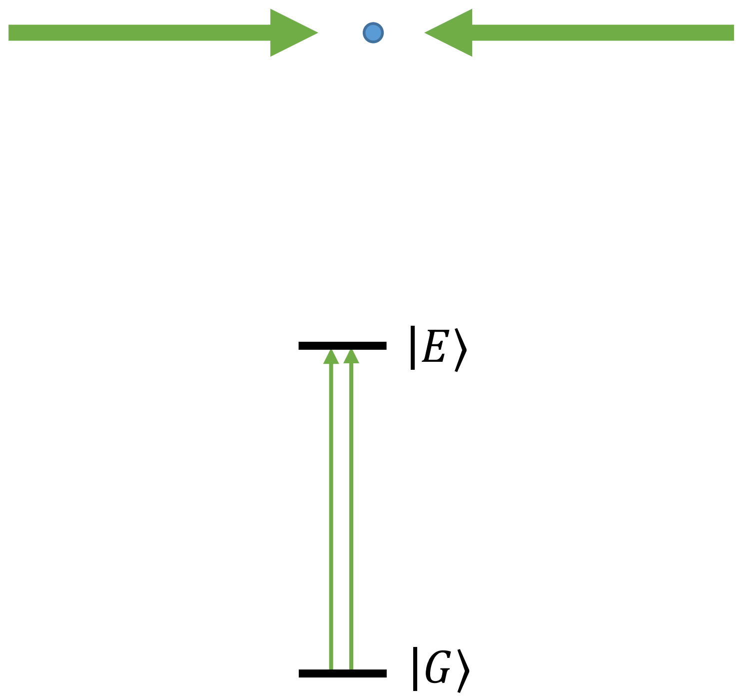


0.25 Pa
0.5 Pa
2.5 Pa
10 Pa
AMSTERDAM
THEORY
CASERTA
HEFEI
relative absorption (10^{-12} cm^{-1})
$f + 217\,105\,181.895$ (MHz)
2.0
1.5
1.0
0.5
0.0
−0.5
−1.0
−1.5
−2.0
−1.5
−1.0
−0.5
0.0
0.5
1.0
1.5
2.0

Interpretation

Crossovers in saturation spectroscopy

Doppler free Lamb-dip



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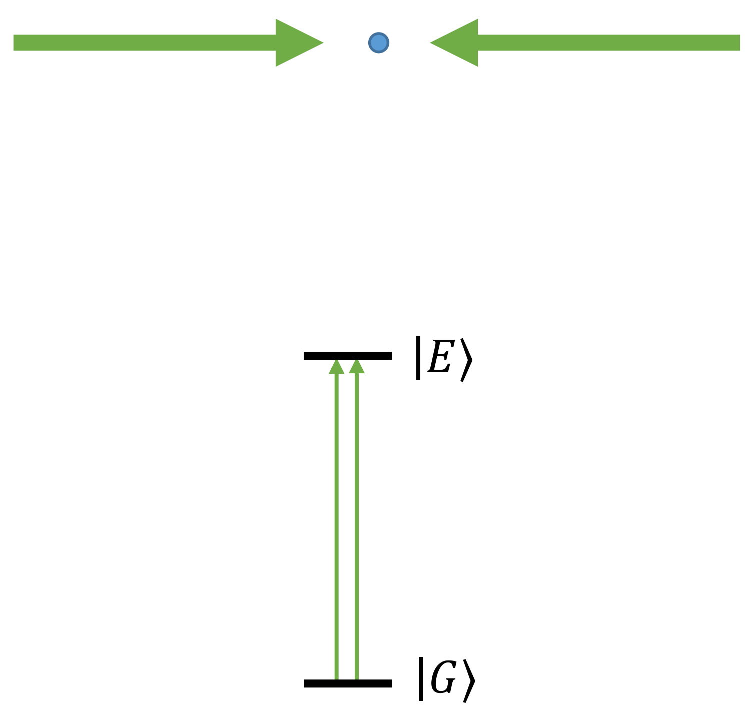
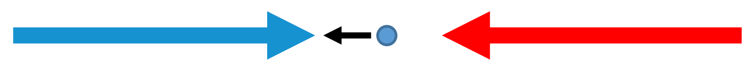


Crossover resonances



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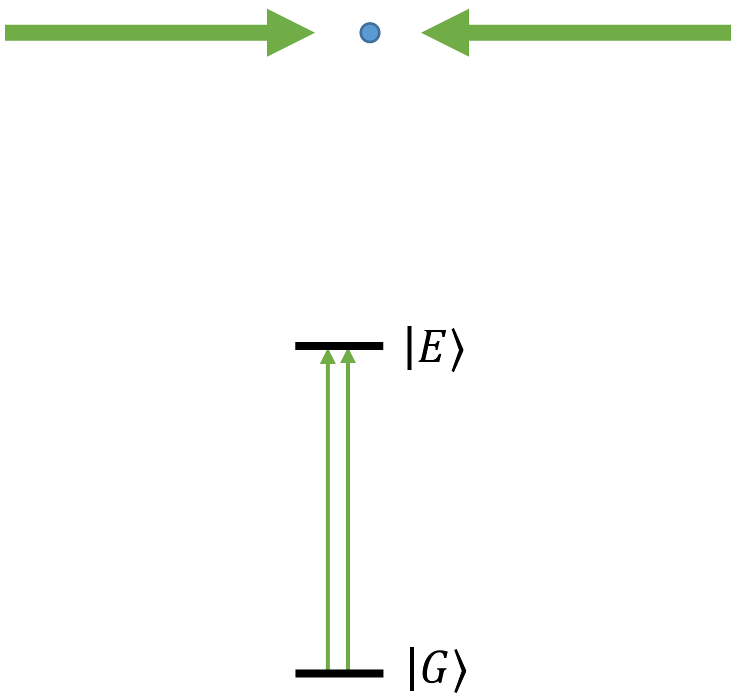

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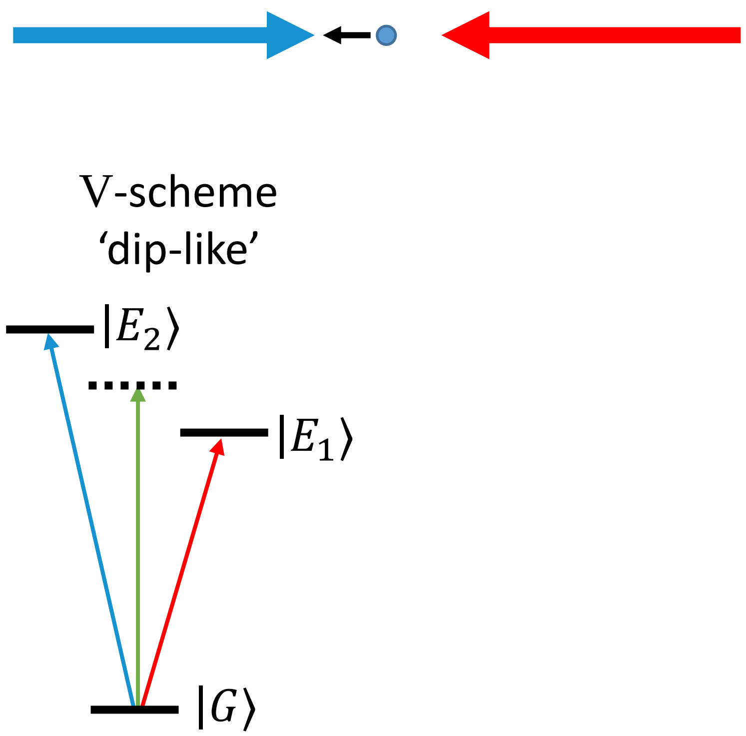

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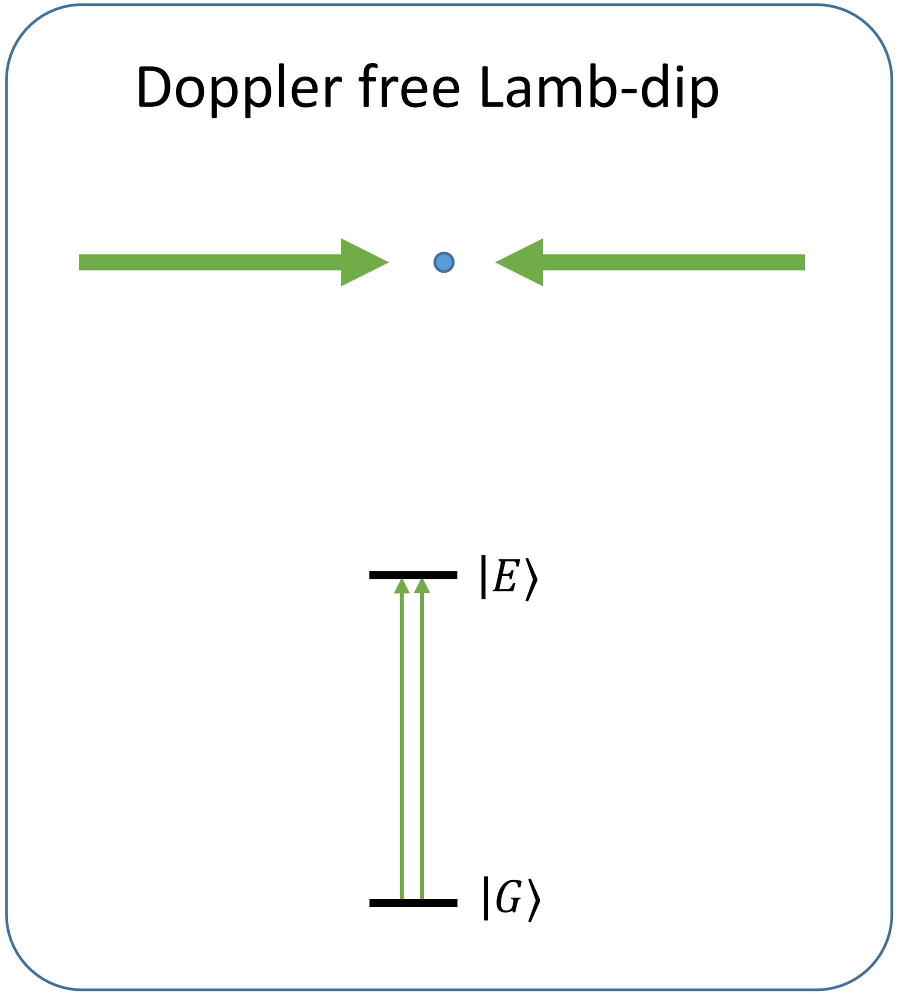
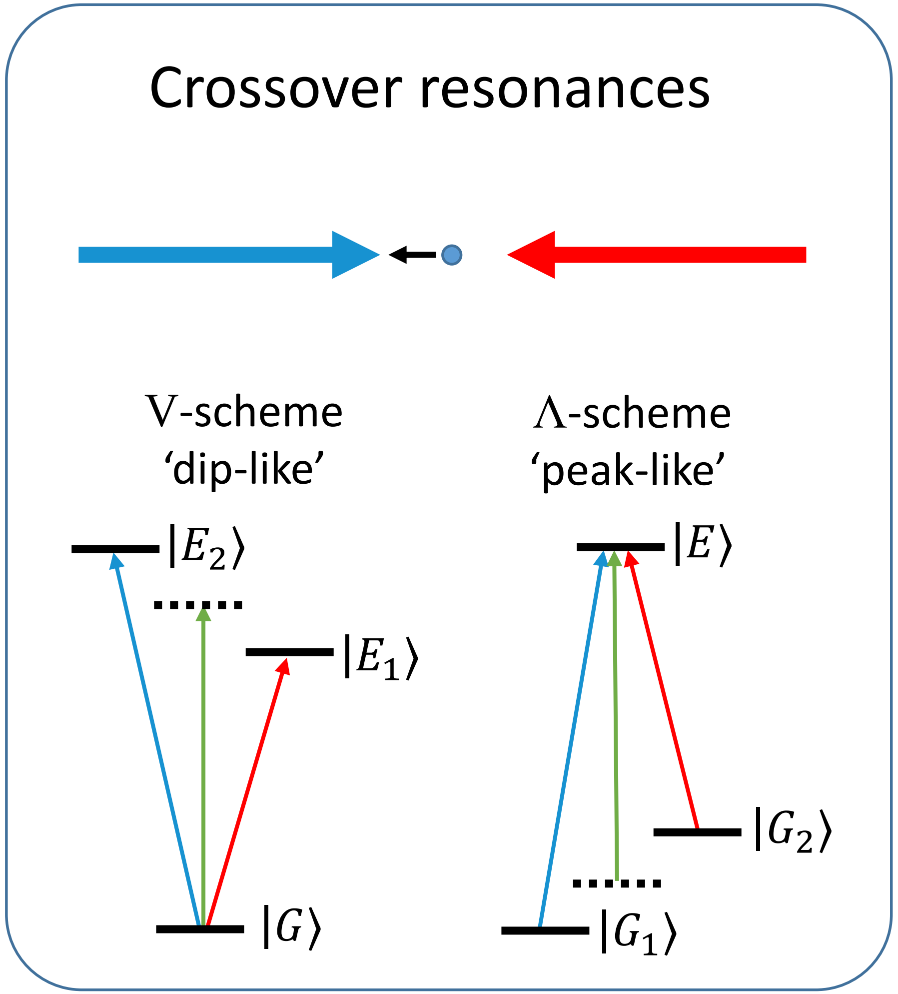
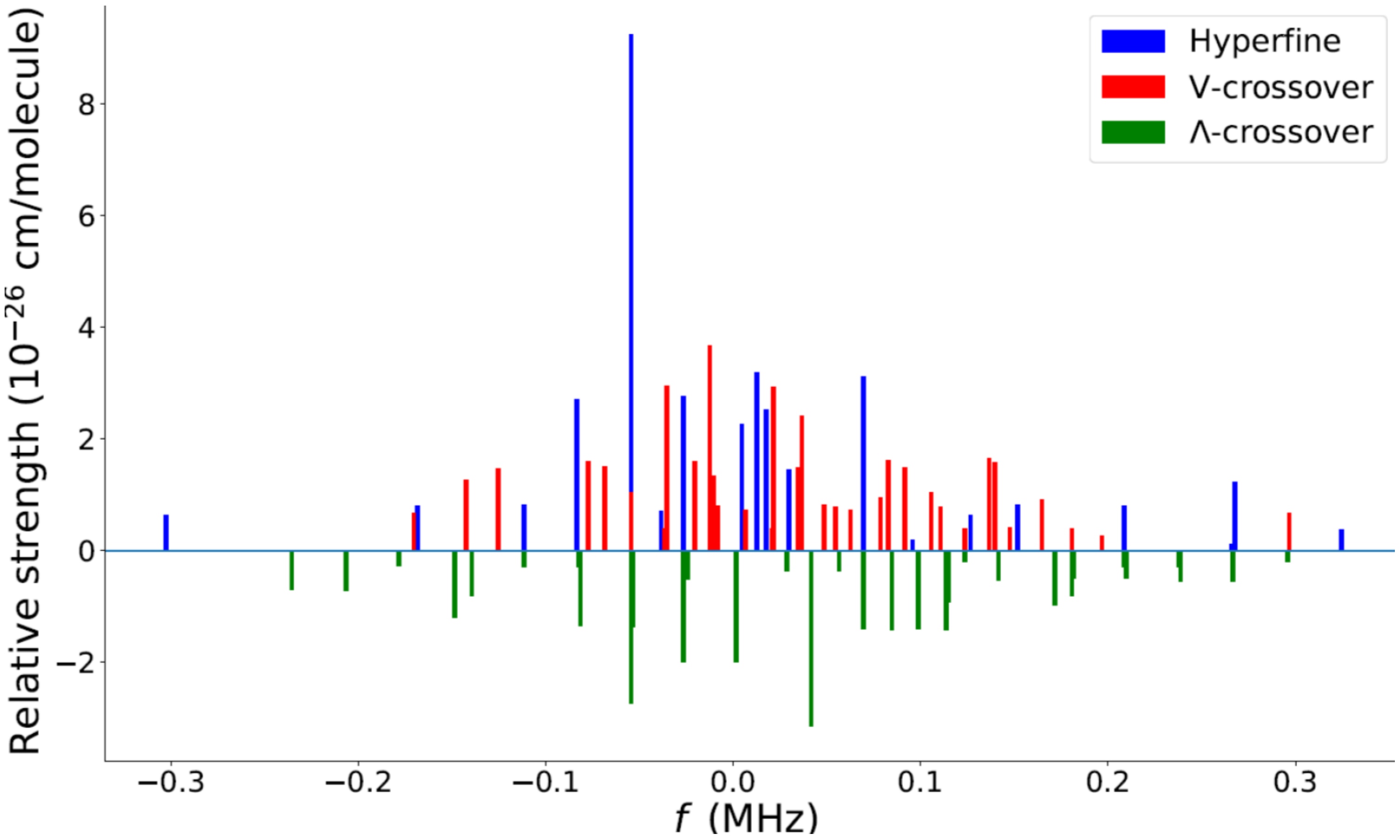




Crossovers effect



21 hyperfine transitions

36 V-crossovers

32 Λ-crossovers

Crossovers strength approximate

Density matrix simulation

- Hamiltonian $H = H_0 + \mu.\in$:
- $\frac{2\pi}{h}\mu_{ij}.\in \rightarrow \Omega_{ij}$, the rabi frequency between 2 states i and j
- Relaxation rates matrix γ
- **Liouville equation:**

$$\dot{\boldsymbol{\rho}} = -\frac{\mathbf{2}\boldsymbol{\pi}}{\boldsymbol{h}}\boldsymbol{i}[\boldsymbol{H}, \boldsymbol{\rho}] + \frac{\mathbf{1}}{\mathbf{2}}\{\boldsymbol{\gamma}, \boldsymbol{\rho}\}$$

  - Doppler profile : The spectrum is determined by summing different velocity of The Maxwell-Boltzmann distribution

Simple Bloch equations

$$\dot{\rho}_{ii} = \sum_j \frac{i}{\hbar}\left(V_{ij}\rho_{ij} - V_{ji}\rho_{ji}\right) + \gamma_{pop,ij}\rho_{jj}$$

$$\dot{\rho}_{jj} = \sum_i -\frac{i}{\hbar}\left(V_{ij}\rho_{ij} - V_{ji}\rho_{ji}\right) - \gamma_{pop,ij}\rho_{jj}$$

$$\dot{\rho}_{ij} = \sum_j (\frac{i}{\hbar} V_{ij}\left(\rho_{jj} - \rho_{ii}\right)) - (i\Delta_{ii} + \gamma_{coh,i,j}\rho_{ij})$$

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$$\dot{\rho}_{jj} = \sum_i -\frac{i}{\hbar}\left(V_{ij}\rho_{ij} - V_{ji}\rho_{ji}\right) - \gamma_{pop,ij}\rho_{jj}$$

$$\dot{\rho}_{ij} = \sum_j (\frac{i}{\hbar} V_{ij}\left(\rho_{jj} - \rho_{ii}\right)) - (i\boldsymbol{\Delta}_{ii}+\gamma_{coh,i,j}\rho_{ij})$$

Fixed $\gamma_{pop,ij} = 65kHz$ and varied $\gamma_{coh,i,j}$

$\gamma_{coh,i,j}$=$\gamma_{pop,i,j} + \gamma_p . P$

Why is a computer cluster needed ?

- Stationary solutions needed -> time of integration needs to be long enough

  Around 850 μ_s

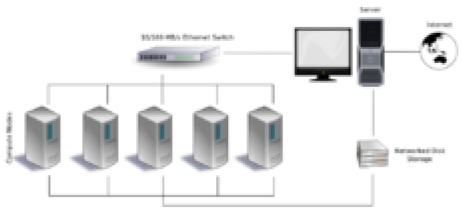
- Integration of the rate equation is done for each velocity
- The distance between two velocity components need to be smaller than the γ, the HWHM
  - For HD, let's assume γ=50Khz, the Doppler broadened γ is 750 MHz
  - So we need more than 15000 velocity components
- For each velocity the equation needs to be solved for all the detuning
  - Detuning needs to be smaller than the γ

So we need a calculation time of 15000x15000 at least.

Between 10 and 20 secs each

Lisa

Parallelization needed with the different Nodes



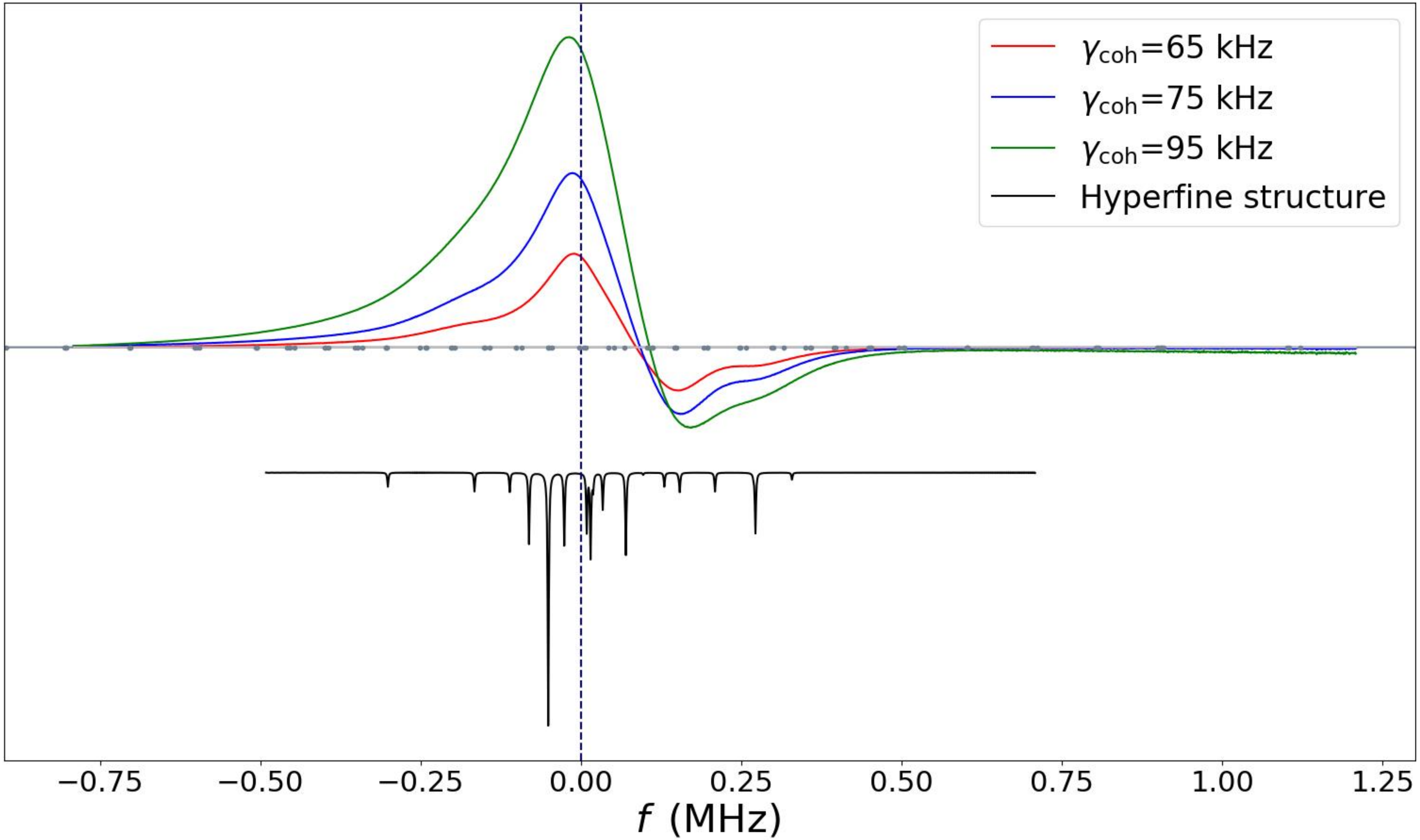
Normal pc task done in one Hour

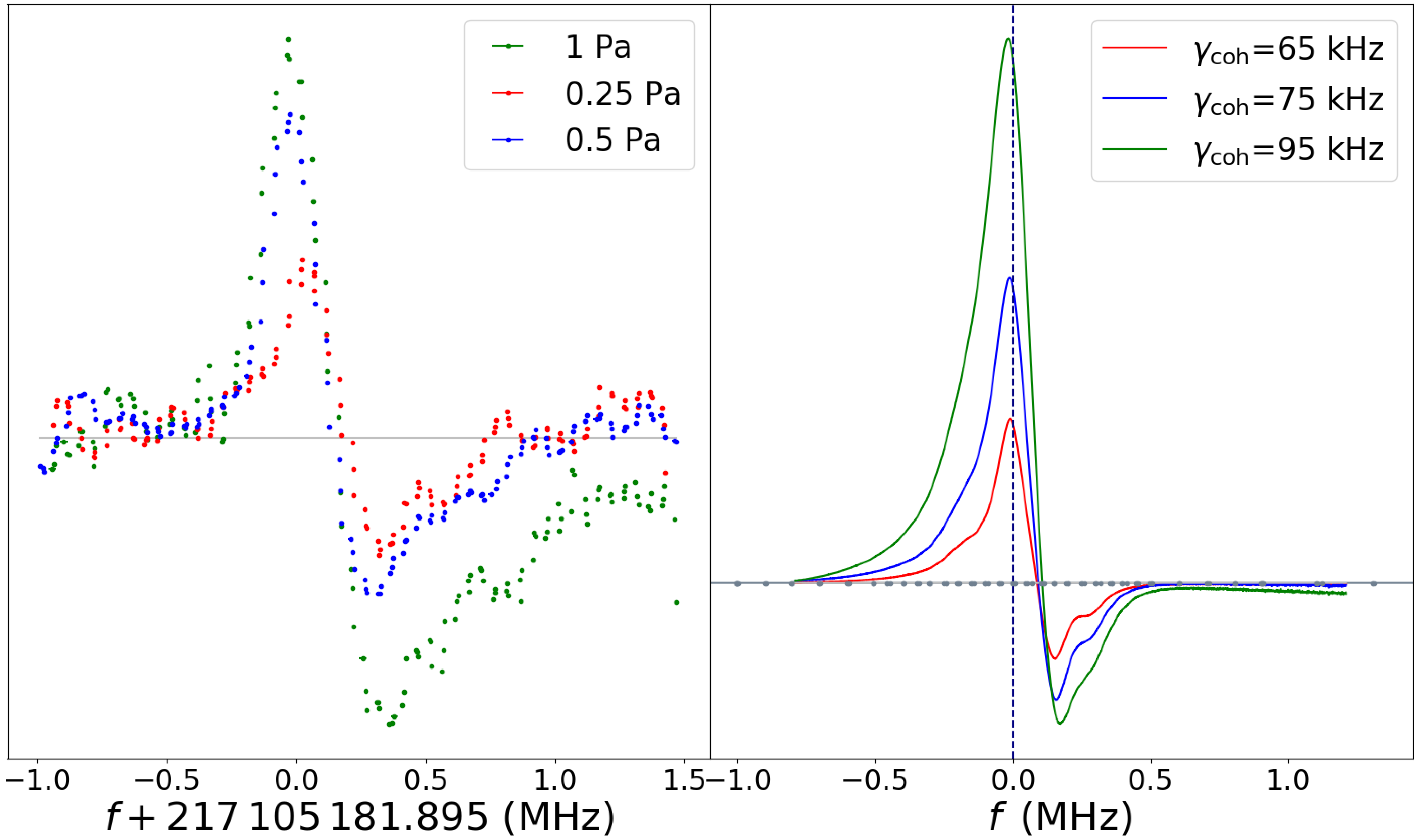
In Lisa : 1 min done

1 whole spectrum done in 18 hours

More info on:
https://userinfo.surfsara.nl/systems/lisa

Calculation outcome





1 Pa
0.25 Pa
0.5 Pa
−1.0
−0.5
0.0
0.5
1.0
1.5
f + 217 105 181.895 (MHz)
γcoh=65 kHz
γcoh=75 kHz
γcoh=95 kHz
−1.0
−0.5
0.0
0.5
1.0
f (MHz)

Remarks

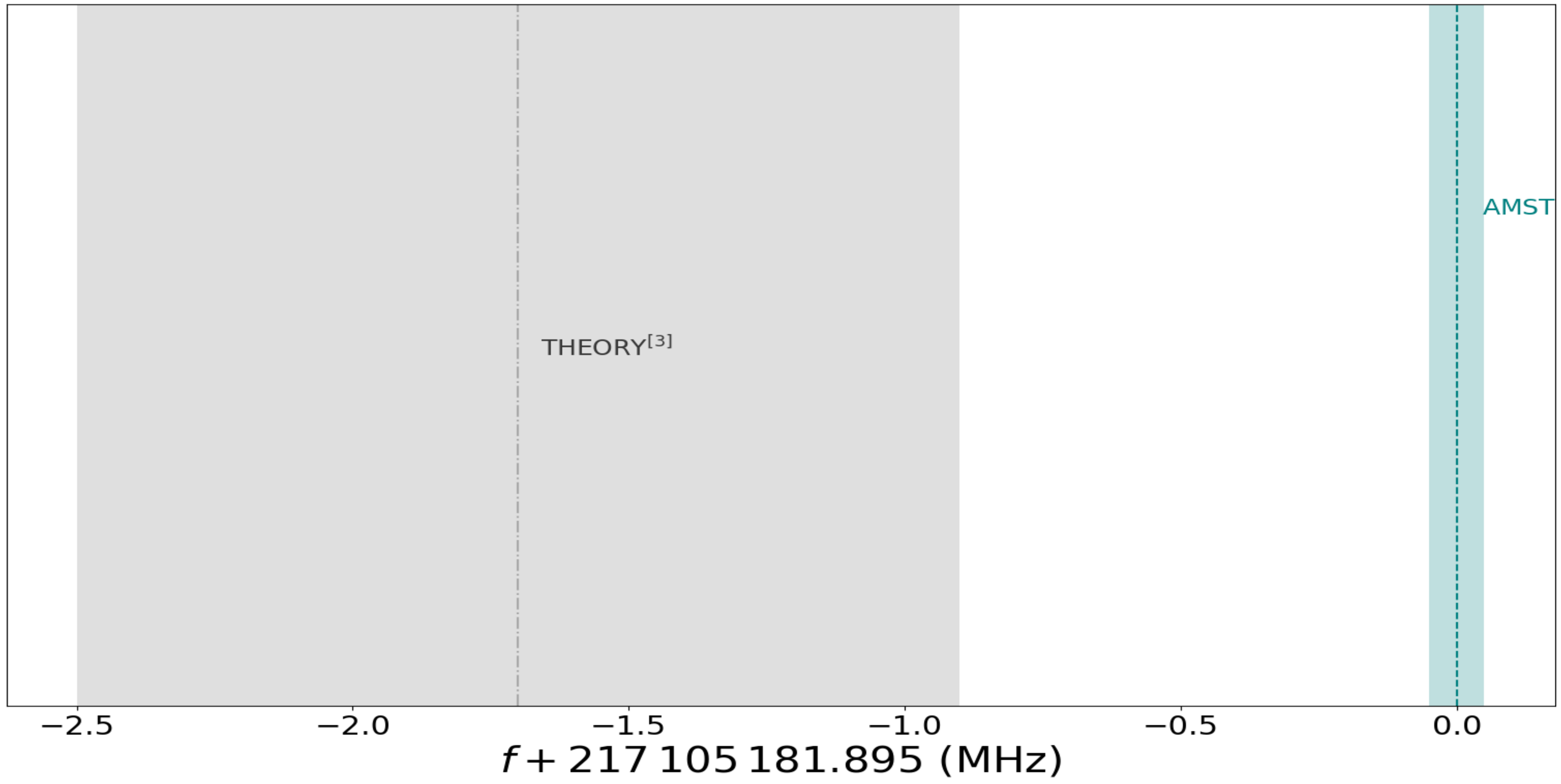
- The Lamb peak extrapolation **8kHz** from the Hyperfineless center

Peak frequency:

217 105 181.895 [50] MHz

- Theory : 217 105 180.200 [900] MHz

2σ



Conclusion

- HD spectroscopy gives rise to a complicated line profile
- Explanation of previous discrepancy
- Complex hyperfine structure gives rise to crossover
- Bloch equation can mimic that shape with certain parameters

Outlook

- Improve the Bloch equation model
- Cryogenic cooled cell is getting build
  - Enhancement of population of the lower states
  - Transit time longer
  - R(0) (top of water line) can therefore be reached
- R(0) hyperfine structure simpler

Acknowledgements

Science

European
Research
Council

NICE-OHMS team

- Frank Cozijn
- Meissa Diouf
- Edcel Salumbides
- Wim Ubachs
- Patrick Dupré
- Benoit Darquié

Technician

- Rob Kortekaas

Thank you