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Mid-infrared frequency-agile dual-comb spectroscopy

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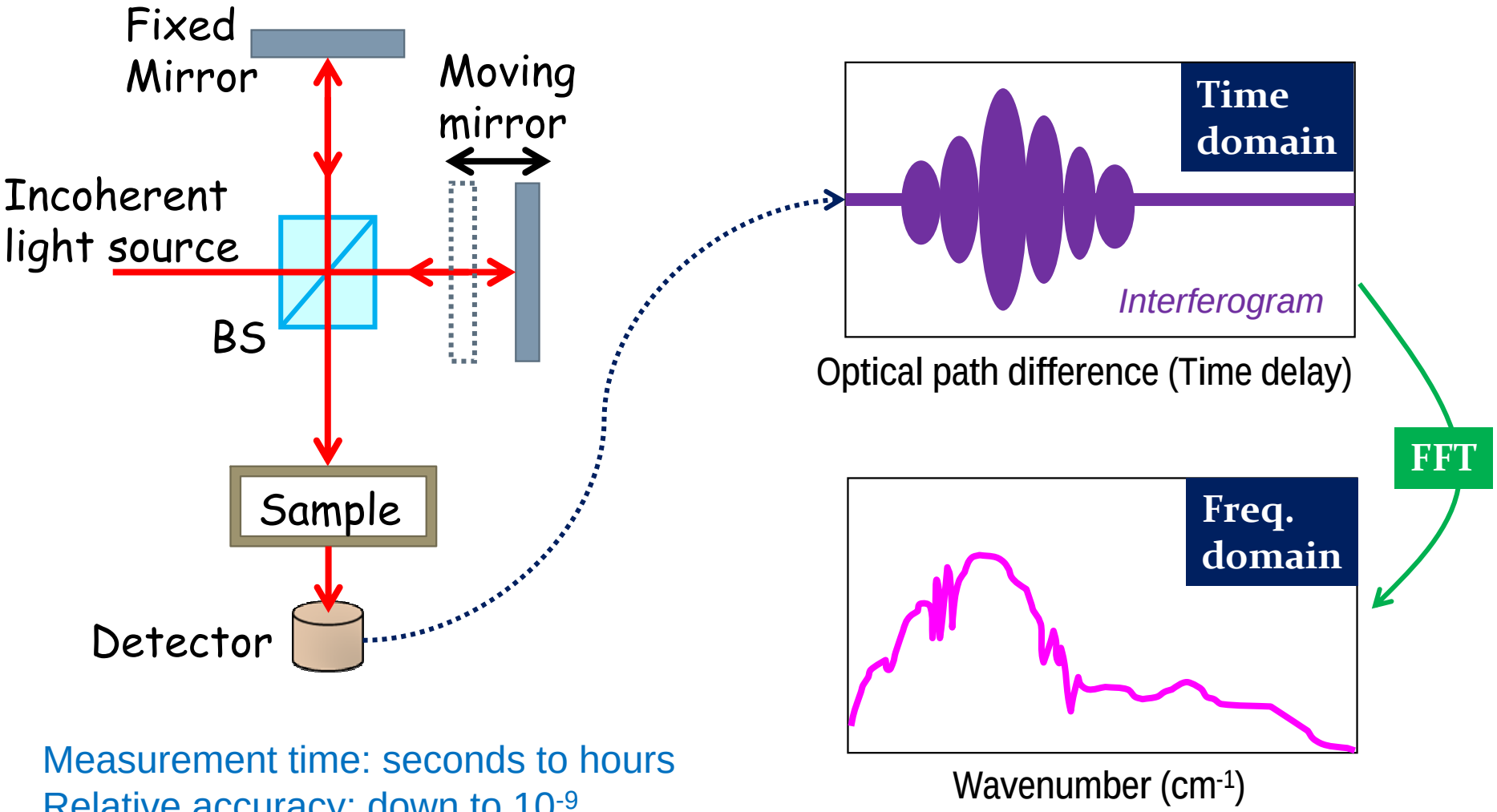
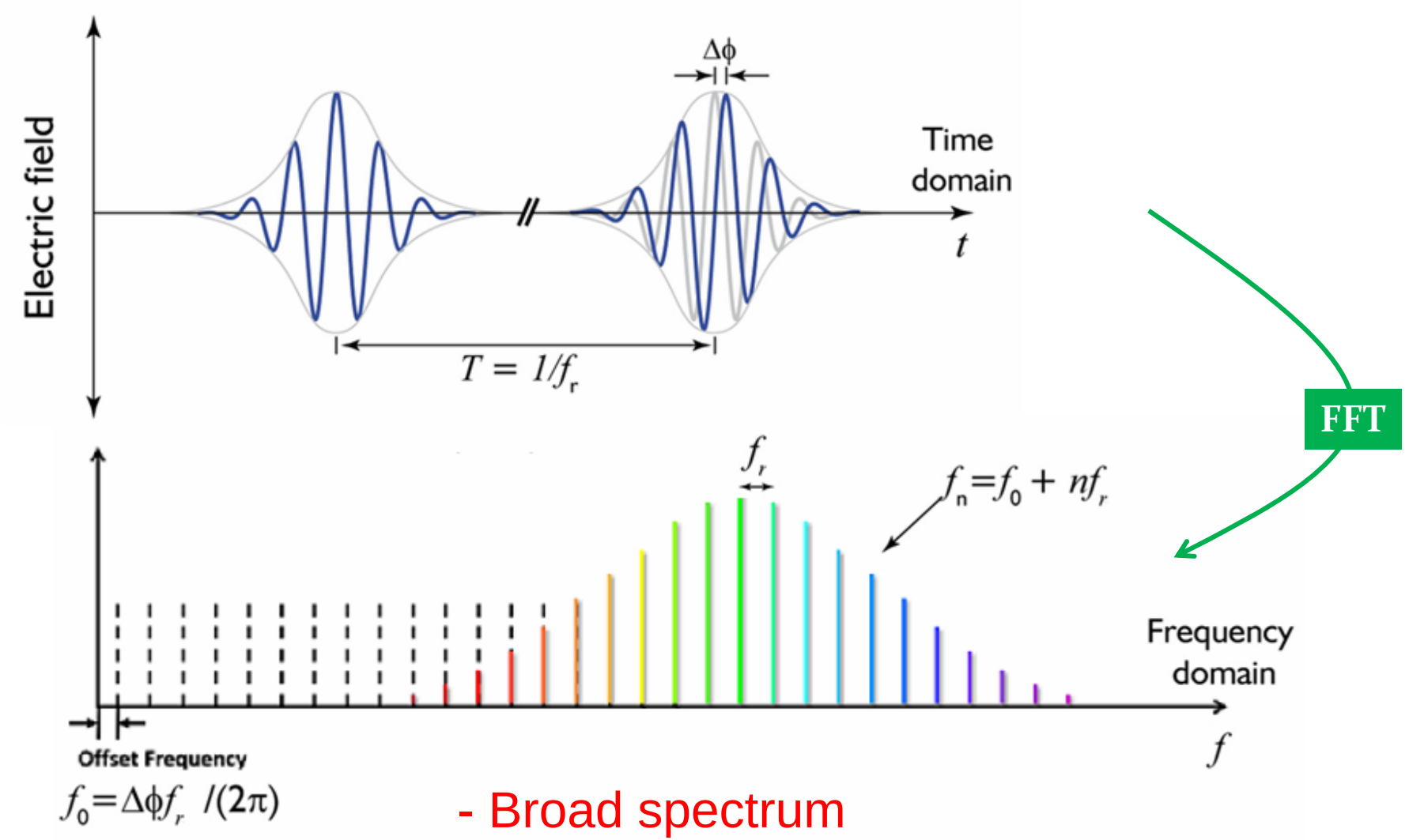
Most widely used spectrometer in mid-infrared

Fourier transform spectrometer (FTS)



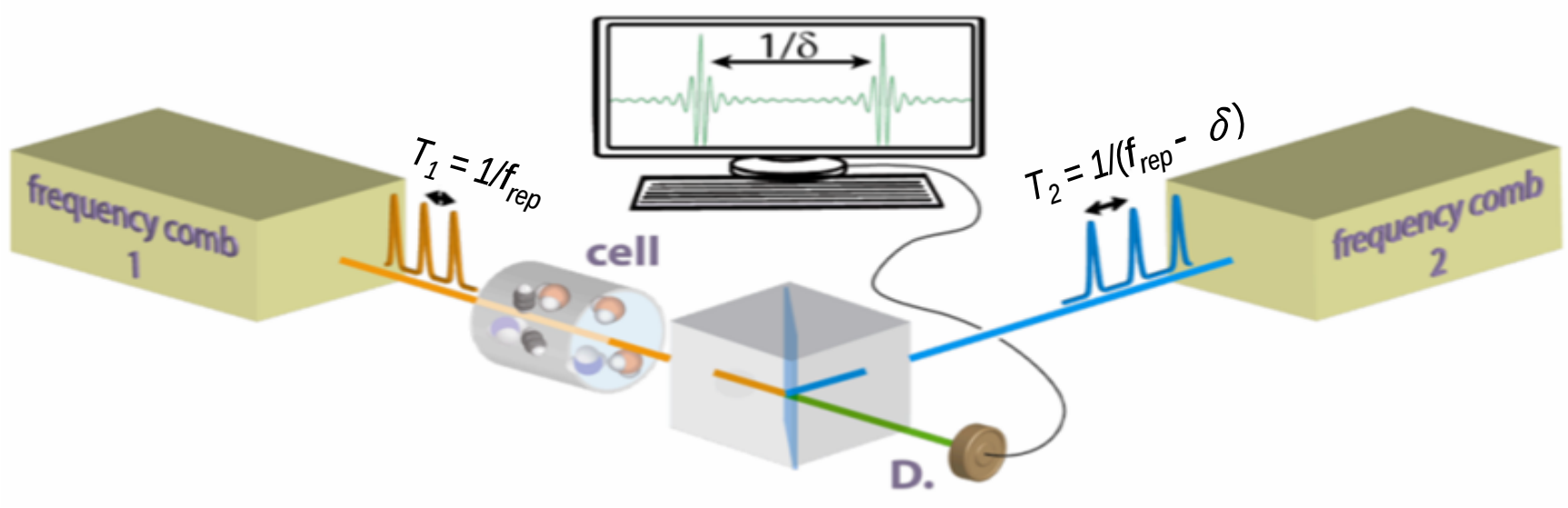
Measurement time: seconds to hours
Relative accuracy: down to 10^{-9}
Resolution: down to 0.001 cm^{-1}

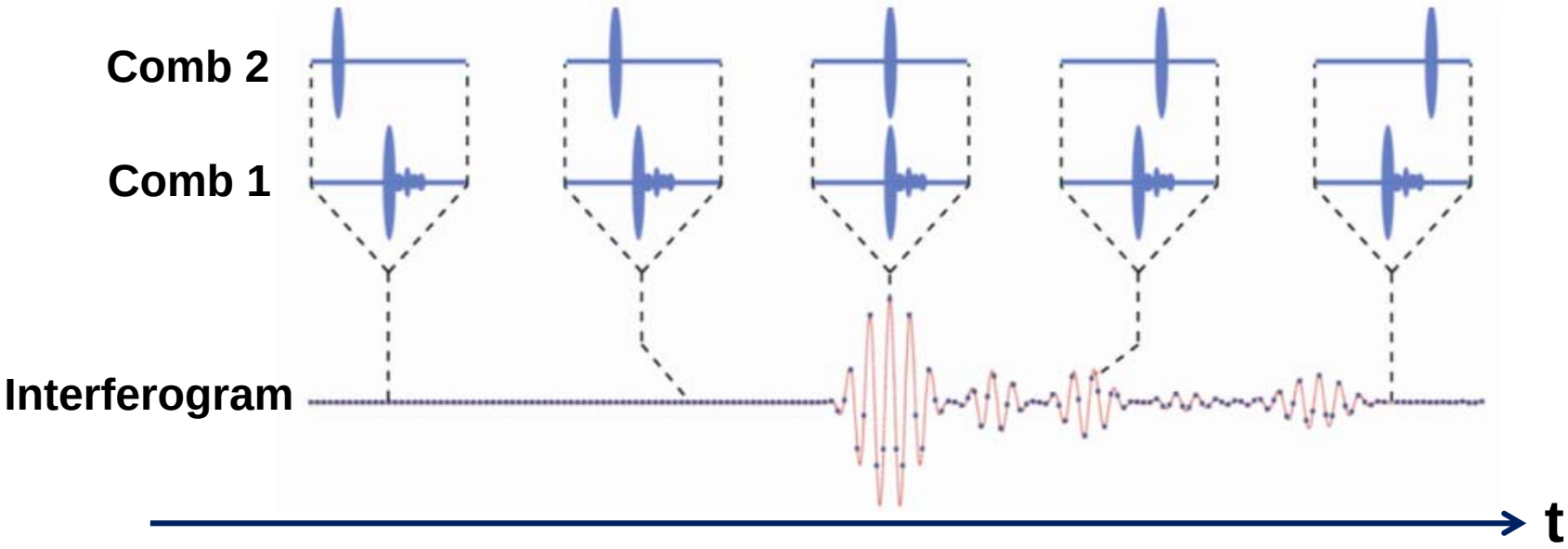
Optical Frequency combs



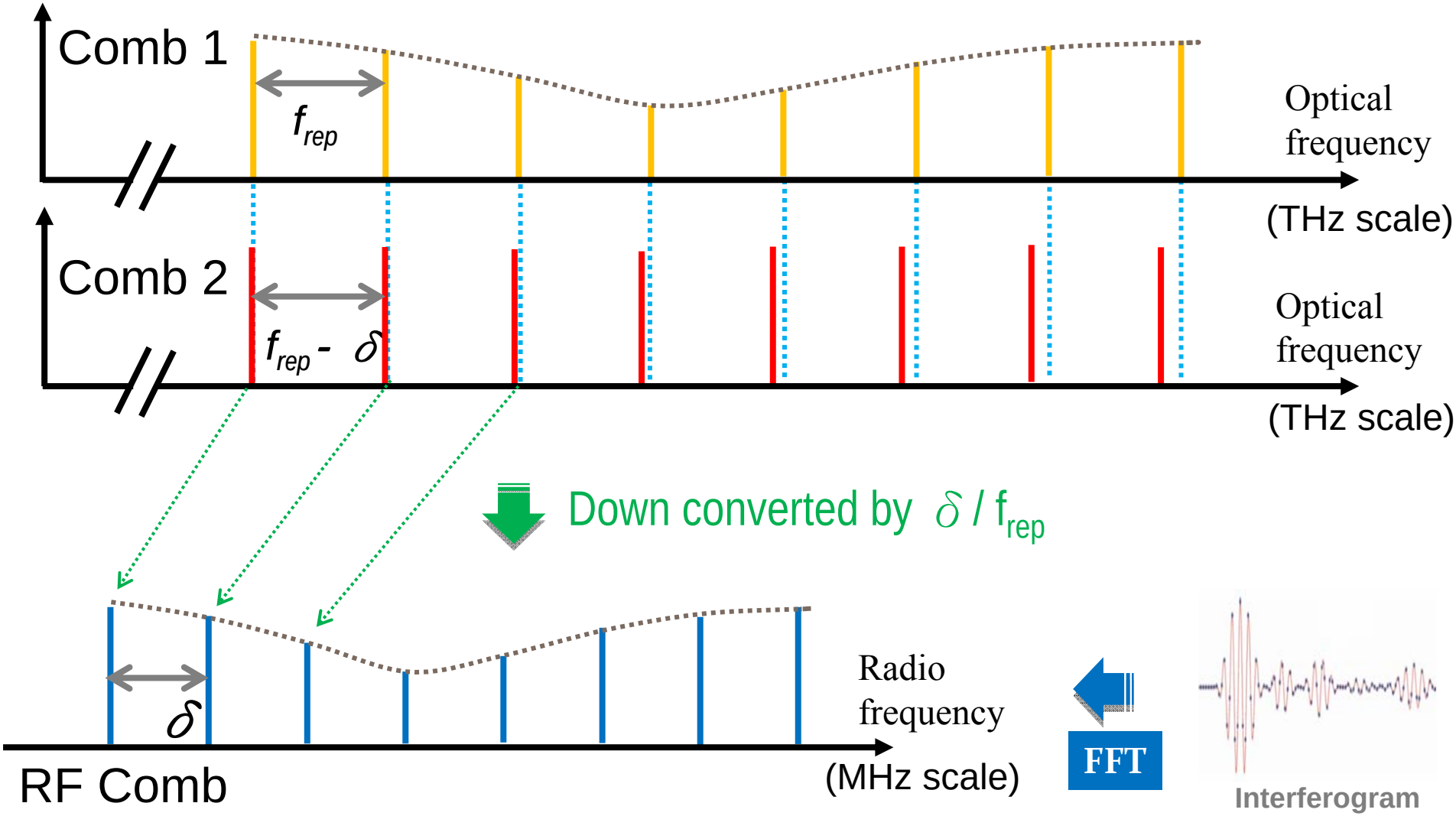
- Broad spectrum
- Many narrow comb lines

Principle of dual-comb spectroscopy in time domain





Principle of dual-comb spectroscopy in frequency domain



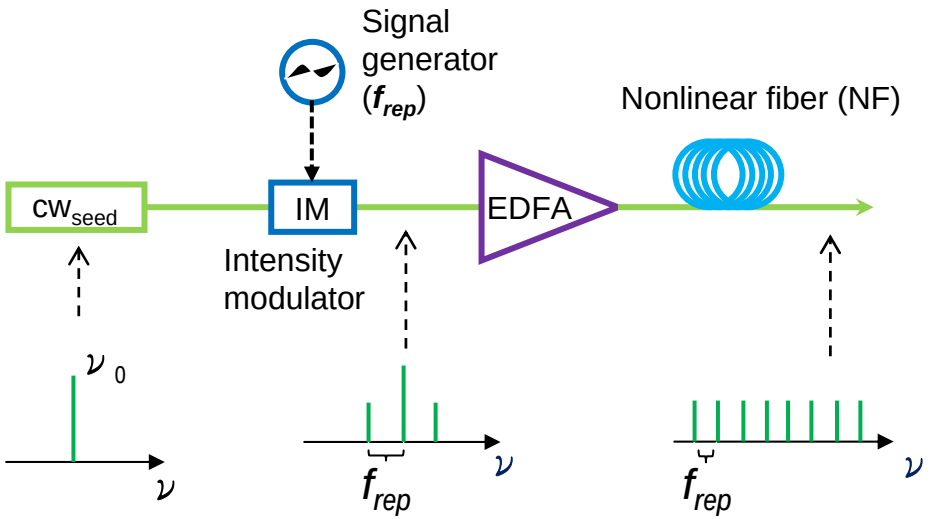
Dual-comb spectroscopy

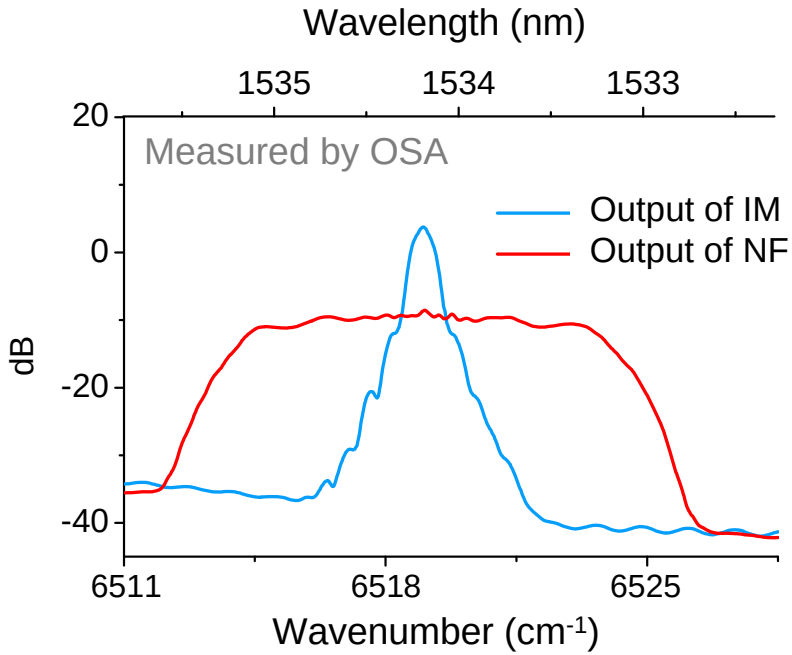
New approach to MIR dual-comb spectroscopy

Without mode-locked lasers and phase-lock electronics

NIR Frequency-agile laser combs + Difference frequency generation to MIR + Multi-heterodyne detection

Frequency-agile laser combs in NIR





☑ Simplicity without mode-locked lasers

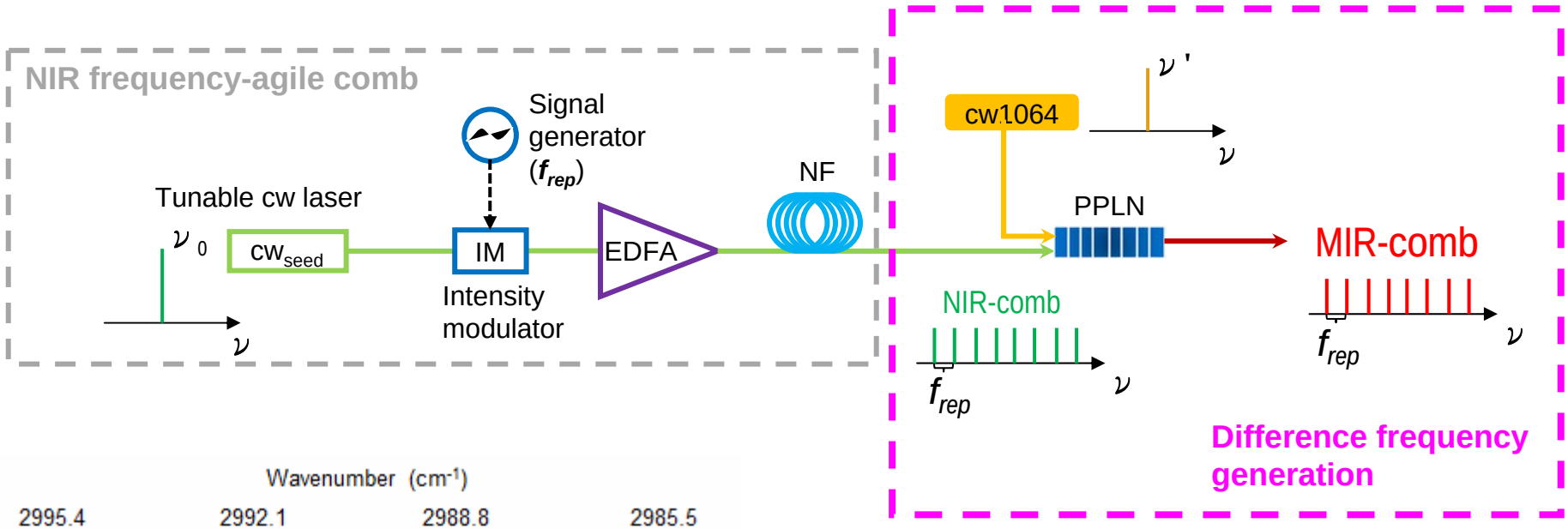
☑ Flat-top spectrum with up to 2000 comb lines (within 3 dB)

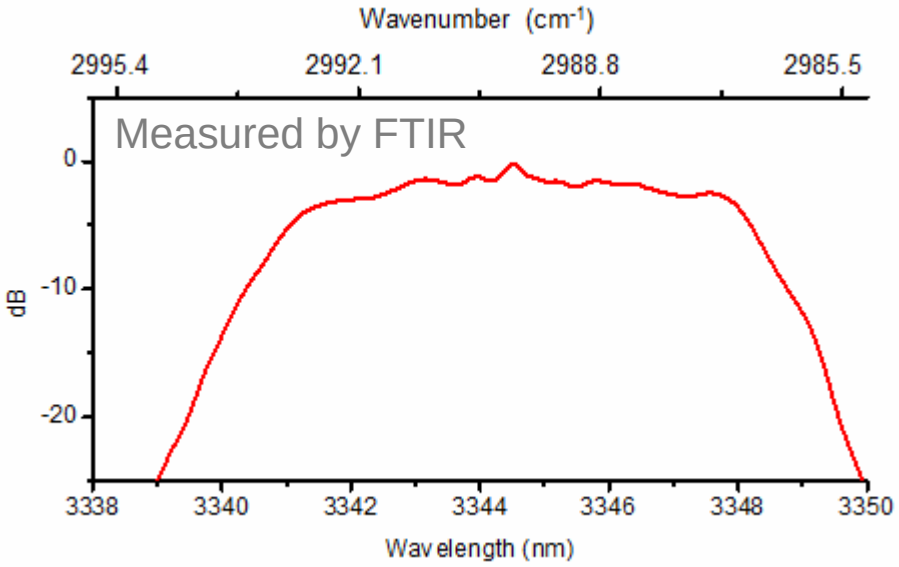
☑ Widely & simply tunable comb line spacing (100 MHz ~ 1000 MHz)

(3.3×10^{-3} cm^{-1} ~ 3.3×10^{-2} cm^{-1})

Nat. Photonics, 10, 27 (2016)

Mid-infrared Frequency-agile laser combs

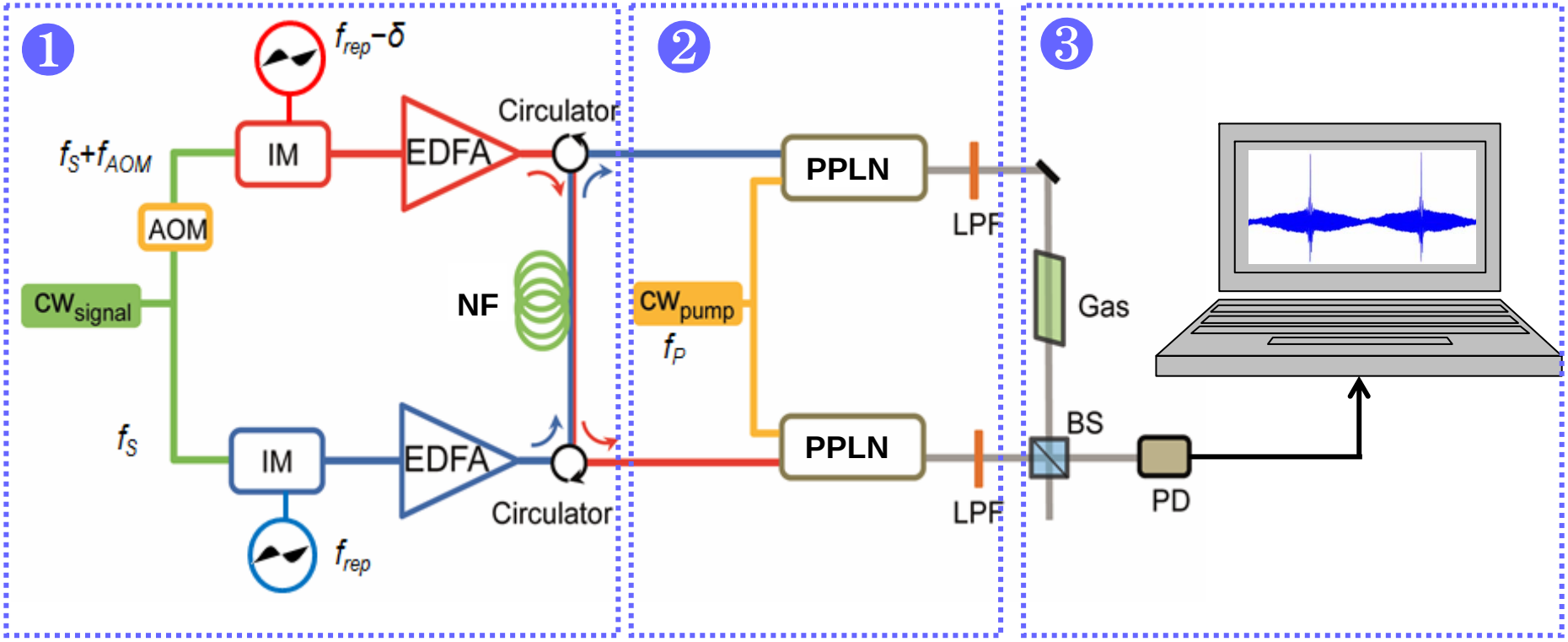




Optical resolution: 6 GHz (0.2 cm^{-1})

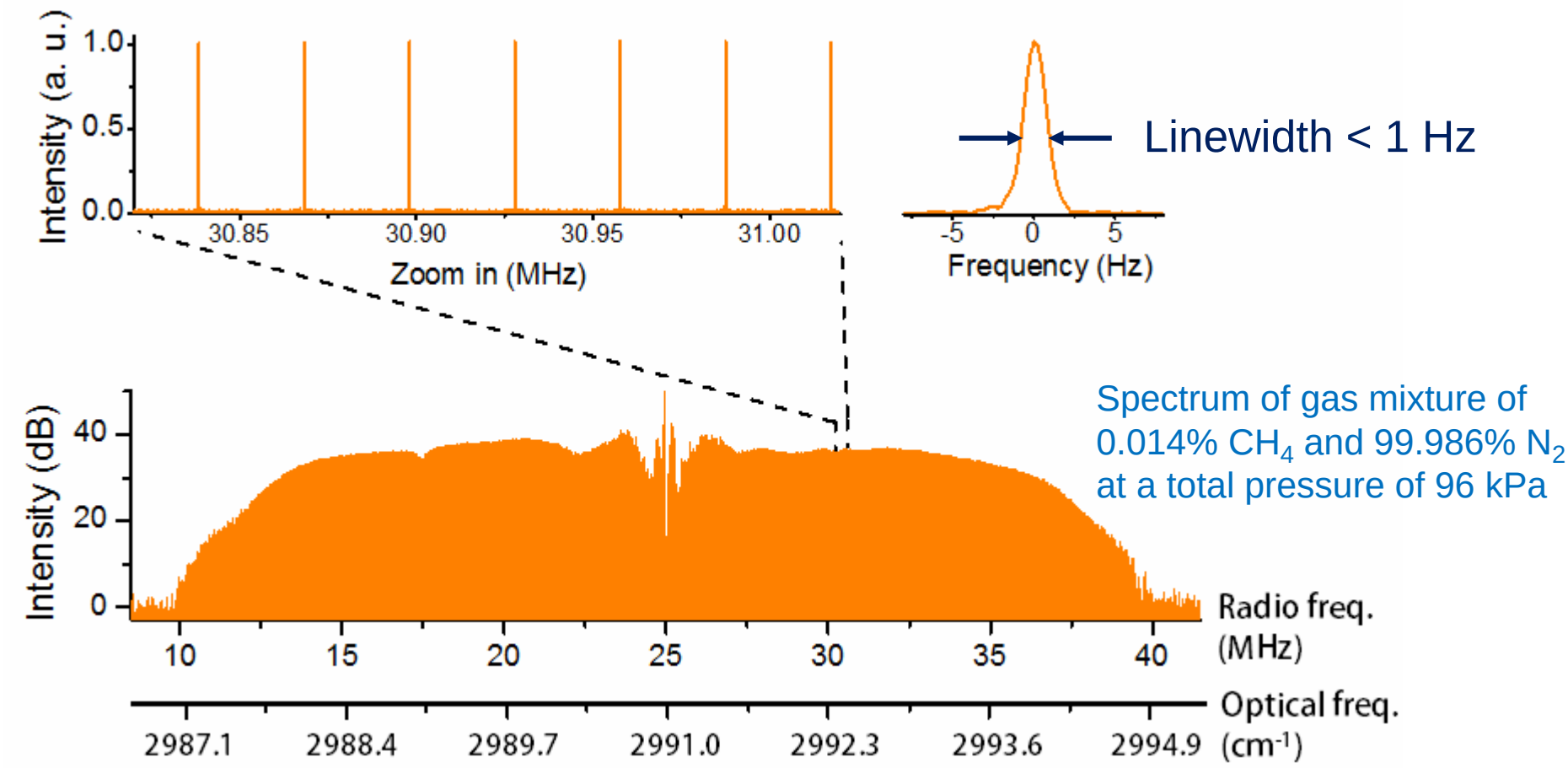
- ☑ Average power: > 500 μW
- ☑ Spectral bandwidth: 7~10 cm^{-1}
- ☑ Wavelength tunable: 2800~3200 cm^{-1} (3100~3500 nm)

MIR Frequency-agile dual-comb spectroscopy



1. Generation of two NIR frequency combs with flat-top spectral profiles
2. MIR frequency comb generation by the difference frequency generation
3. Dual-comb Fourier transform spectroscopy

MIR Frequency-agile dual-comb spectroscopy



Measurement time: 1.1 sec; Optical resolution: 230 MHz (~7.7×10^{-3} cm^{-1})

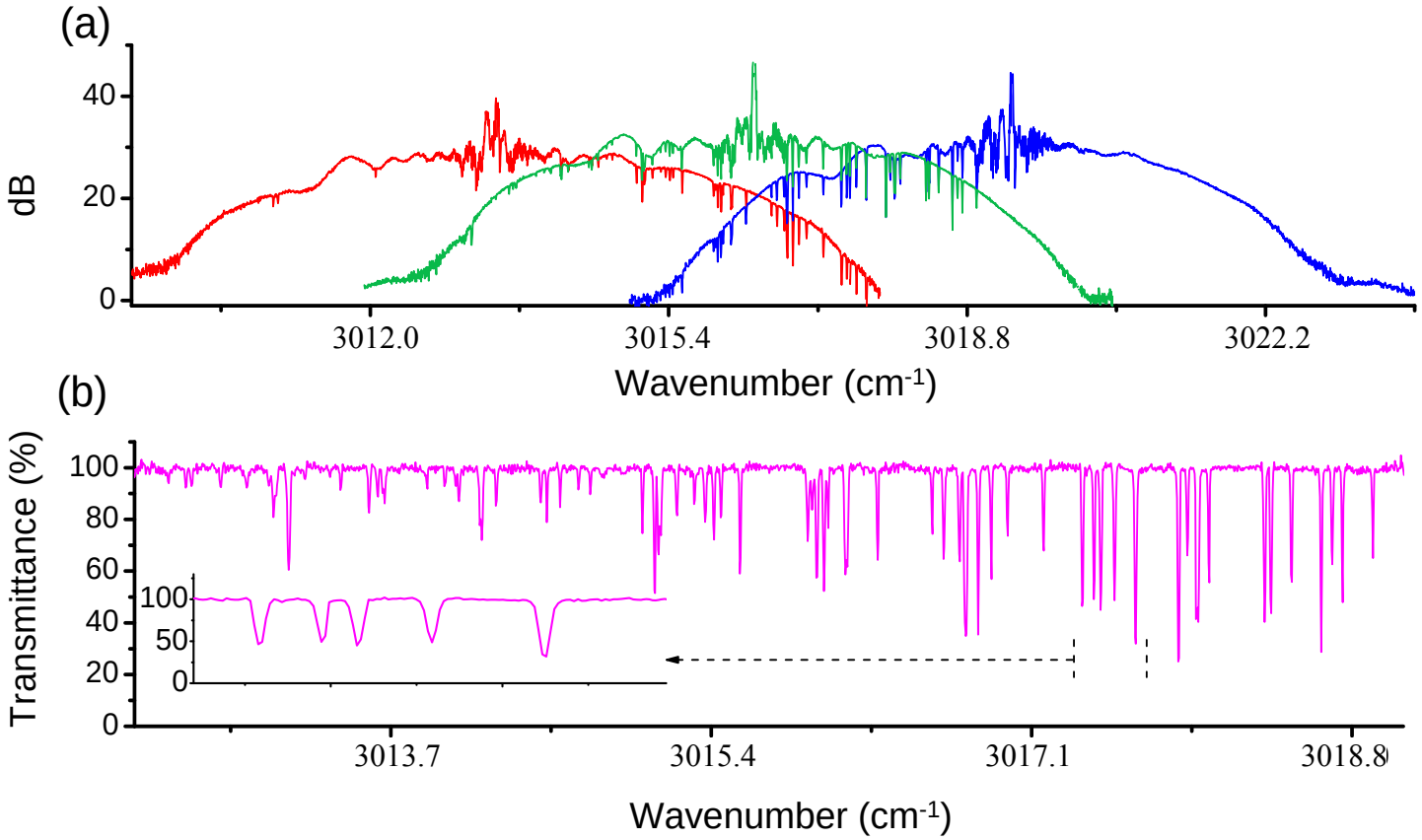
Rules for RF domain → Optical frequency domain

Center peak → f (CW_{pump}) − f (CW_{signal})

Comb line spacing in RF domain × f_{rep}/δ → Optical frequency domain

Mid-infrared dual-comb spectra with frequency agility

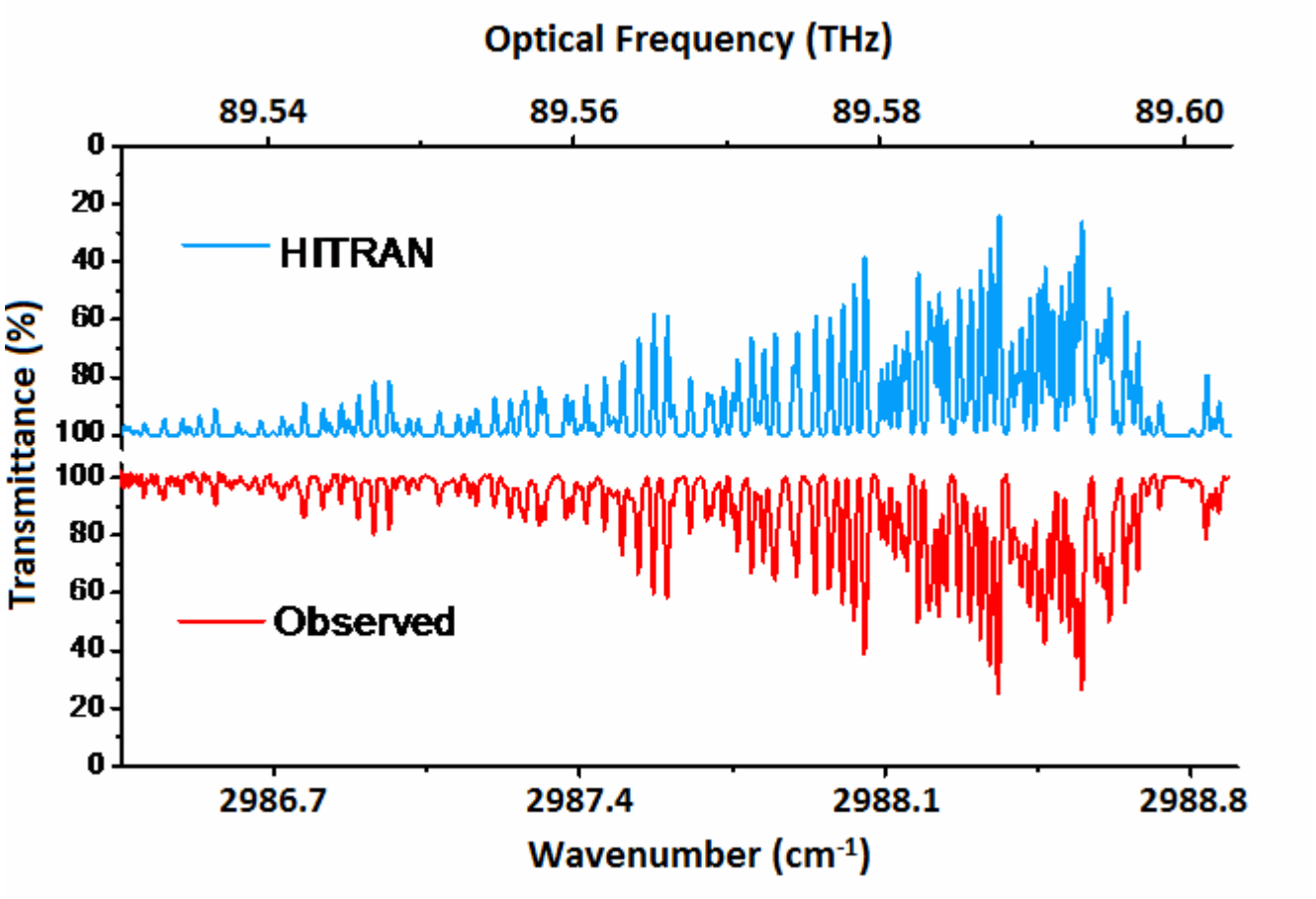
Q-branch of methane gas (ν_3 band)



- Measurement time for each spectrum: 72 ms (728 μs × 100)
- Resolution: 115 MHz (~5×10^{-3} cm^{-1})

Mid-infrared dual-comb spectrum with Doppler-limited resolution

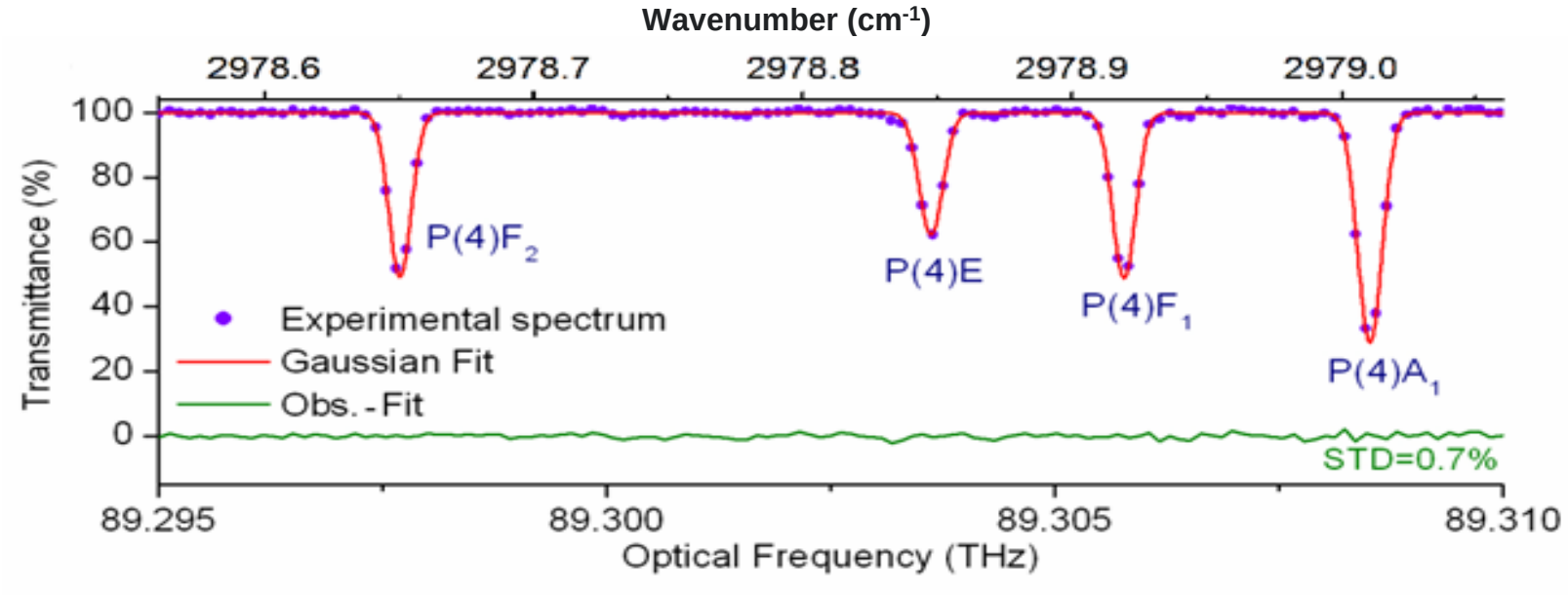
Q-branch of ethylene gas (ν_{11} band)



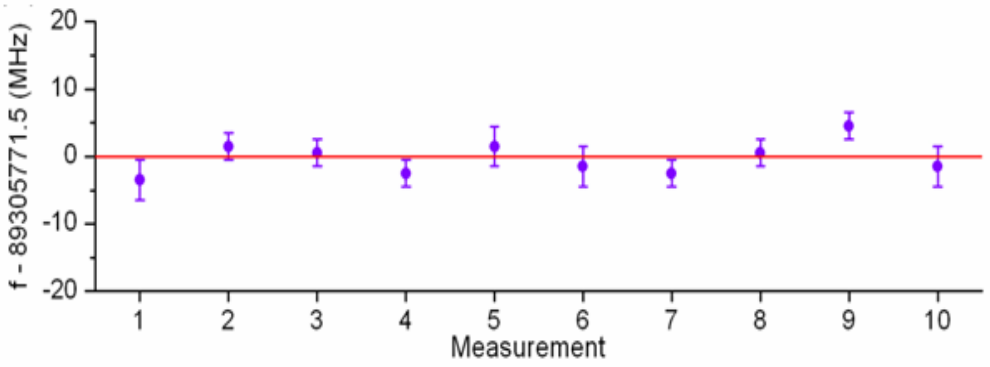
- Total measurement time: 72 ms (728 μs × 100)
- Resolution: 100 MHz (~3.3×10^{-3} cm^{-1})

Precision measurements of line position and intensity

The v_3 band P(4) lines of CH_4



- Measurement time: 72 ms
- Resolution: 115 MHz (~5×10^{-3} cm^{-1})



10 measurements of the $P(4)F_1$ line position

☑ Line position measurements for Doppler-limited spectra

→ Relative accuracy of 10^{-8}

(a few MHz uncertainty)

☑ Line intensity measurements

→ 1~3% uncertainty

Precision measurements of line position and intensity

Line positions

Line	This work (MHz)	Ref. 1 (MHz)	This work – Ref. 1 (MHz)
P(4) F_2	89297694.9(1.2)	89297695.2635(21)	-0.4
P(4) E	89303617.9(1.3)	89303620.0492(25)	-2.1
P(4) F_1	89305771.5 (1.1)	89305771.6093(21)	-0.1
P(4) A_1	89308513.3(1.1)	89308512.2568(20)	1.0

Ref. 1: Absolute frequency list of the ν_3-band transitions of methane at a relative uncertainty level of 10^{-11}, Opt. Express 19, 23878-23888 (2011)

Line intensities

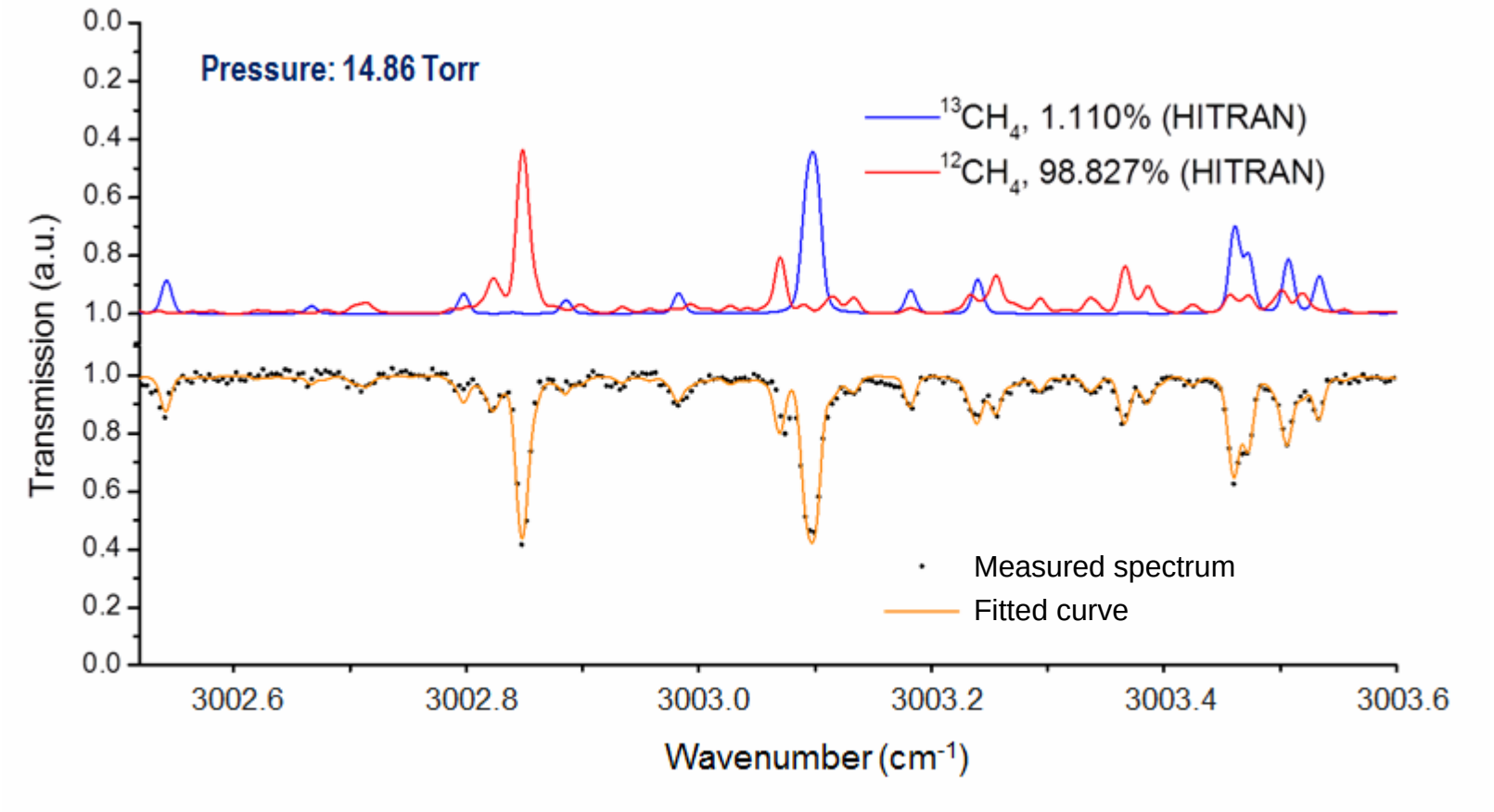
Line	This work* ($cm^{-1}/mol*cm^{-2}$)	Ref. 2 ($cm^{-1}/mol*cm^{-2}$)	Difference (%)
P(4) F_2	$7.134(133)\times10^{-20}$	$7.310(147)\times10^{-20}$	-2.41
P(4) E	$4.796(132)\times10^{-20}$	$4.887(98)\times10^{-20}$	-1.86
P(4) F_1	$7.245(132)\times10^{-20}$	$7.339(147)\times10^{-20}$	-1.28
P(4) A_1	$1.2060(196)\times10^{-19}$	$1.215(25)\times10^{-19}$	-1.71

* The intensities here have been corrected for the purity of methane (99.95%) and the isotopic abundance (98.827% for $^{12}CH_4$). The uncertainty includes 1% pressure error, 1% cell length error, and 1 K temperature error.

Ref. 2: N_2 and Ar broadening and line mixing in the *P* and *R* branches of the v_3 band of CH_4, J. Quant. Spectrosc. Radiat. Transf. 57, 157–176 (1997)

Accurate Concentration measurements of isotopes ($^{12}C/^{13}C$) in methane

☑ Concentration measurements → ~2% uncertainty



- Total measurement time: 72 ms (728 μs × 100)
- Resolution: 115 MHz (~5×10^{-3} cm^{-1})

Summary

- **Mid-IR frequency-agile comb**
  - ✓ Without mode-locked lasers & phase-lock electronics
  - ✓ Flat-top spectral profile with 1000 comb lines
  - ✓ Widely tunable comb line-spacing & center wavelength

- **Mid-IR frequency-agile dual-comb spectroscopy**
  - ✓ Dual-comb Fourier transform spectrometer
    - Broadband measurement available
    - Resolved comb line without instrumental lineshape
  - ✓ Precision measurements of Doppler-limited dual-comb spectra in tens of ms measurement time
    - Line position measurement → relative accuracy of 10^{-8}
    - Line intensity measurement → 1~3% uncertainty
  - ✓ Accurate concentration measurements of ($^{12}C/^{13}C$) in methane

Outlook

- ✓ Turnkey system for environment tracing
- ✓ Real-time gas sensing (gas concentration & composition)