(UV) Dual Comb Spectroscopy: Ultra-High Resolution Molecular Spectra in Milliseconds

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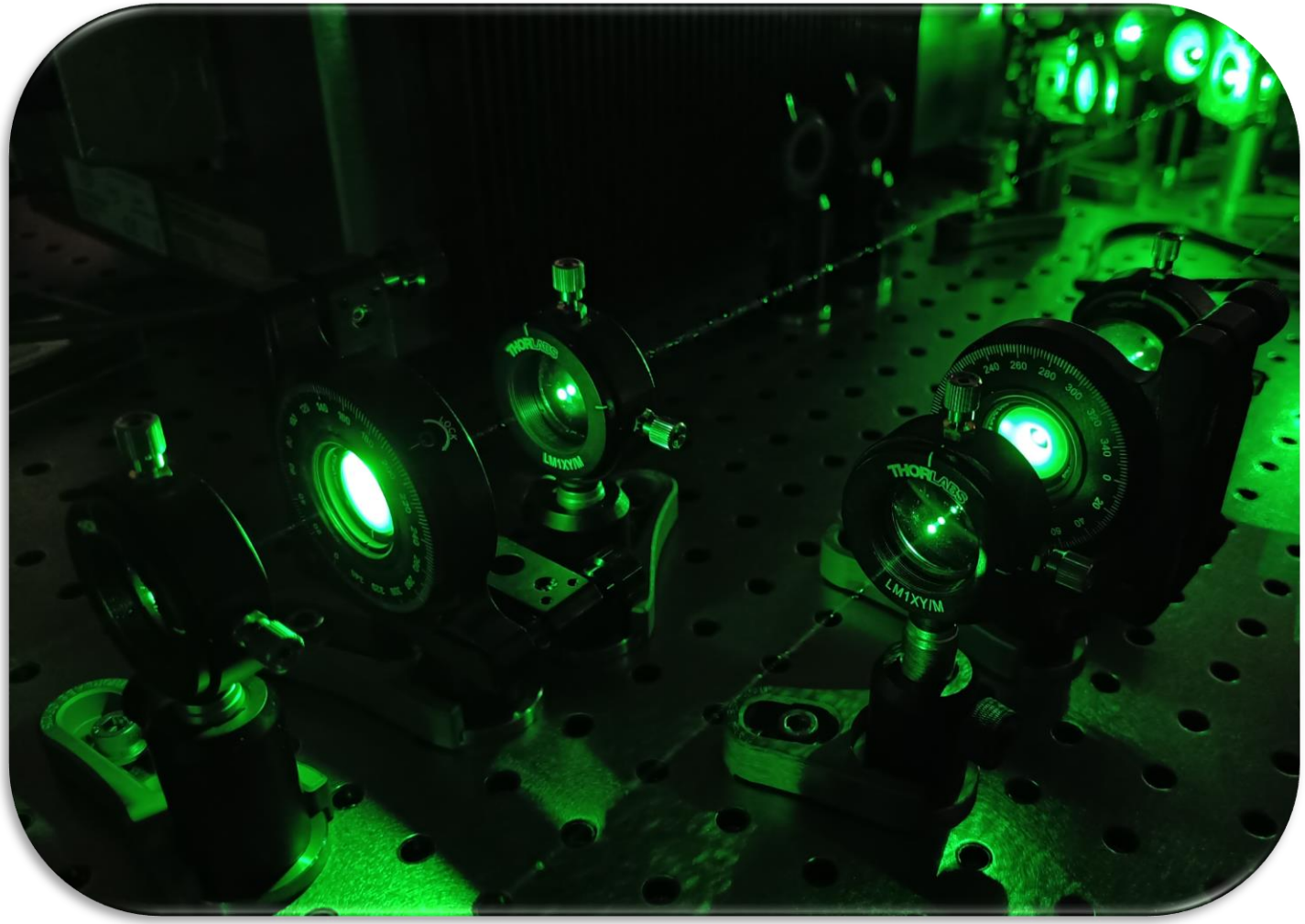
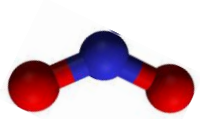
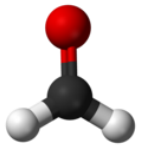
[1]Graz University of Technology,
[2]OeAW Space Research Institute

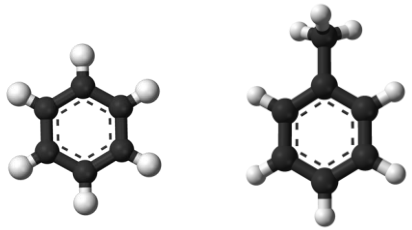


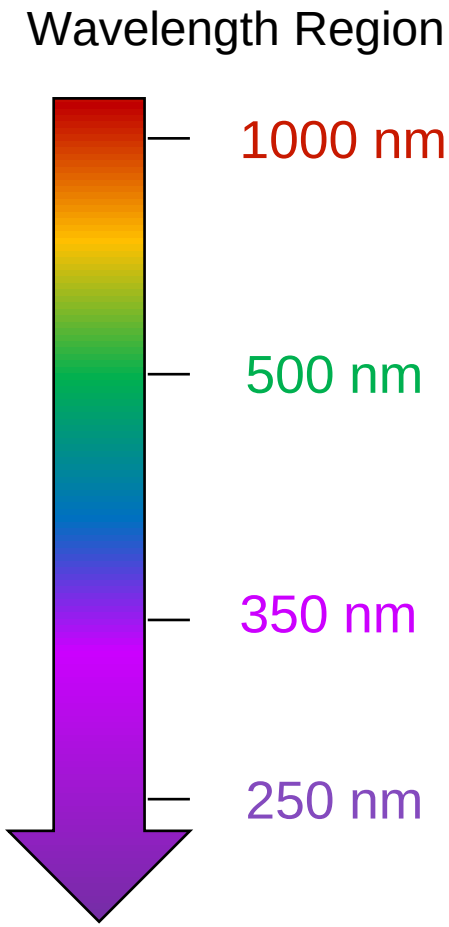
Outline

- Dual Comb Spectroscopy
- Nitrogen Dioxide (VIS)
- Formaldehyde (NUV)
- Benzene & Toluene (DUV)









Why High Resolution & High Speed?

Current methods:

or

Slow & accurate

Resolve rotational structure

✓

Time-dependent measurements

✗

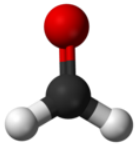
Fast & low resolution

Resolve rotational structure

✗

Time-dependent measurements

✓



Dual Comb Spectrometer

Resolve rotational structure

✓✓✓

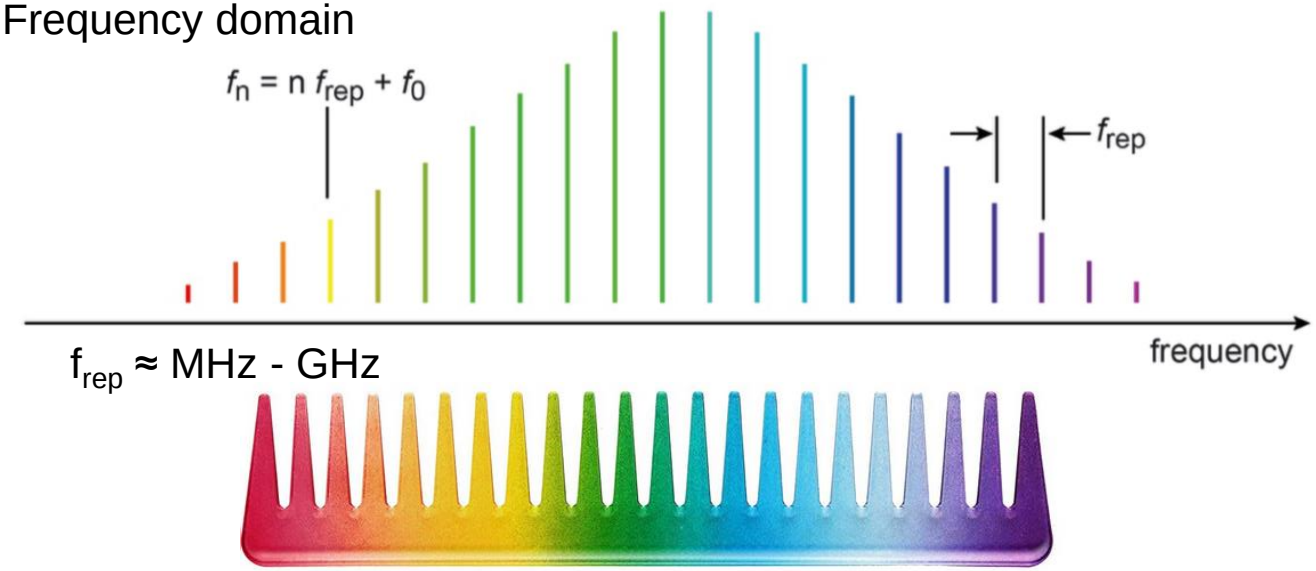
Time-dependent measurements

✓

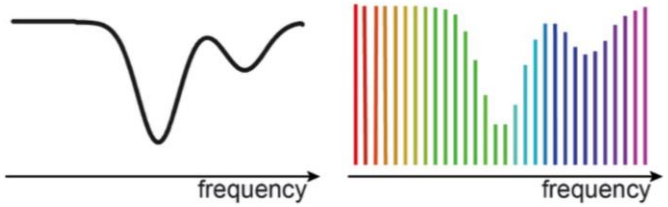
Frequency Combs

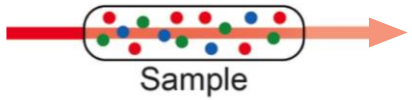
Glauber, Hall & Hänsch, 2005

Frequency domain



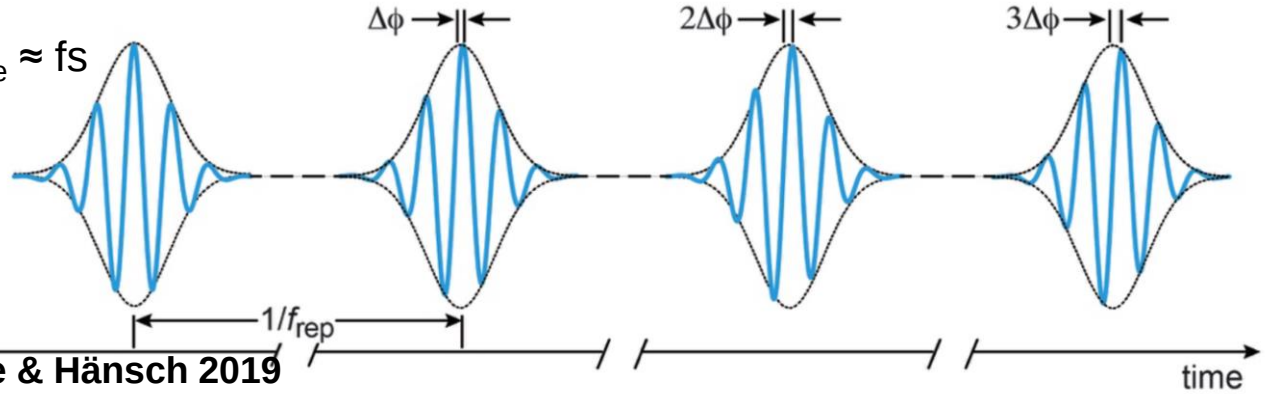
$f_{rep} \approx$ MHz - GHz





Time domain

$t_{pulse} \approx$ fs



Picque & Hänsch 2019
DOI:10.1038/s41566-018-0347-5

High absolute precision! 10^{-14}

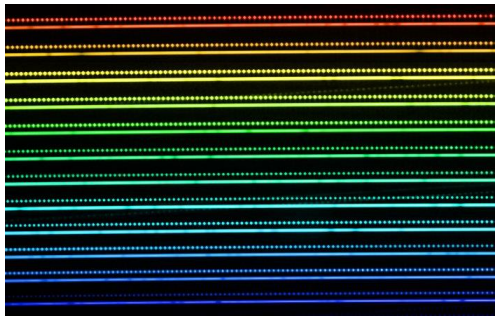
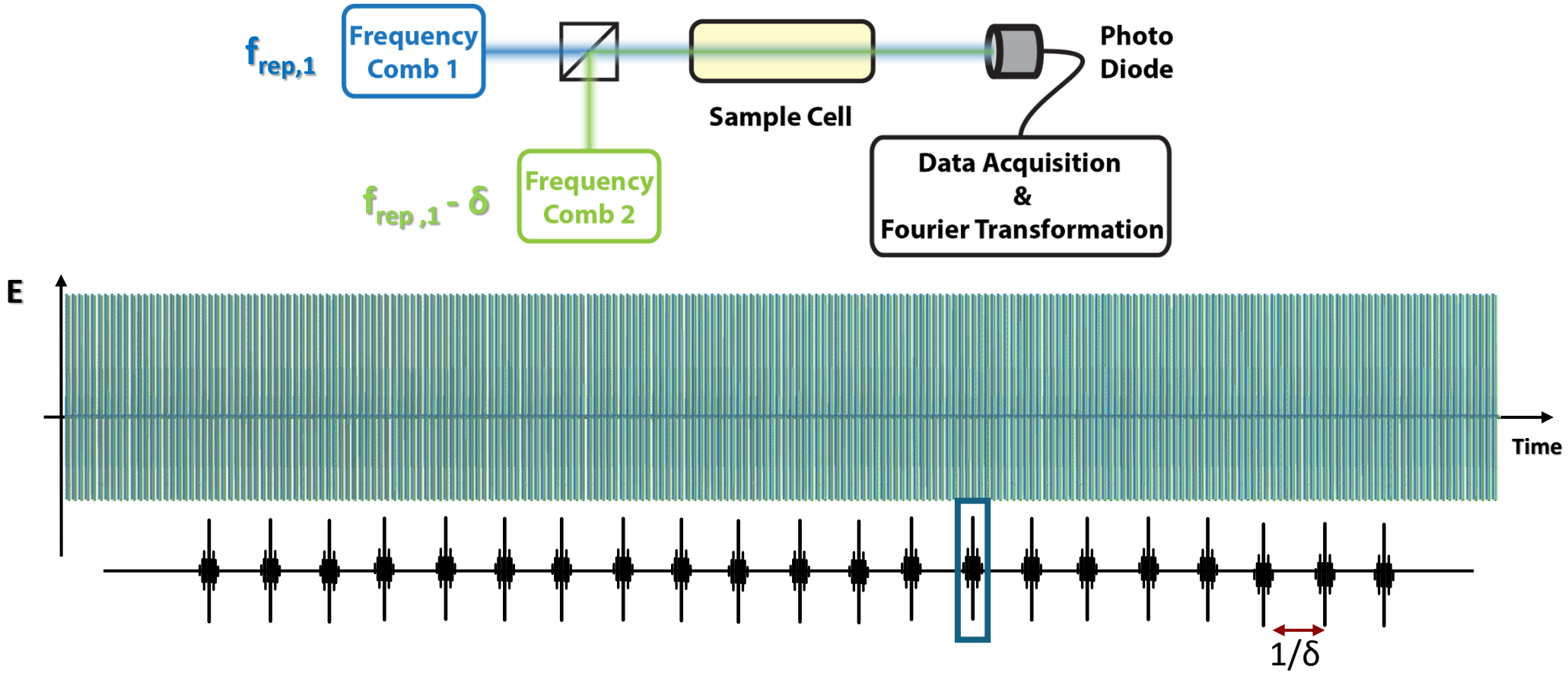
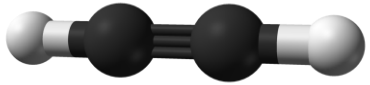


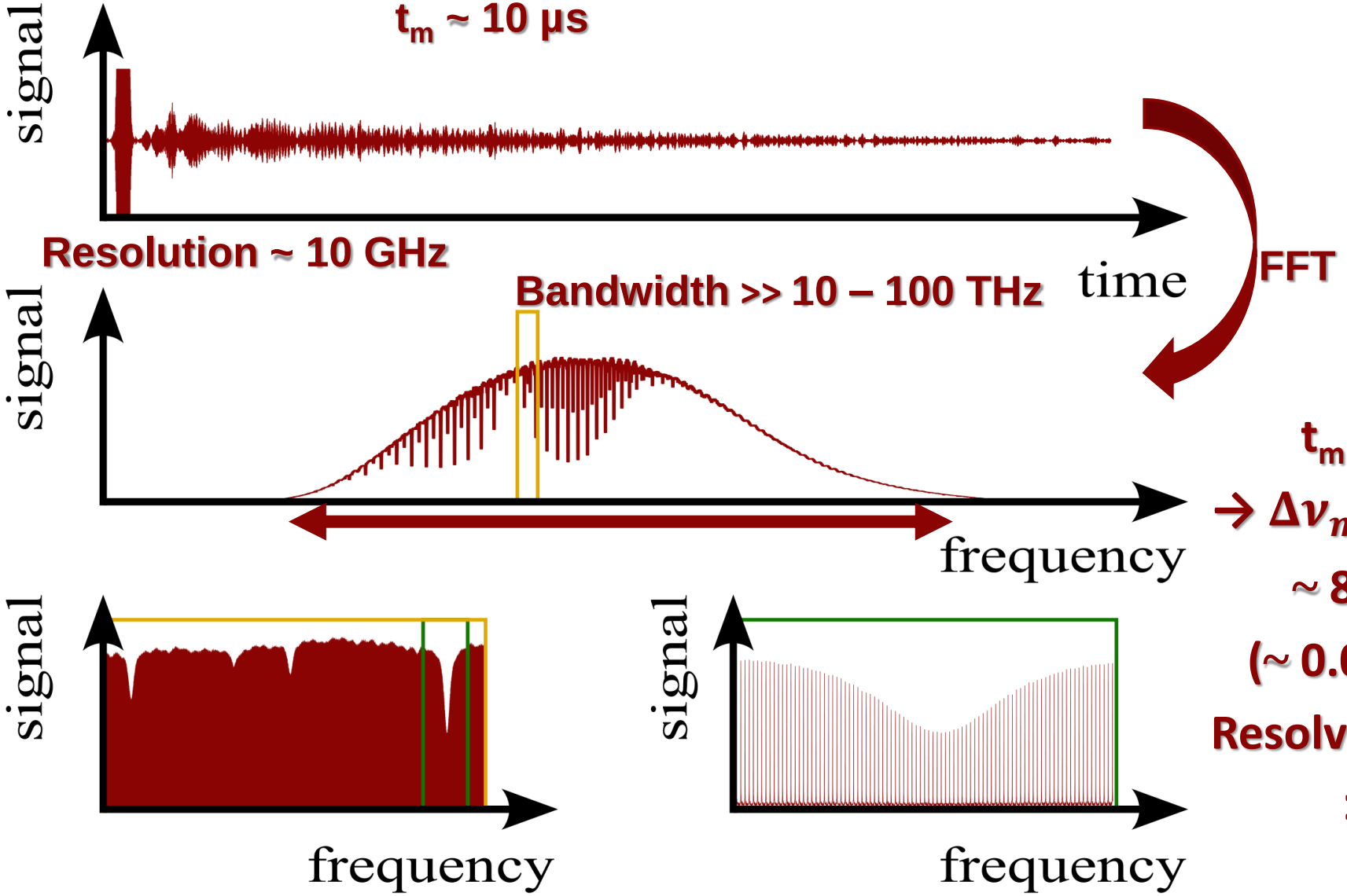
Image credit: ESO

Dual Comb Spectroscopy (DCS)



Strengths of DCS





t_m ~ 10 s

→ $\Delta\nu_{max} = f_{rep}$

~ 80 MHz

(~ 0.003 cm^{-1})

Resolving Power:

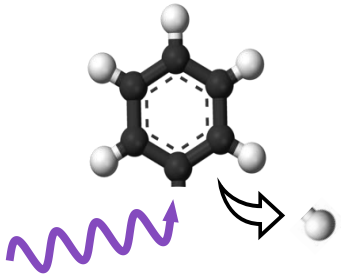
>10^6

Pal et al. submitted
DOI: 10.48550/arXiv.2412.05972

Why UV & Visible?

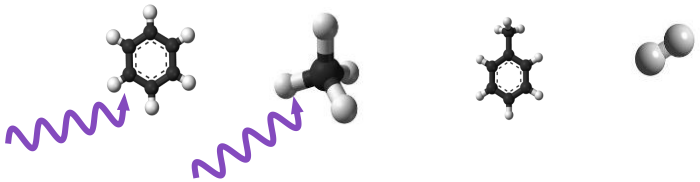
Atmospheric chemistry

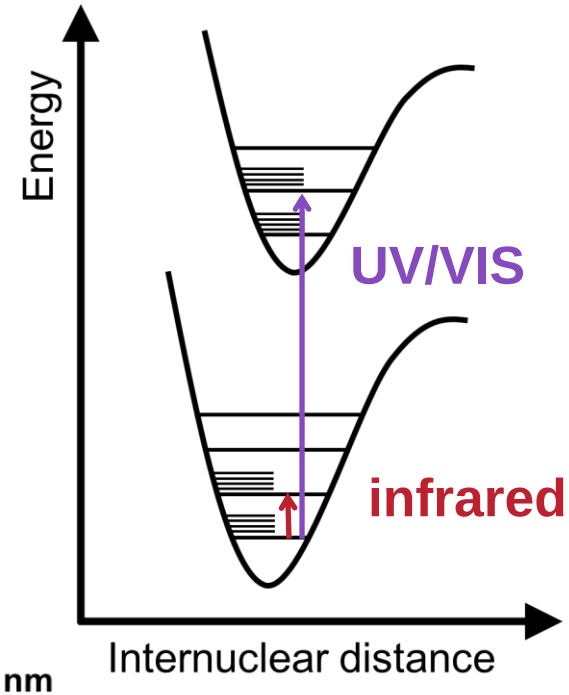
- Photodissociation



- Forming new molecules

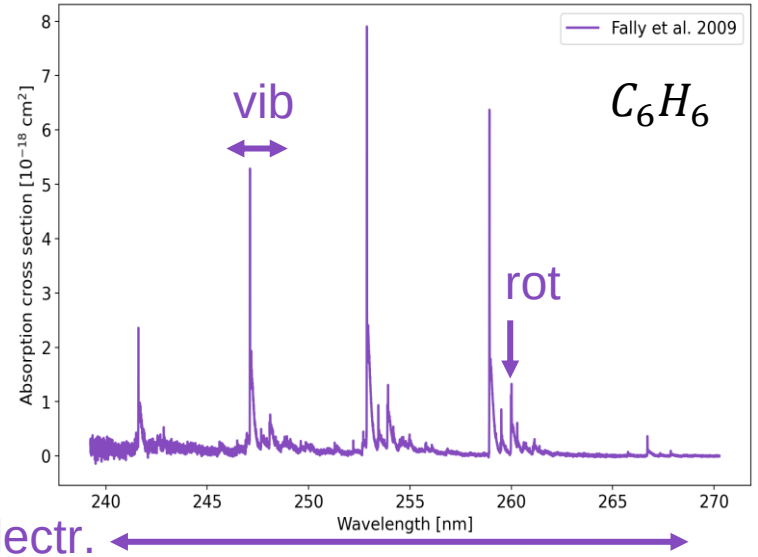
$$C_6H_6 + CH_4 \xrightarrow{UV} C_6H_5CH_3 + H_2$$

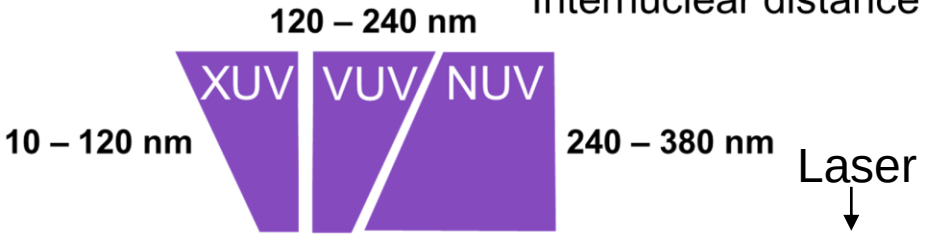




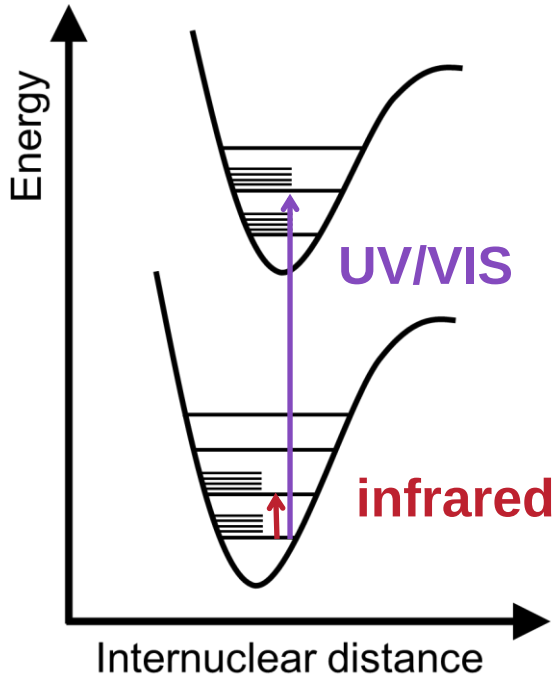
Uniquely identify molecules

- Vibrational, rotational and electronic transitions

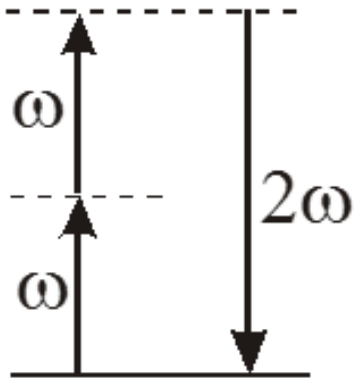




Accessing New Wavelengths



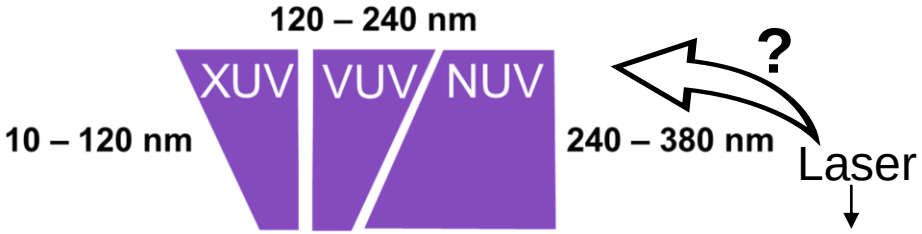
- Nonlinear frequency upconversion



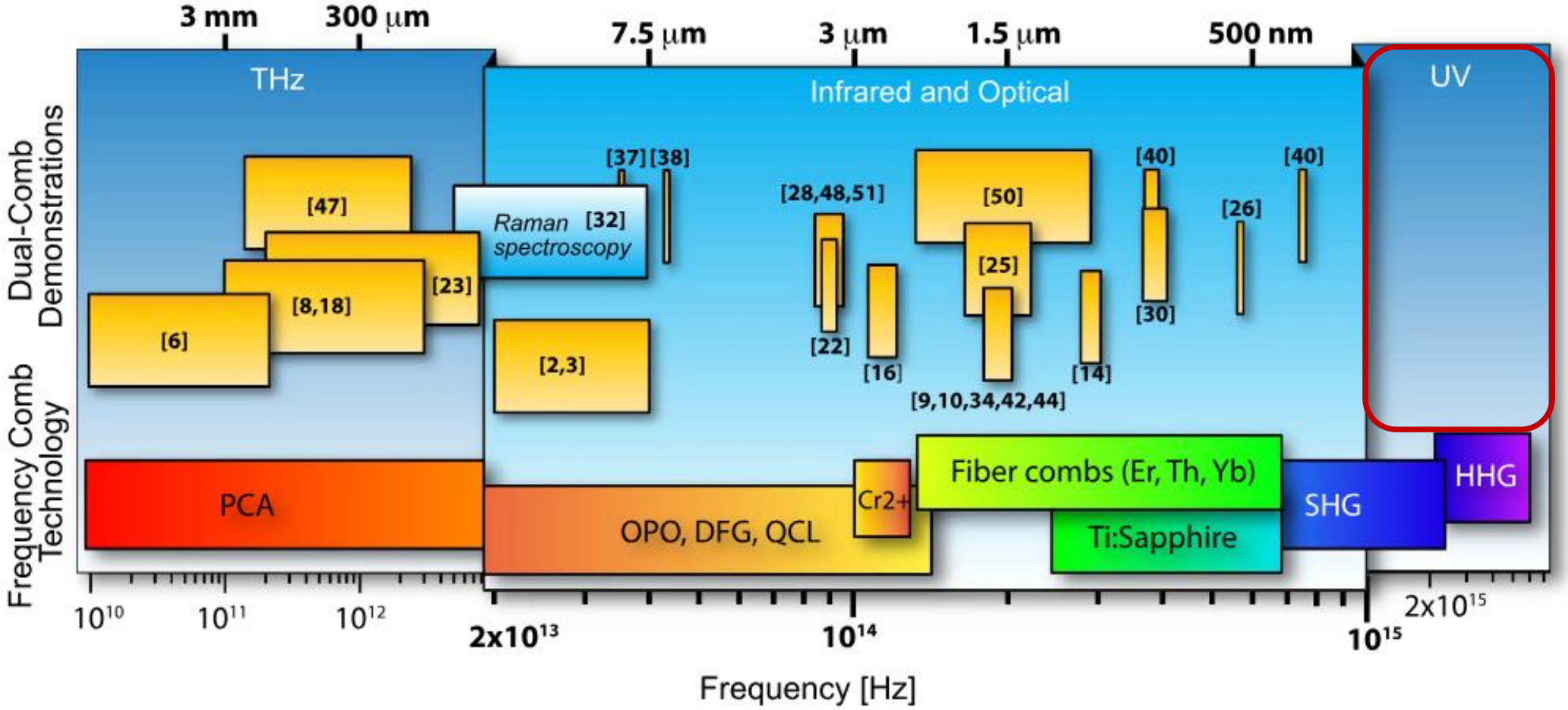
1050 nm → 525 nm **2nd Harmonic (SHG)**

1050 nm → 350 nm **3rd Harmonic (THG)**

1050 nm → 262 nm **4th Harmonic (FHG)**



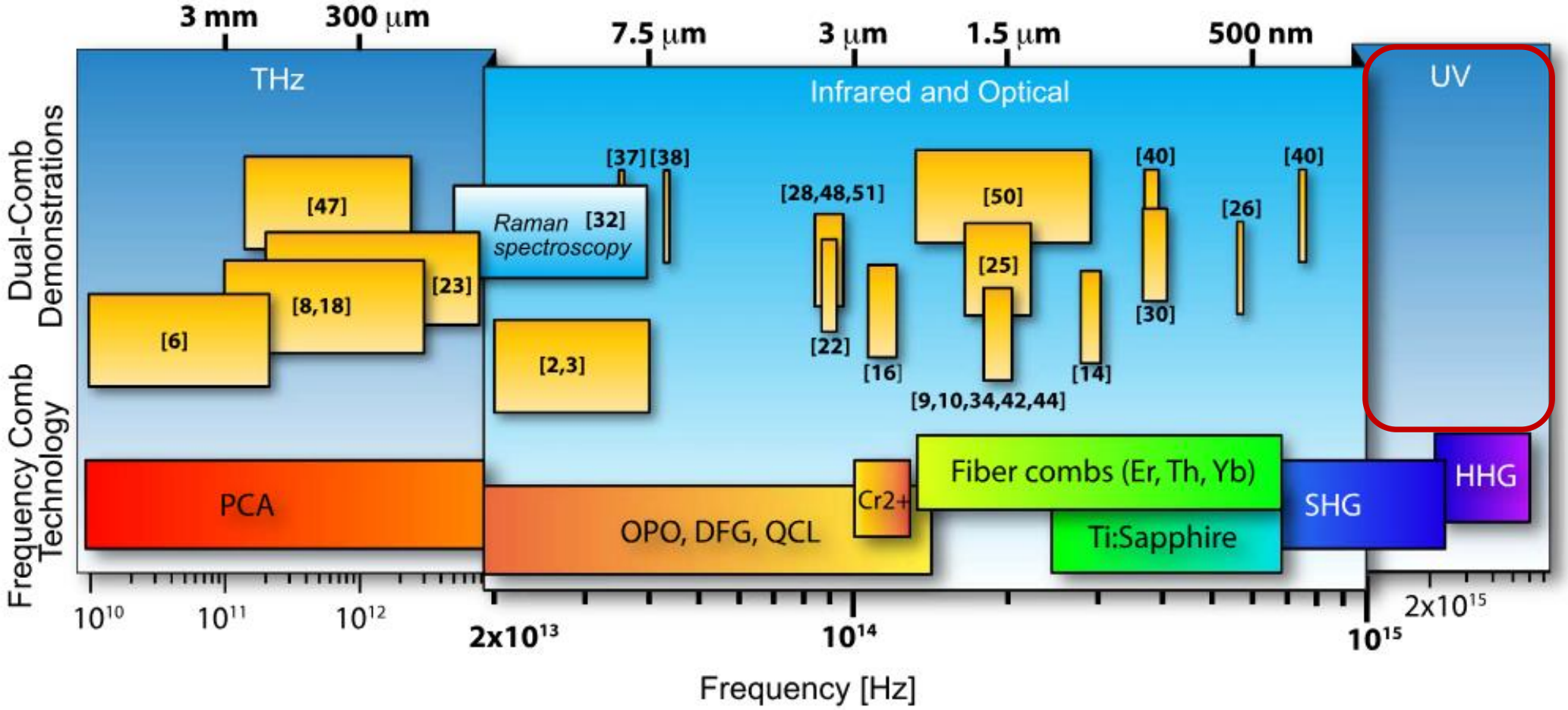
DCS State-of-the-Art



Coddington et al. 2016
DOI:10.1364/OPTICA.3.000414

2016

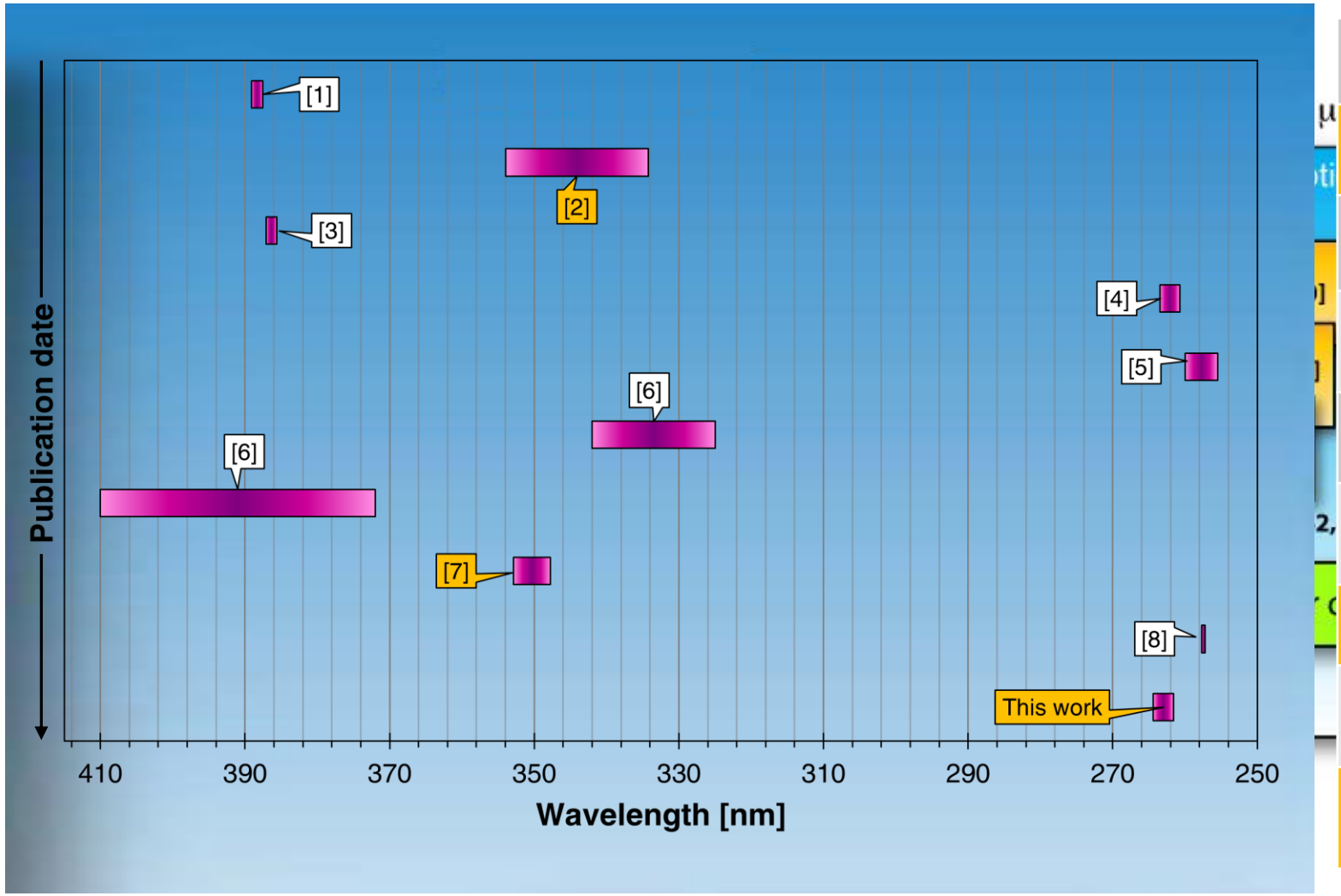
UV DCS State-of-the-Art



Coddington et al. 2016
DOI:10.1364/OPTICA.3.000414

2022

UV DCS State-of-the-Art

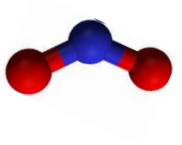


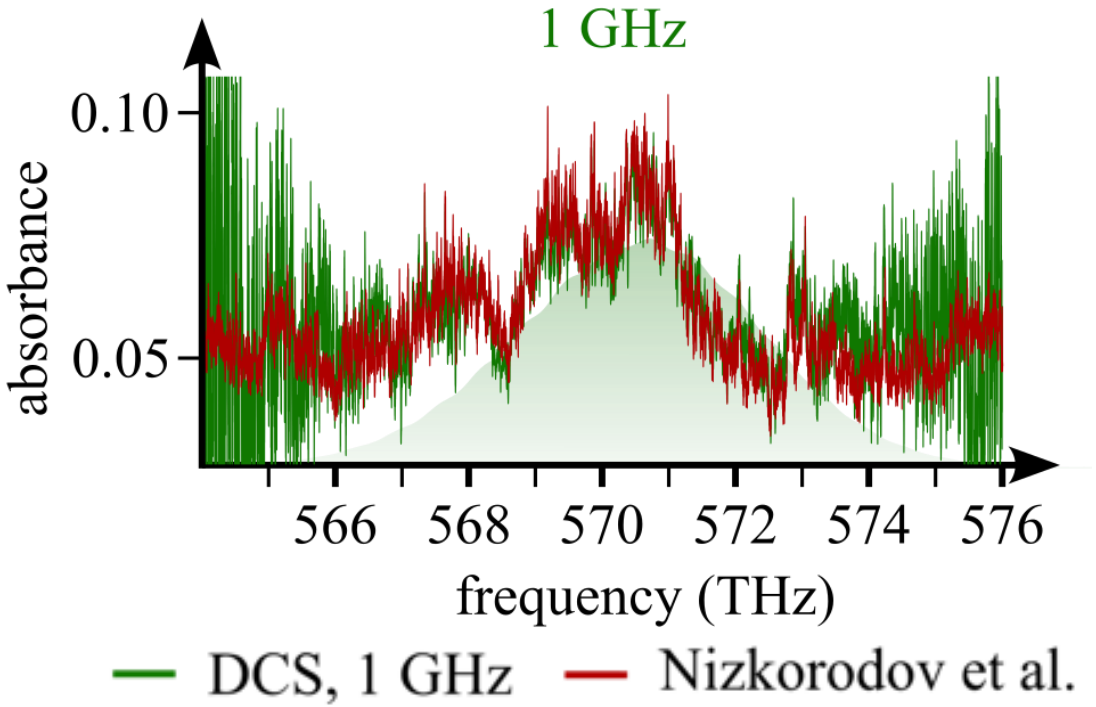
[1]	Xu et al. **2024** DOI: 10.1038/s41586-024-07094-9	**MPQ Garching, GER**
[2]	Fuerst et al. **2024** DOI: 10.1364/OPTICA.516783	**TU Graz, AUT**
[3]	Chang et al. **2024** DOI: 10.1364/OL.515776	**NIST, USA**
[4]	McCauley et al. **2024** DOI: 10.1364/OPTICA.516851	**University of Arizona, USA**
[5]	Li et al. **2025** DOI: 10.34133/ultrafastscience.0087	**Aerospace Information Research Institute, CHN**
[6]	Muraviev et al. **2024** DOI: 10.1364/OPTICA.536971	**University of Central Florida, USA**
[7]	Fuerst et al. **subm.** DOI: 10.48550/arXiv.2501.07350	**TU Graz, AUT**
[8]	Hofer et al. **2025** DOI: 10.1088/1361-6455/add4d7	**HSU Hamburg, GER**
This work	Ehl et al. **in prep**	**TU Graz, AUT**

Coddington et al. 2016
DOI:10.1364/OPTICA.3.000414

2025

VIS (526 nm) – NO_2





Alexander Eber

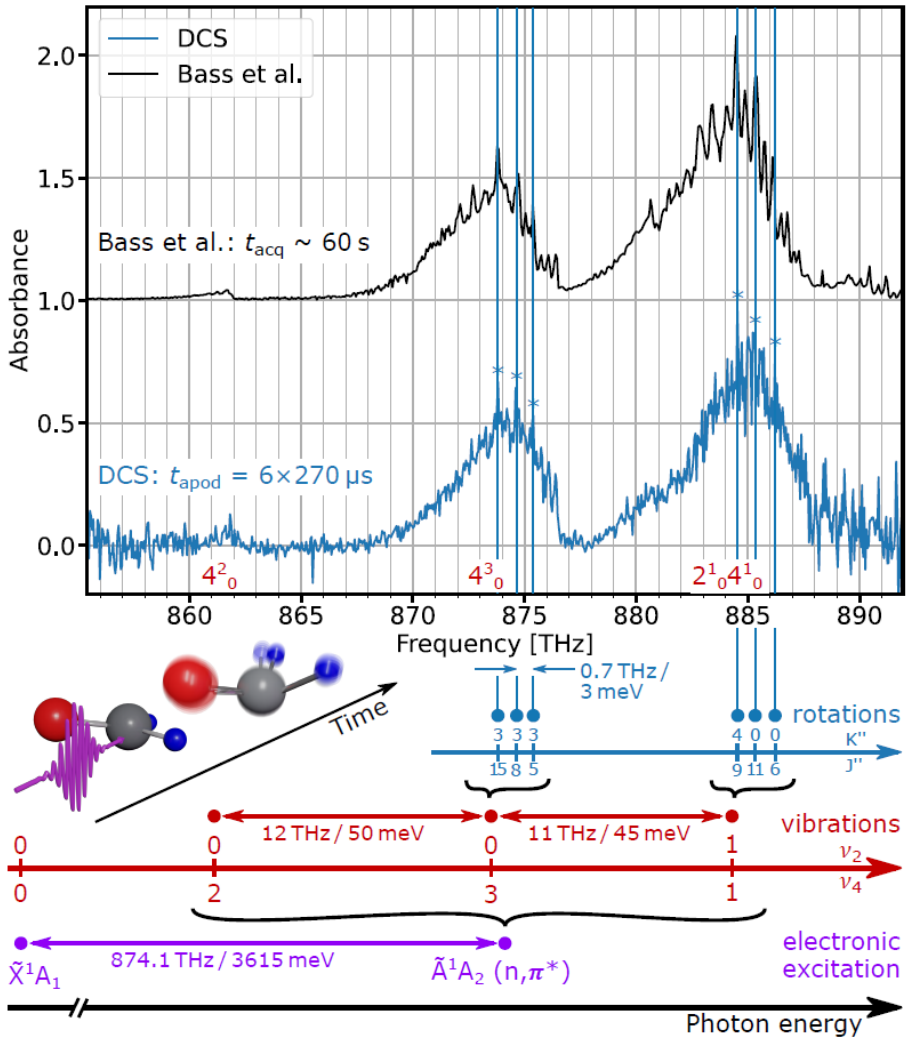
$$\lambda[nm] = \frac{3 \cdot 10^5}{f\ [THz]}$$

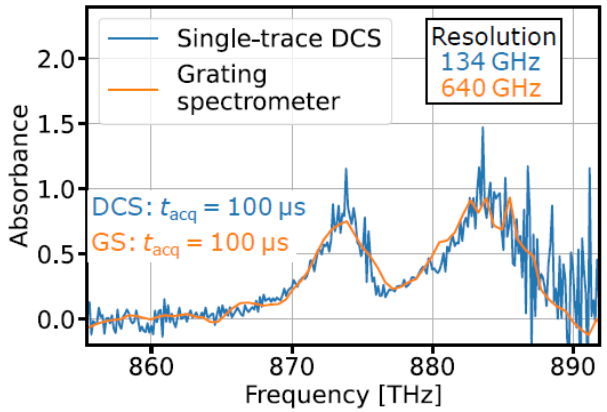
Doppler Width: 1 GHz

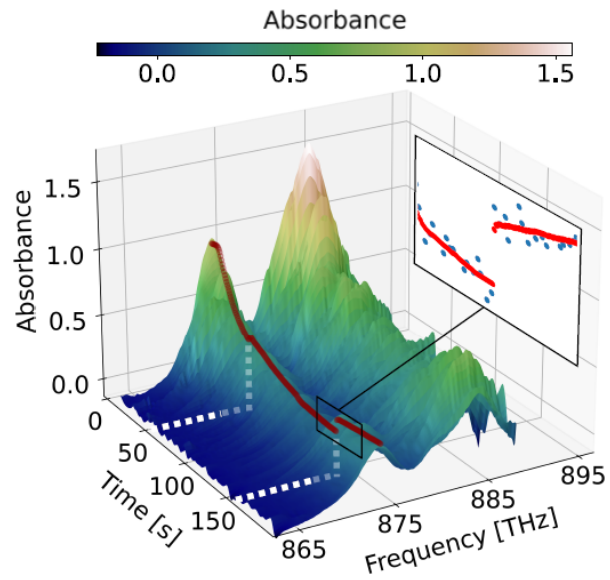
	Resolution	Acquisition Time
DCS (1 GHz)	1000 MHz	1 s
FTS (Nizkorodov et al. 2004)	890 MHz	80 min

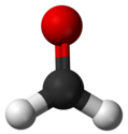
Pal et al. submitted
DOI: 10.48550/arXiv.2412.05972

NUV (350 nm) – CH_2O









Comparable resolution in a fraction of the time

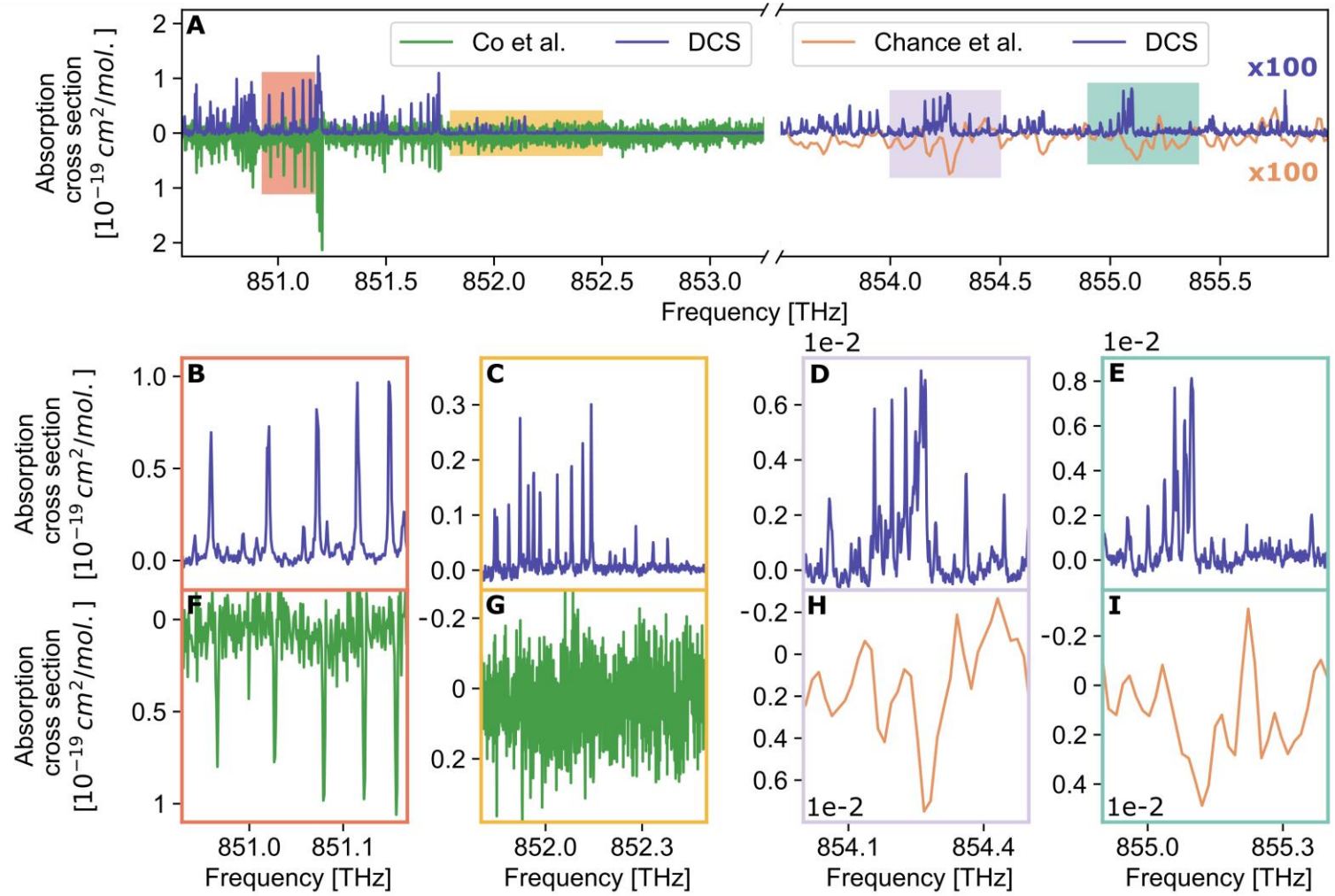
Observed rotations in just 100 µs

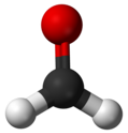
Real-time tracking of concentration with DCS!

Lukas Fürst

Lit: **Bass et al. 1980**, DOI: 10.1016/0032-0633(80)90112-9
Fuerst et al. 2024, DOI:10.1364/OPTICA.516783

NUV (350 nm) – CH_2O





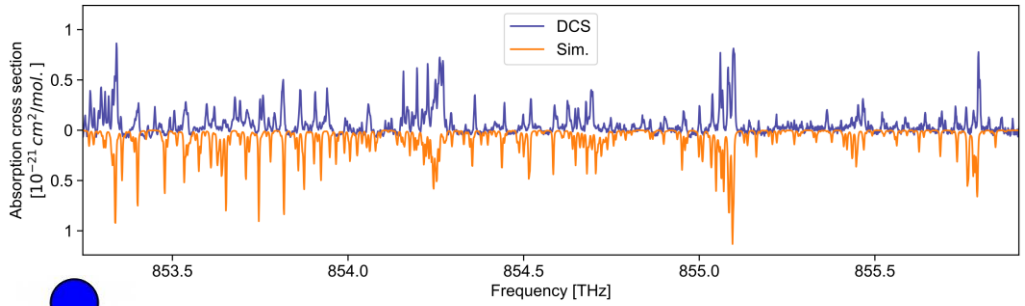
Lukas Fürst

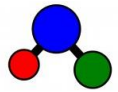
1 GHz resolution in 500 ms
(~ 0.03 cm^{-1})

Resolving power 10^6

>100 prev. unresolved lines

Line assignments & simulation

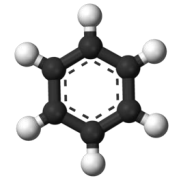


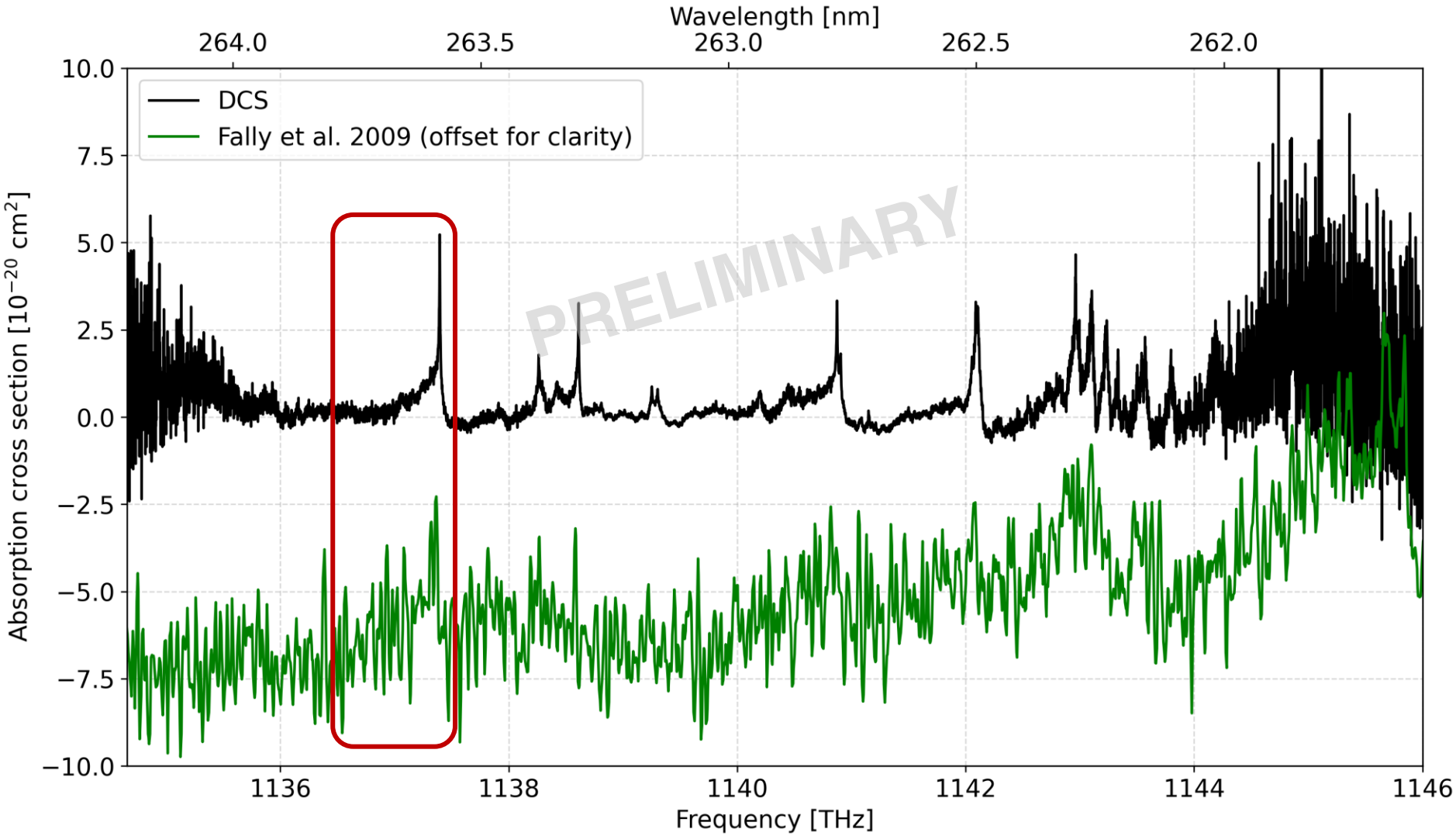


PGOPHER
DOI: 10.1016/j.jqsrt.2016.04.010

Lit: **Co et al. 2005**, DOI: 10.1021/jp053466i
Lit: **Chance et al. 2011**, DOI: 10.1016/j.jqsrt.2011.02.002
Fuerst et al. submitted, DOI: 10.48550/arXiv.2501.07350

DUV (263 nm) – Benzene





Doppler-limited!

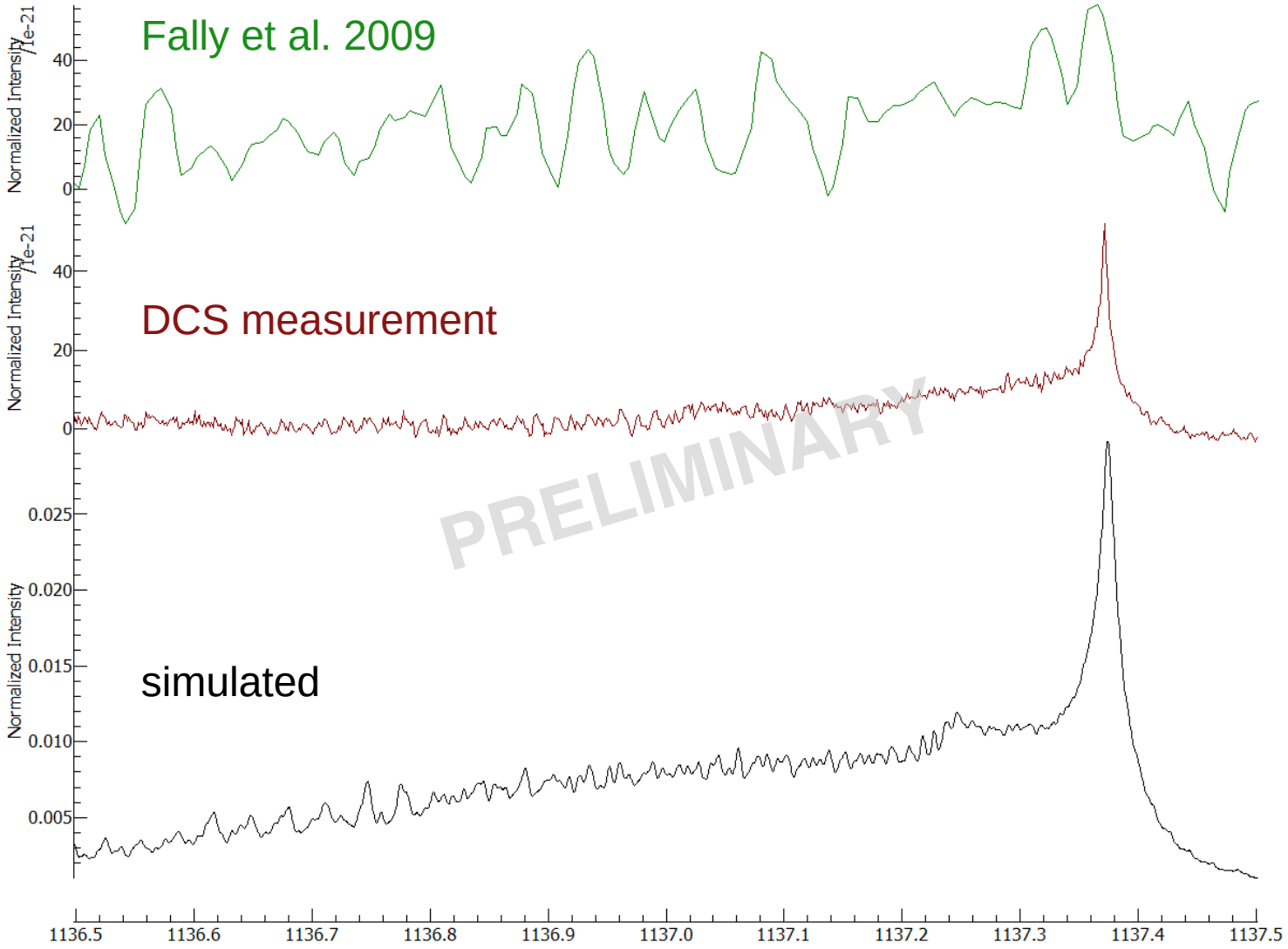
25x higher resolution

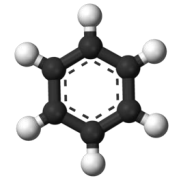
< 10^{-20} cm² sensitivity

Doppler Width:
1.5 GHz

Ehl et al. in prep

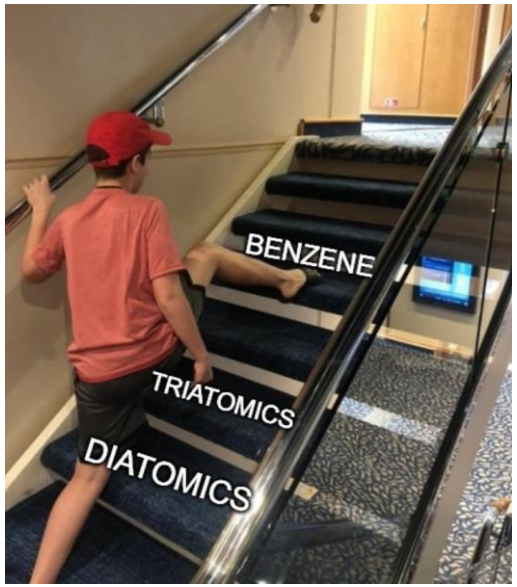
DUV (263 nm) – Benzene





Several unassigned lines

Ab initio calculations, line assignments and modelling in progress

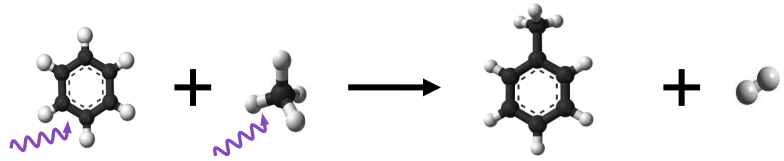


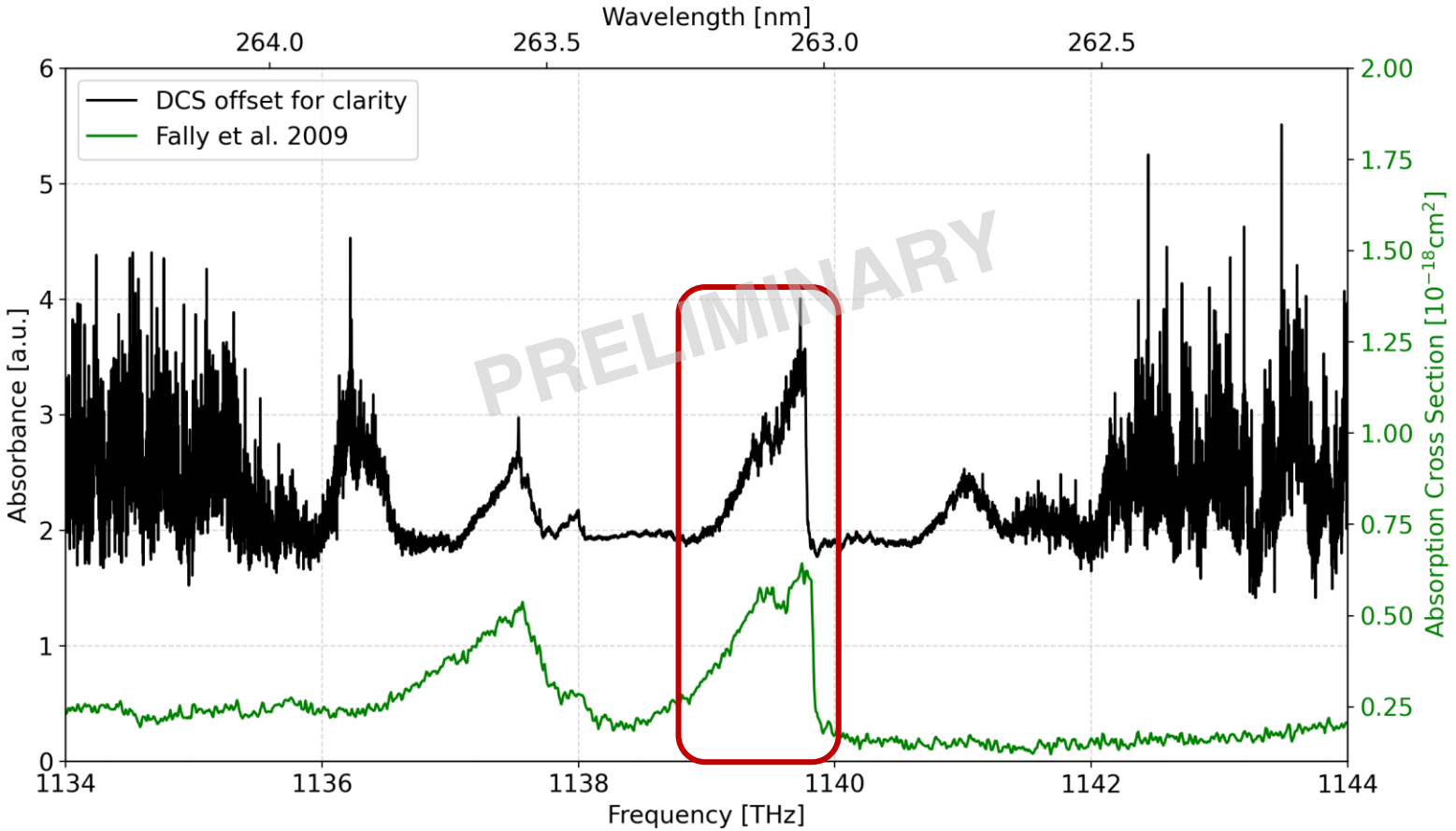
PGOPHER

Ehl et al. in prep

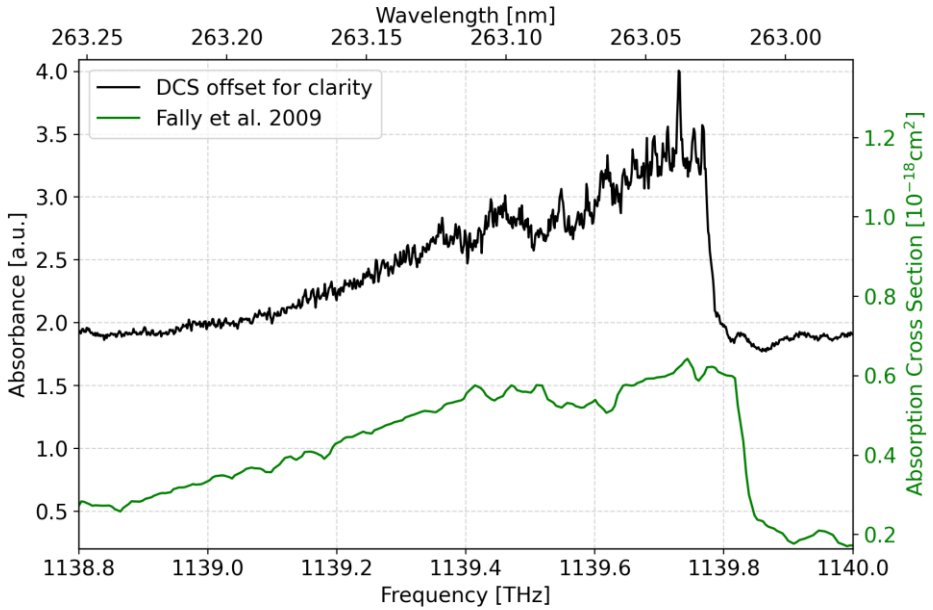
DOI: 10.1016/j.jqsrt.2016.04.010

DUV (263 nm) – Toluene





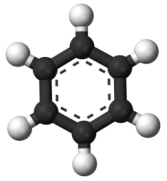
Unresolved structure



Ehl et al. in prep

Outlook

- **Benzene details!**

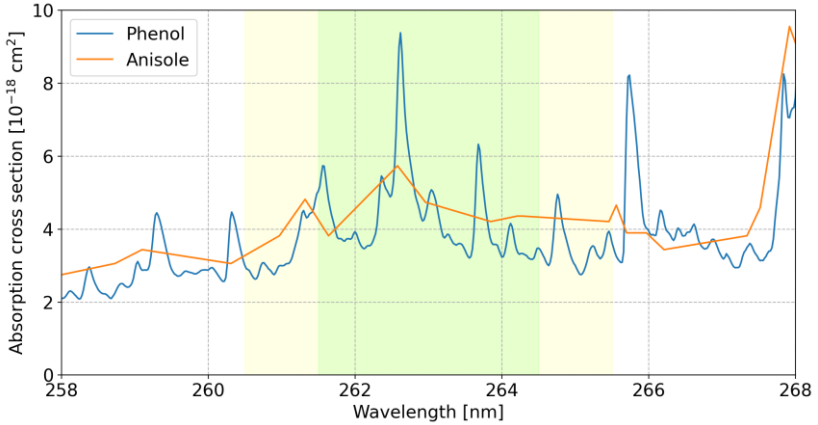


  - Benzene-d6
  - T dependence

- **New samples**

SUGGESTIONS?

  - SO_2
  - Aromatics



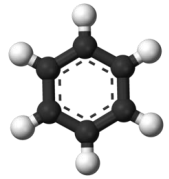
- **HWO & Pollux**

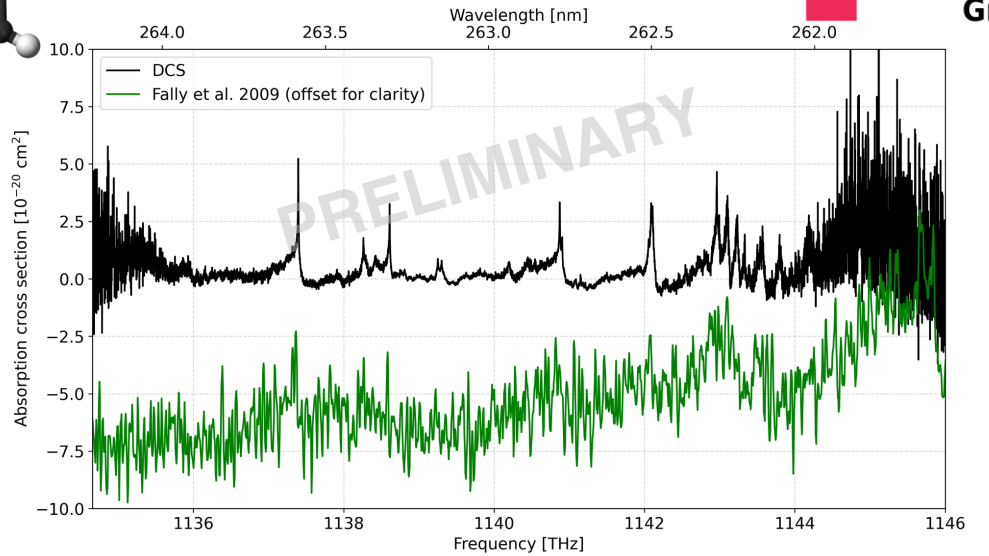
  - Launch 2040
  - Spectropolarimeter
  - 100 – 1900 nm
  - R ~ 100,000

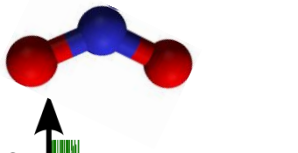
- **Wishlist**
  - SO_2 line list until 40,000 cm^{-1}
  - NO line list until 85,000 cm^{-1}
  - NO_2 & H_2CO
  - Benzene?

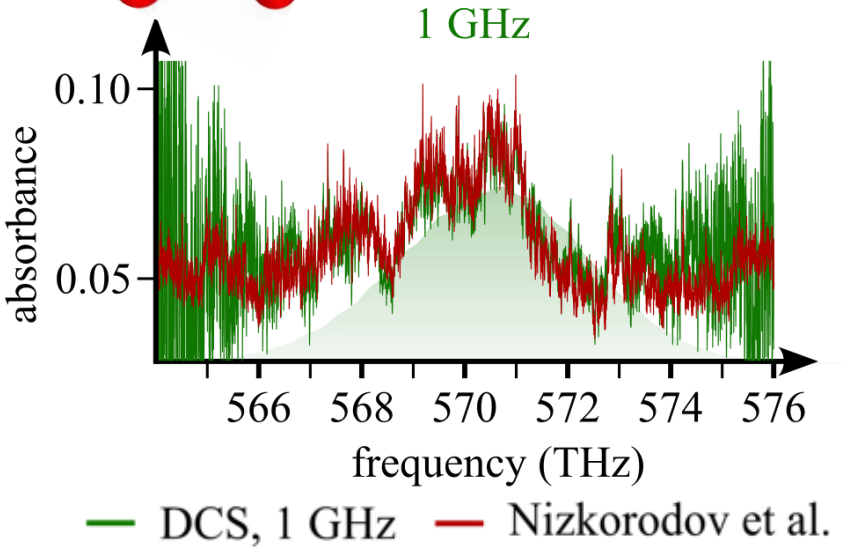
Summary

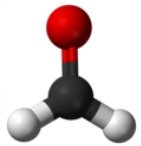
- High spectral resolution (~MHz – GHz)
- Resolving power > 10^6
- Fast acquisition times (~ms – s)

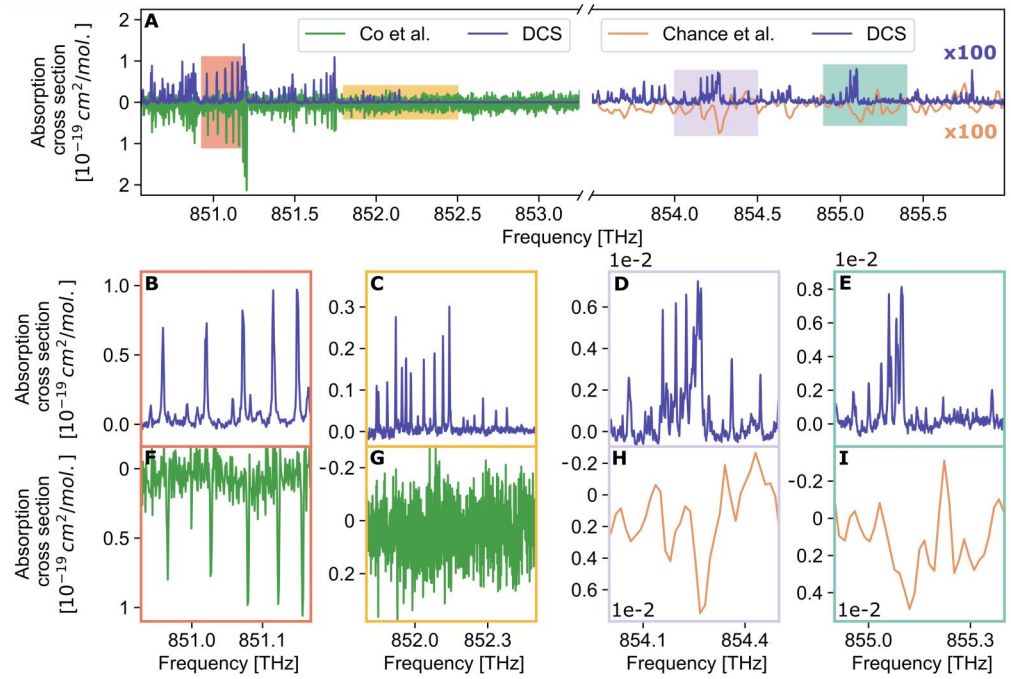




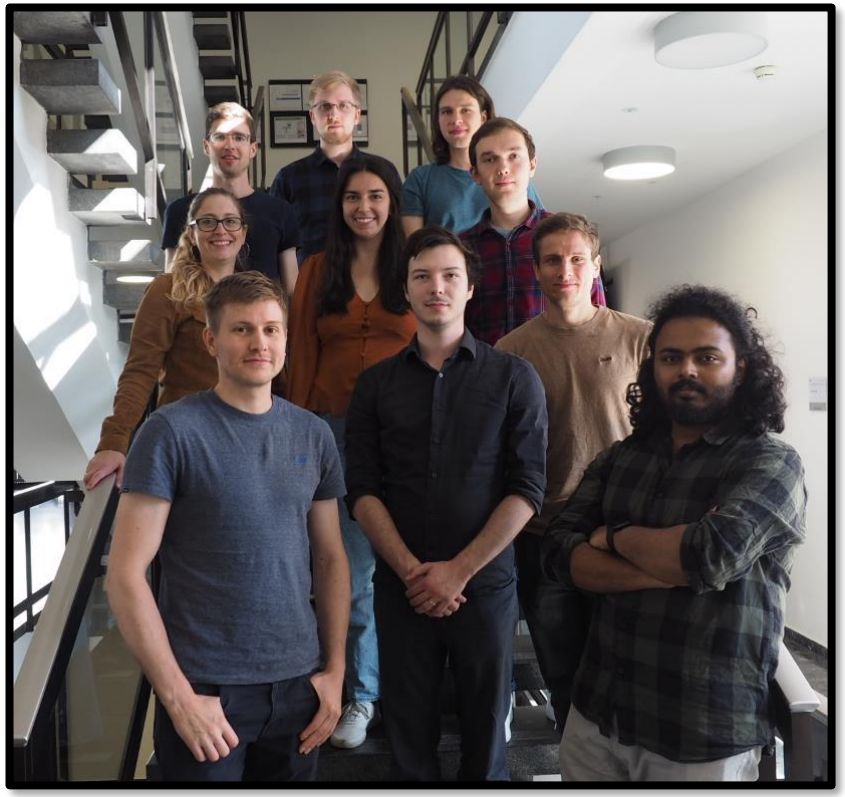








Acknowledgements



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Extra Slides

Setup Sketch