# Frequency comb spectroscopy in the THz range

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# Motivation

- Rotational transitions of small polar compounds and Vibrational transitions of floppy molecules showing low-frequency modes may be probed with the mm-wave/THz radiation.
- At the Doppler limit (low-pressure), individual rotational lines without overlapping are observed. The selectivity available in the THz allows to study a complex mixture of gases or to discriminate species with close chemical structures! **Sub MHz resolution is required**
- A polar compound may be absolutely quantified from its rotational intensity without any standard of calibration. Moreover a trace gas detection in the THz is possible for many molecules which present strong rotational transition intensities.

However

- At atmospheric pressure, collisional broadening becomes large involving a degradation of the selectivity and the sensitivity.
- At THz frequencies, the absorption of the water vapor continuum is very strong and the THz waves may be propagated only for a distance of several meters.
- The emitted power of the THz sources rest  relatively weak

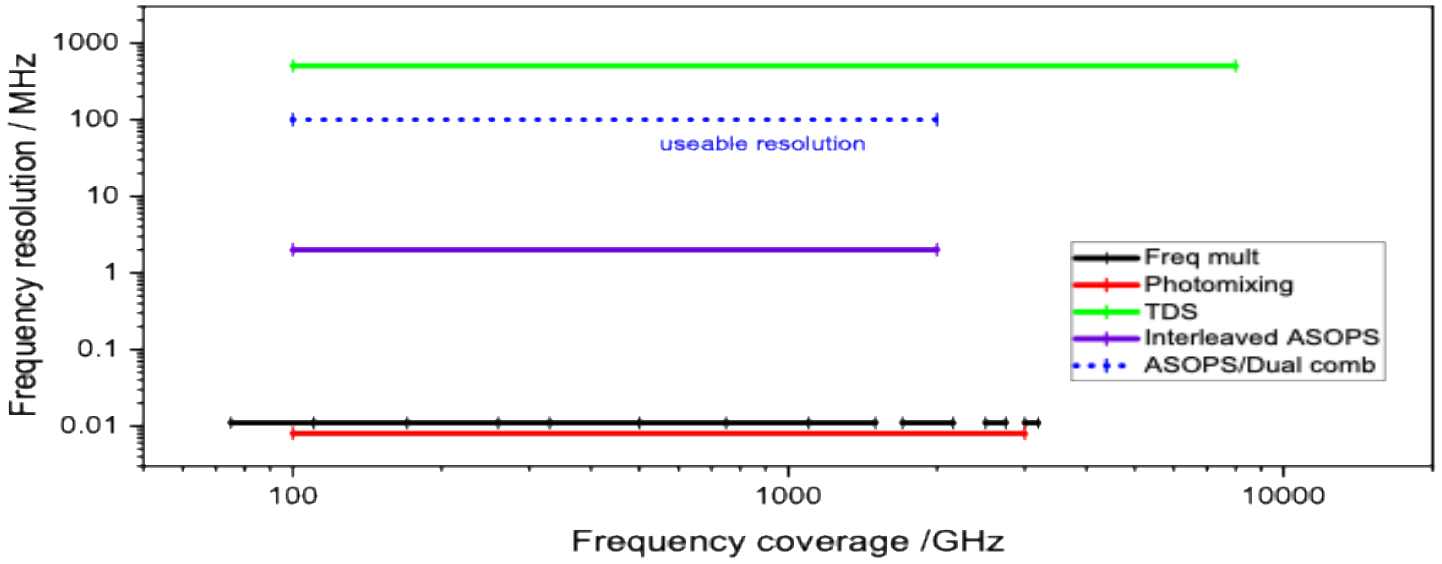
# THz Instrumentation

sensitivity, **resolution, acquisition time**, spectral coverage

- Frequency domain spectroscopy
  - Upconversion – straight forward
  - Downconversion – Photomixing
  - Speed between 1 and 10 GHz/hr

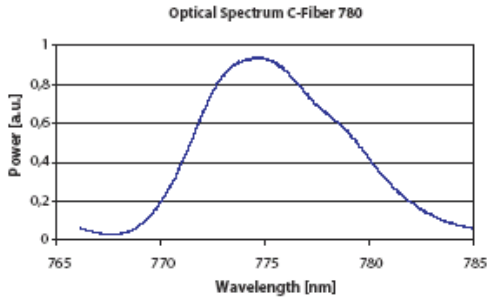
- Time Domain Spectroscopy
  - Resolution limited to around 500 MHz
  - Acquisition time approx. 20 minutes for best performance
  - Complete band is obtained

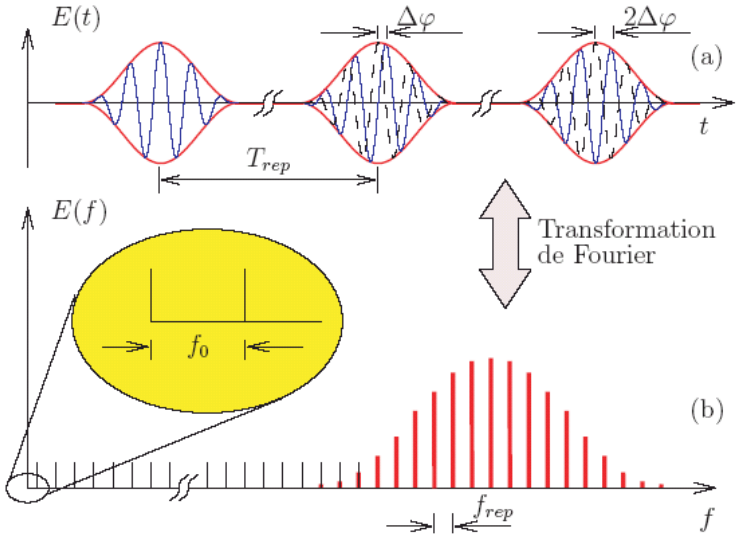
- Dual Comb/ASOPS
  - Useable resolution of $f_{rep}$
  - Interleaving possible




Compromise between resolution and acquisition time.

# Frequency comb



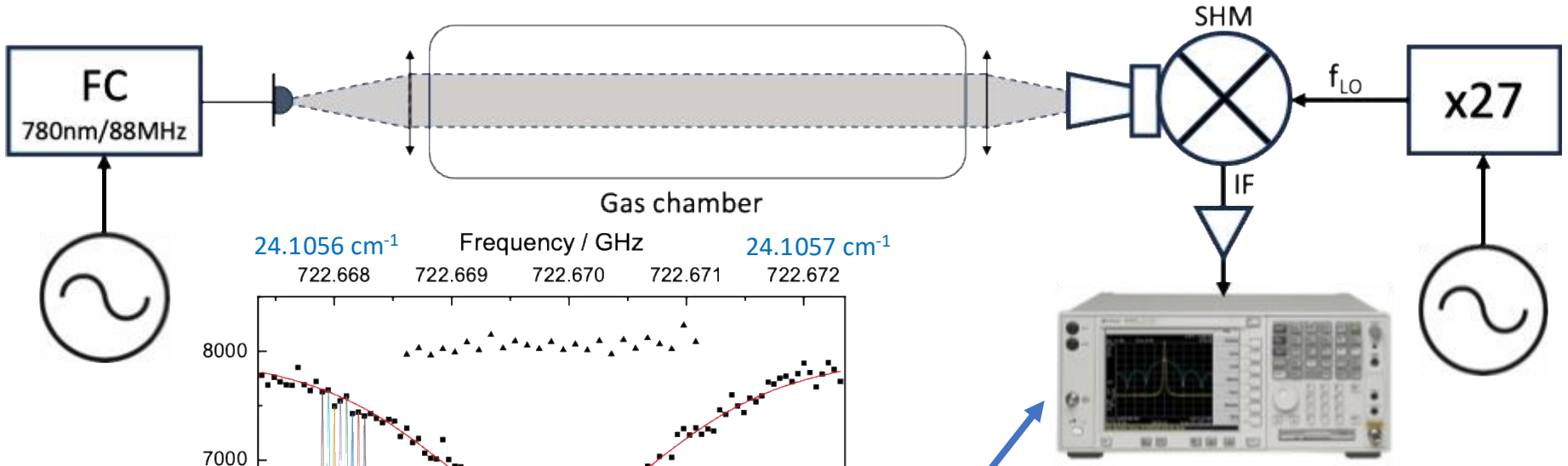



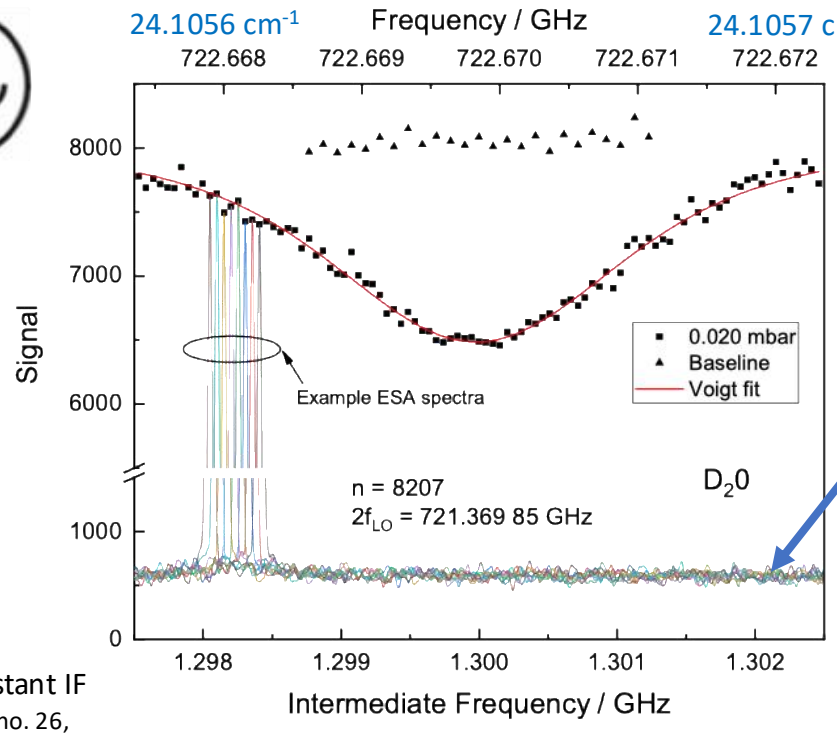


- Frequency combs have excellent resolution capabilities due to the fine line widths
- Easily and directly transferable to the THz domain
- No Carrier Envelope Offset (CEO)
- Point spacing at $f_{rep}$ leads to non-uniform resolution

- Varying the $f_{rep}$ allows all frequencies to be covered,
- Mulitple acquisitions should allow a uniform covergae
- Must have discrimination down to a single THz FC mode

# FC with heterodyne detection – Single mode



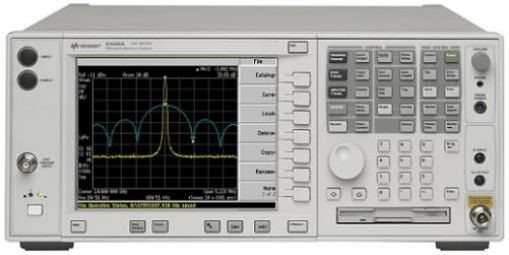




- IF analysis with a standard spectrum analyser
- Variation of $f_{rep}$ to construct spectra
- Many modes may be observed coverage up to 80 GHz
- Becomes very inefficient and time consuming to monitor more than a single mode

mm-wave single line with constant IF

S. Skryl *et al* Opt. Express, vol. 22, no. 26, pp. 32276–32281, 2014, doi: 10.1364/OE.22.032276.

# Detection – Multiple FC modes



Standard ESA, unable to measure two frequencies simultaneously

- multiple FC modes may be observed

**Intermediate Frequency analysis by XFFTS**

"eXtended bandwidth a Fast Fourier Transform Spectrometer"

*developed for radio astronomical applications*

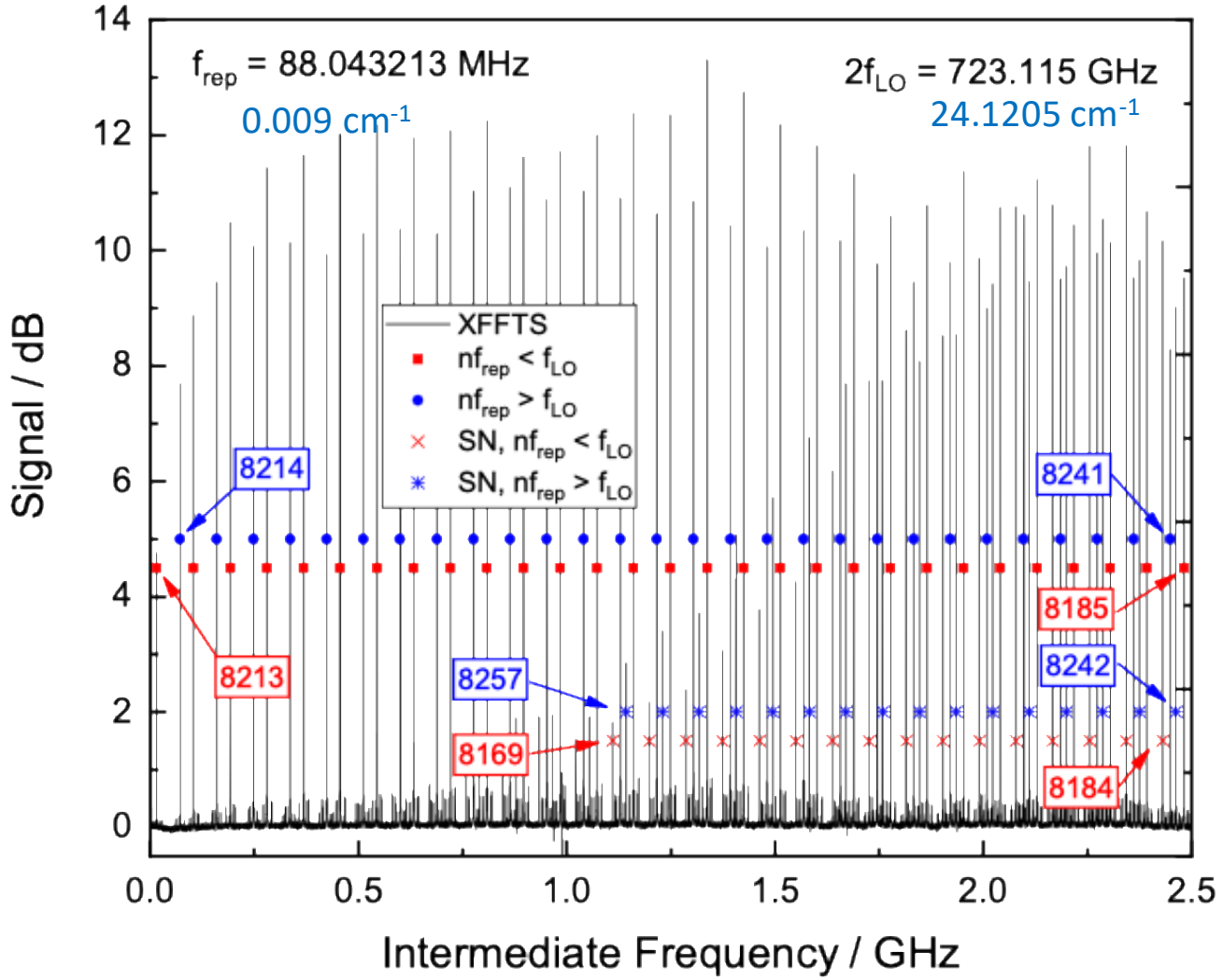
- sensitive
- wide-band digital frequency domain analysers
- high-speed analogue to digital converters and field programmable gate arrays.
- 2.5 GHz of analysis bandwidth
- 32k equal frequency channels (width 76 kHz, $2.5 \times 10^{-6} cm^{-1}$)
- all channel are accumulated simultaneously
- Not particularly user friendly

B. Klein, *A&A*, vol. 542, p. L3, Jun. 2012, doi: 10.1051/0004-6361/201218864.

# Example XFFTS spectrum,

32768 channels
89 FC modes visible (dynamic > 6dB)

| Visible mode | 8169 - 8184 | 8185 - 8213 | 8214 - 8241 | 8242 - 8257 |
|---|---|---|---|---|
| Nyquist image | 2nd | **1st** | **1st** | 2nd |
| FC above below LO | Below | **Below** | **Above** | Above |
| Electrical BW GHz | 2.5 – 5 GHz | **0 – 2.5 GHz** | **0 – 2.5 GHz** | 2.5 – 5 GHz |
| Spectral band GHz | 718.1– 723.1 | **720.6 - 723.1** | **723.1 – 725.6** | 725.6 – 728.1 |
| Modes | 16 | **29** | **28** | 16 |

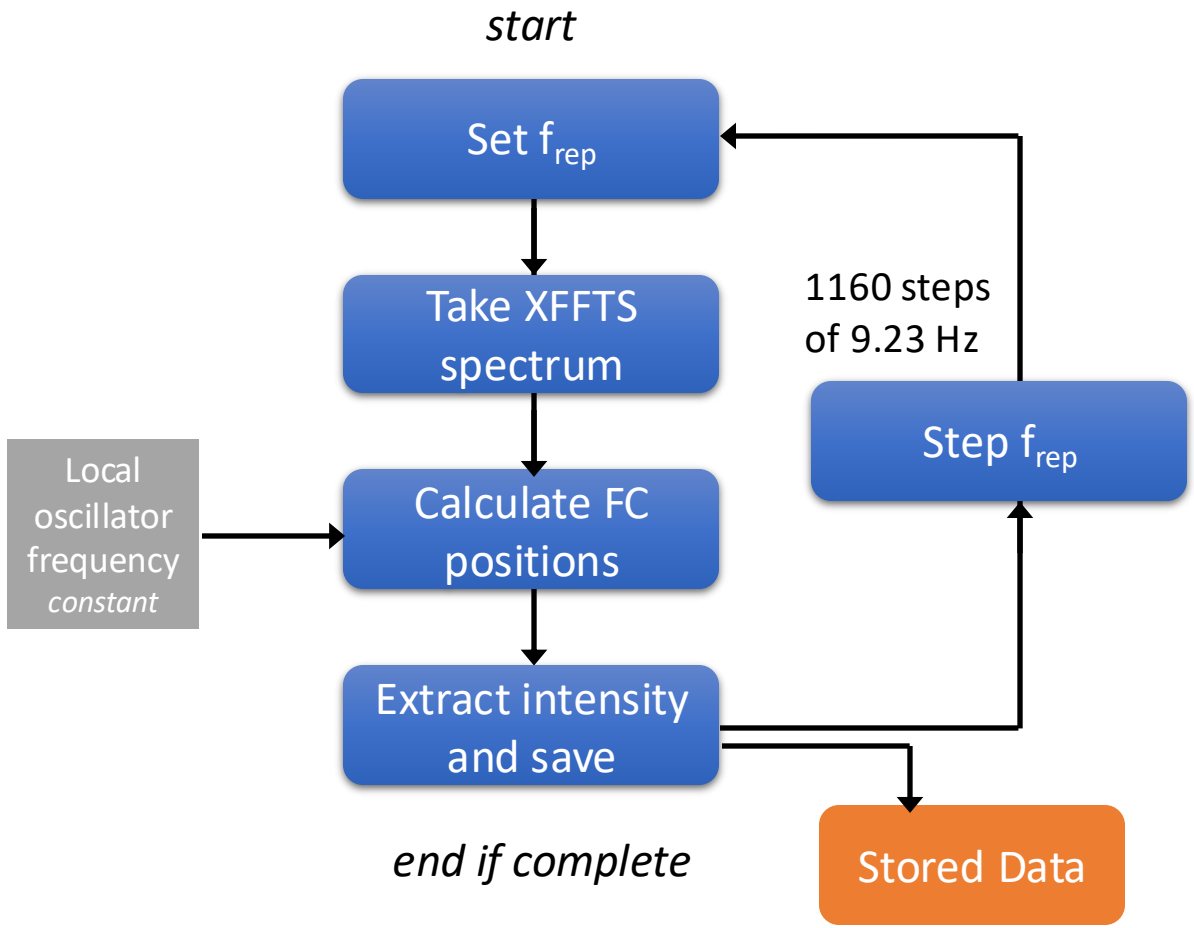


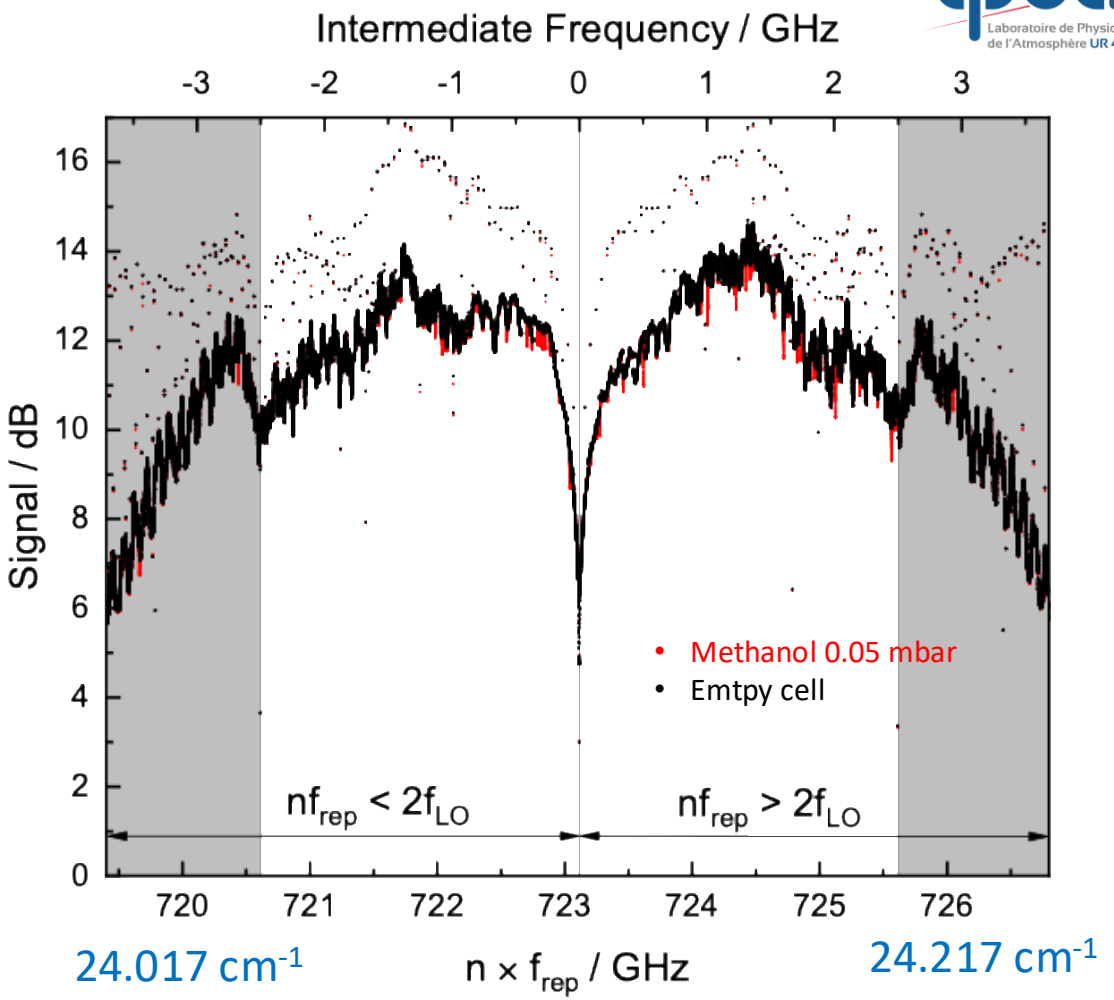

Total spectral bandwidth around 7.5 GHz (0.25cm$^{-1}$)

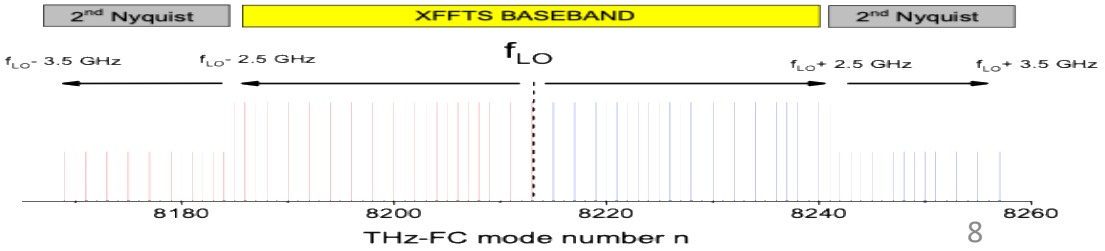
# Vary $f_{rep}$

$$\delta f_{rep} = \frac{{f_{rep}}^2}{\nu_{THz}}$$

Total variation to cover all frequencies at 723 GHz, 10.7 kHz

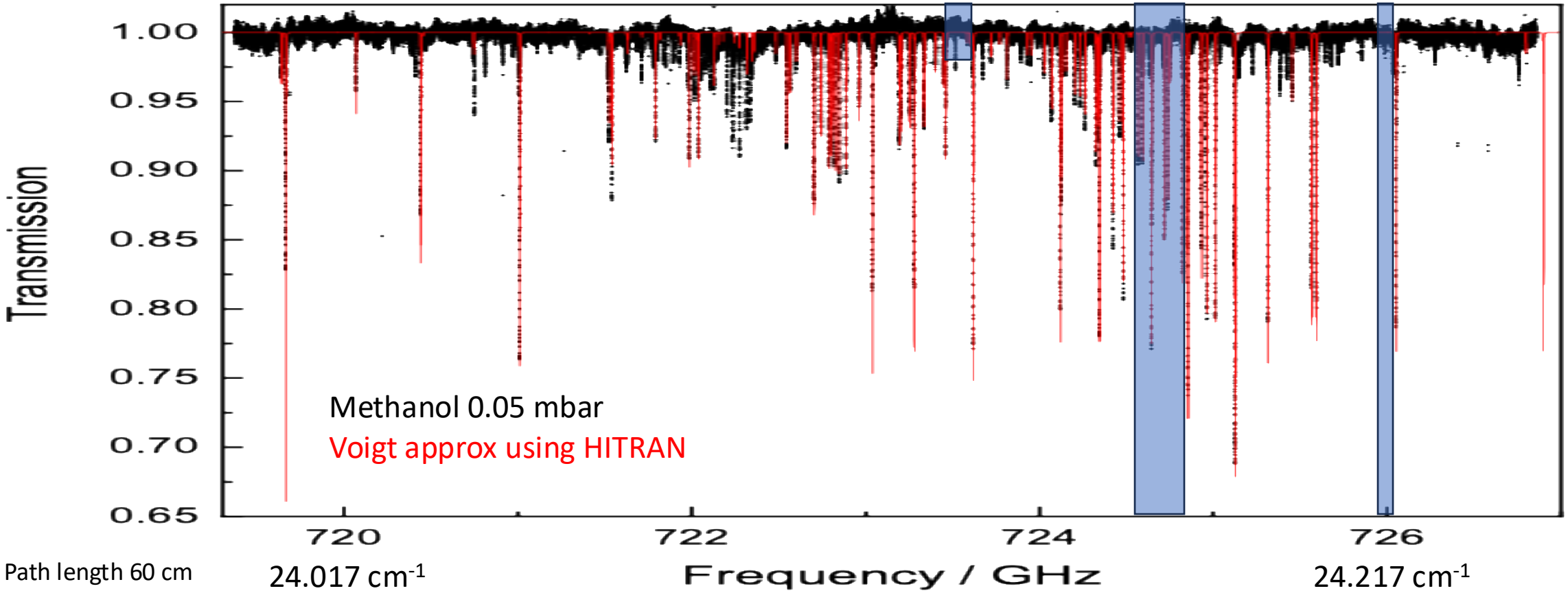


# Spectrum of Methanol

- Coverage 7.48 GHz, 0.25 $cm^{-1}$
- 98600 points
- Spacing 76 kHz, 2.5x$10^{-6}$$cm^{-1}$
- Acquisition time 20 minutes




Path length 60 cm

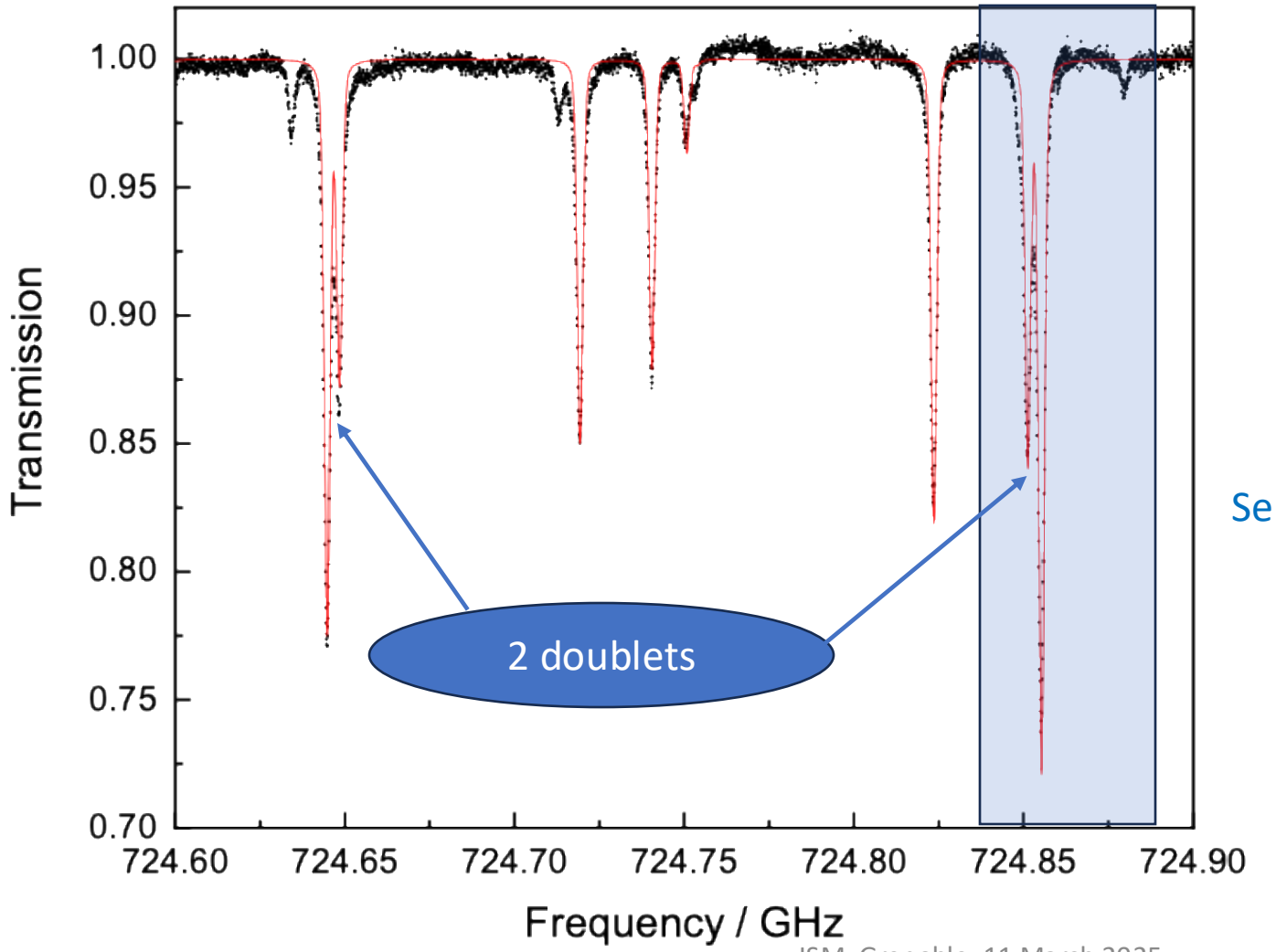
24.017 $cm^{-1}$

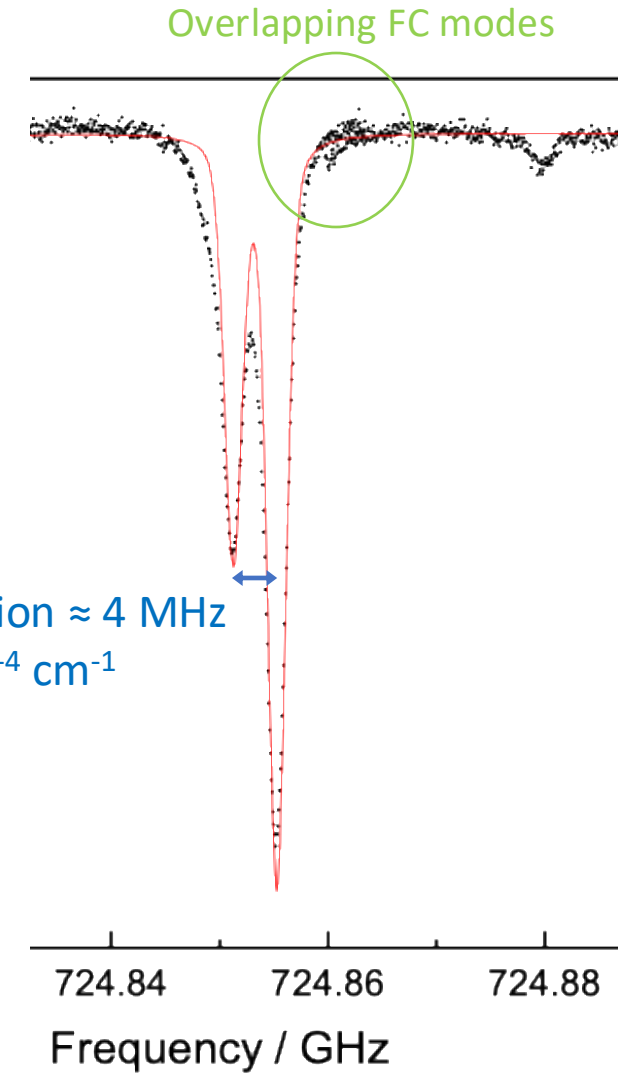
24.217 $cm^{-1}$



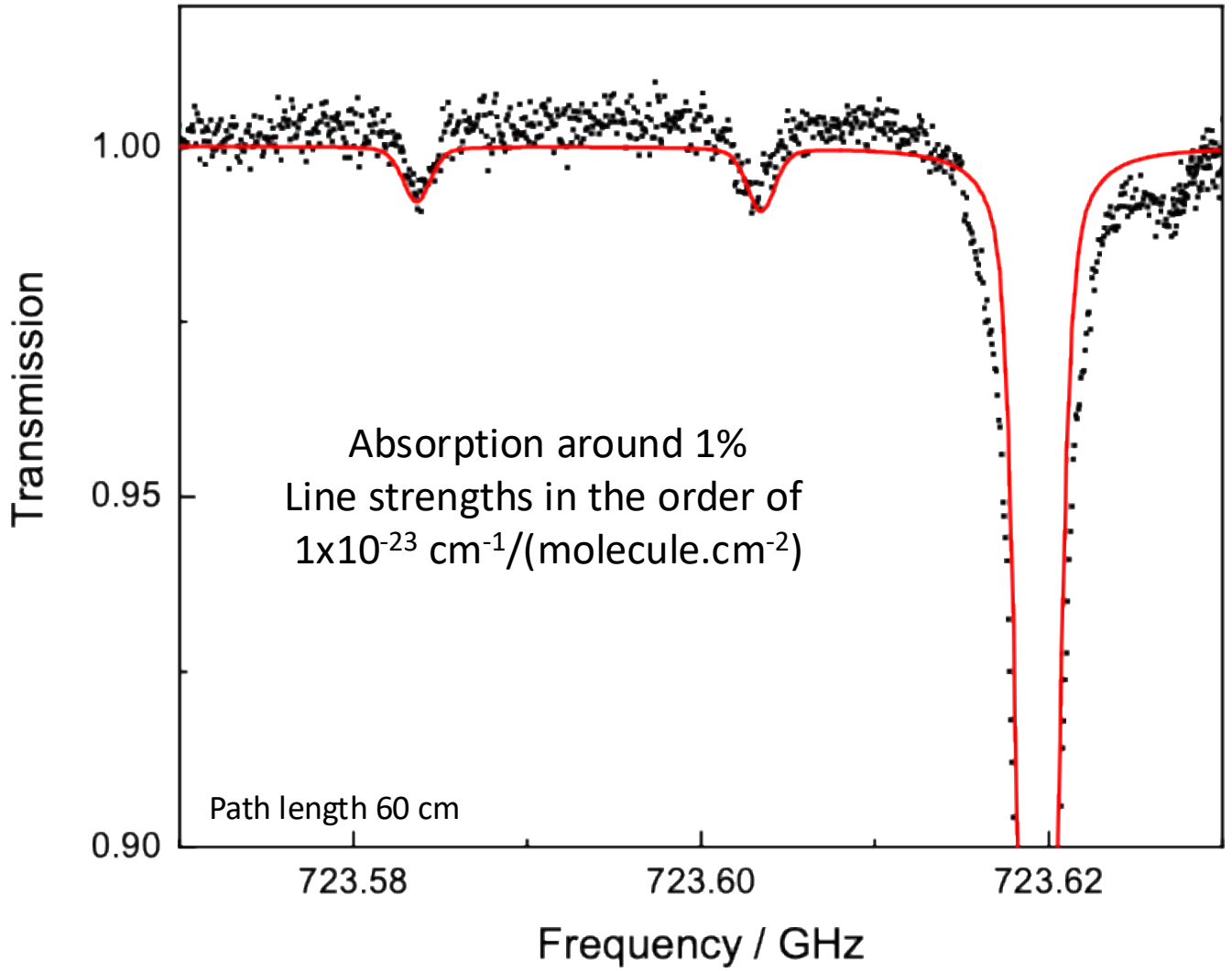
*150 molecular lines are observed, 80 can be directly identified*

# Resolution

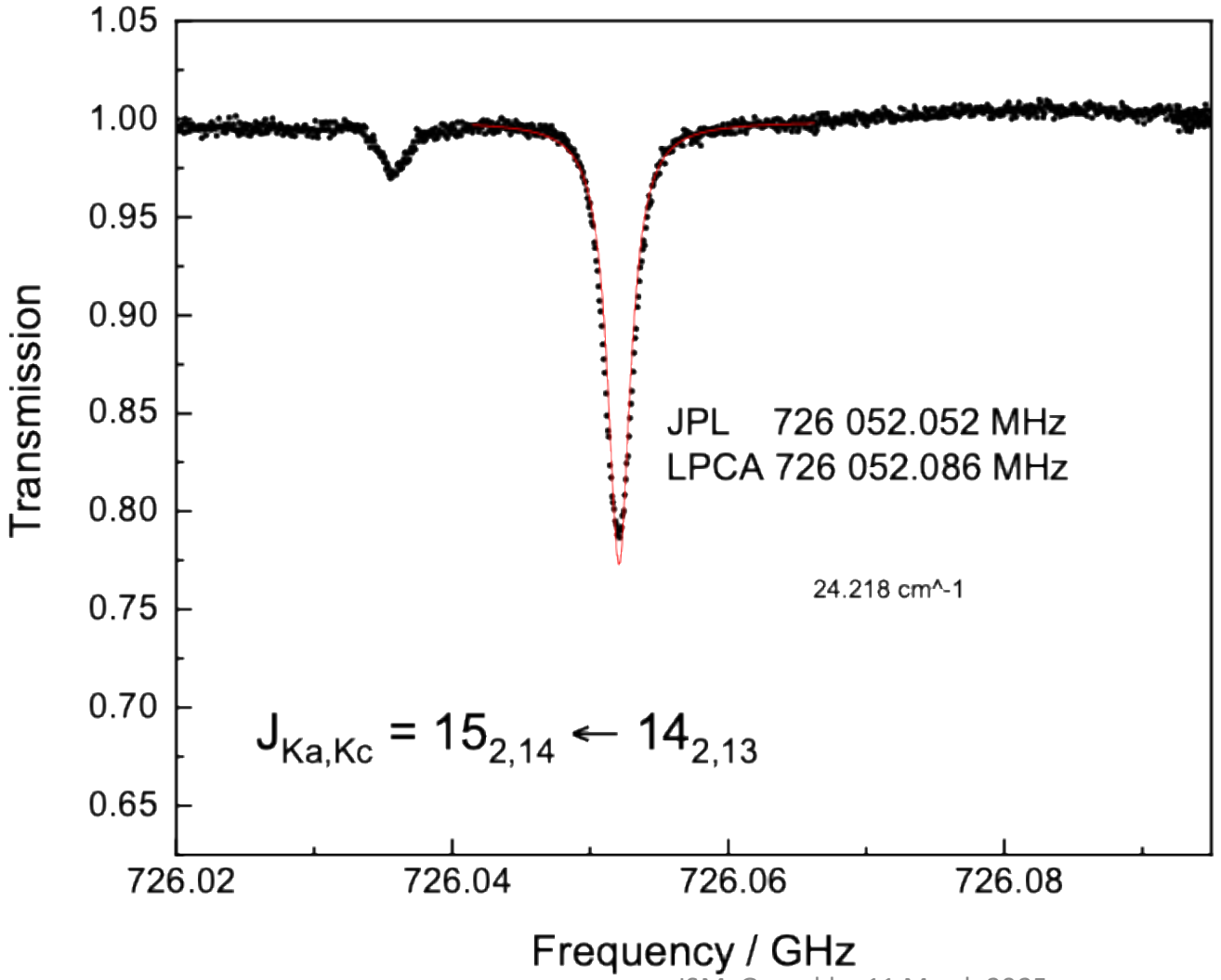

Transmission
1.00
0.95
0.90
0.85
0.80
0.75
0.70
724.60
724.65
724.70
724.75
724.80
724.85
724.90
Frequency / GHz
2 doublets


Overlapping FC modes
Separation ≈ 4 MHz
10-4 cm-1
724.84
724.86
724.88
Frequency / GHz

# Sensitivity



# Line centre measurement




- Fitted with Voigt profile
- 30 kHz from tabulation

L.-H. Xu *et al.*, 'Torsion–rotation global analysis of the first three torsional states ($v_t$ = 0, 1, 2) and terahertz database for methanol', *Journal of Molecular Spectroscopy*, vol. 251, no. 1, pp. 305–313, Sep. 2008, doi: 10.1016/j.jms.2008.03.017.

Lines measured with an experimental accuracy ranging from 50 to 200 kHz

# Perspectives

- Zone available 500 – 750 GHz, dynamic > 10 dB should be exploited for frequency measurement and validation.
- Extension of the bandwidth, no time penalty
  - Additional XFFTS cards, next generation XFFTS (32 GHz of spectral coverage)
  - Mixer electrical bandwidth limit approx. 40 GHz
  - Data processing may become a problem
- Mixer at 1 THz available
- Finer resolution is possible
- FC provides frequency metrology
  - Replace LO by molecular laser to access (higher frequencies 1-4 THz)

Postdoc position currently available