Cavity-Enhanced Optical Frequency Comb Spectroscopy

Aleksandra Foltynowicz
Department of Physics, Umeå University, Sweden

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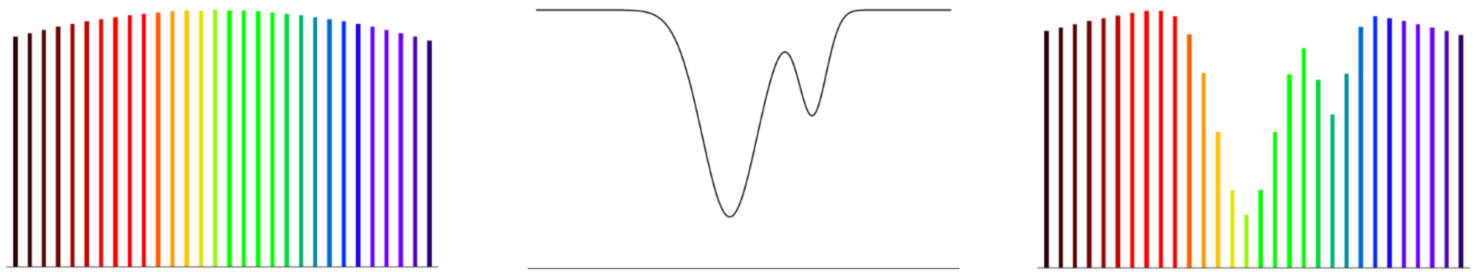
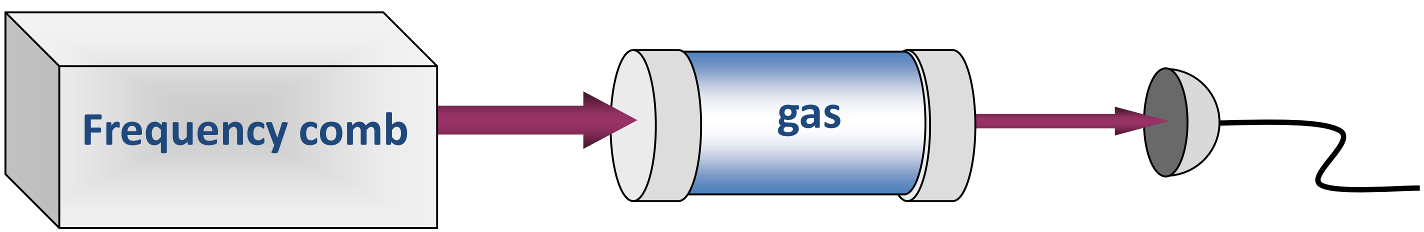
Cavity-Enhanced Optical Frequency Comb Spectroscopy

Comb modes | Molecular absorption | Transmitted spectrum



Acquisition of entire absorption bands
Multispecies detection

High sensitivity
Short acquisition times



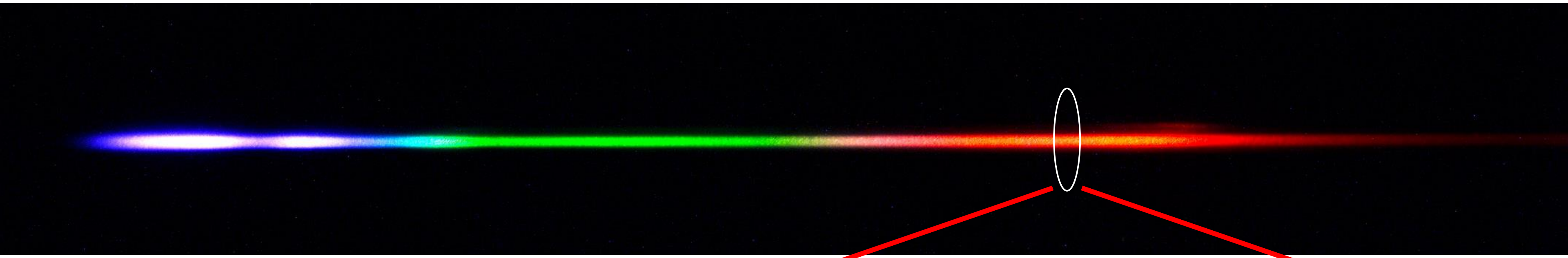
Mode-locked laser
- broad bandwidth
- high resolution
- efficient coupling to cavity

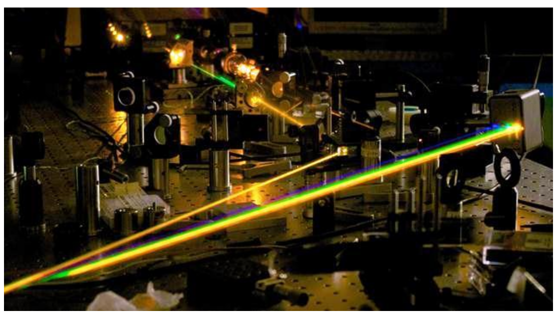
High-finesse optical cavity
- high sensitivity to absorption

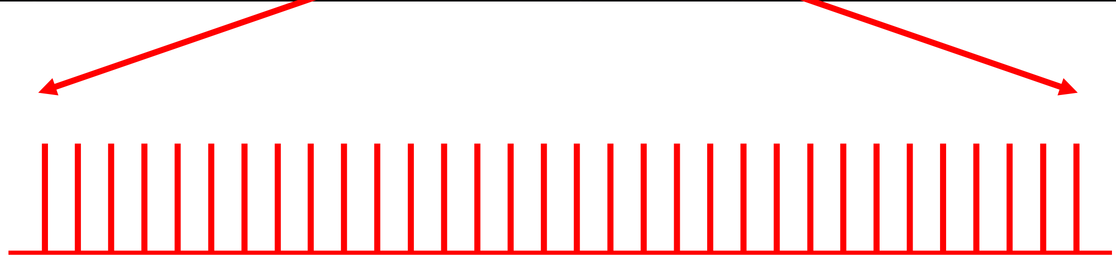
Broadband detection
- rapid data acquisition
- broad bandwidth

Todd Johnson and Scott Diddams

Optical Frequency Comb







Thousands of synchronized laser lines!!

Nobel Prize 2005

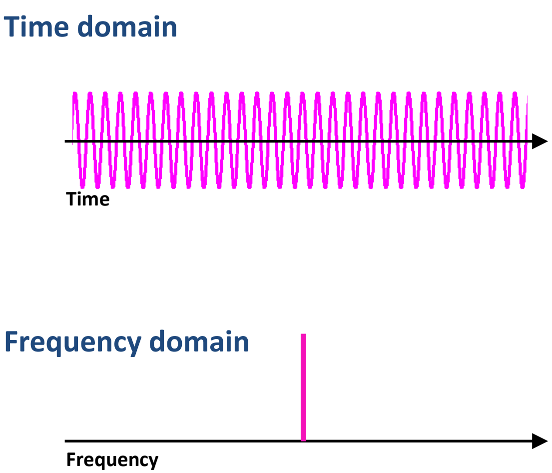
John Hall and Theodor Hänsch

‘for their contributions to the development of laser-based precision spectroscopy, including the optical frequency comb technique’

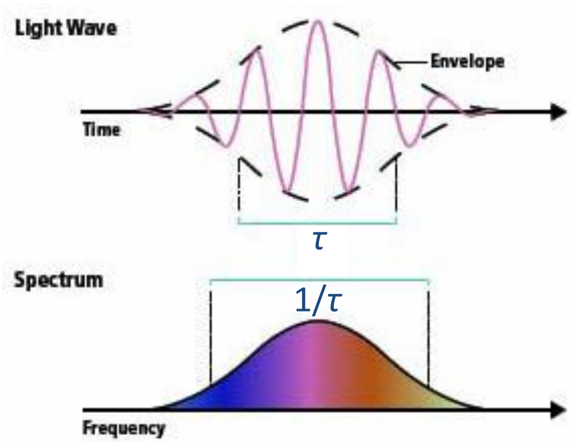
S. Cundiff & J. Ye, Rev. Mod. Phys. 75, 325 (2003)
http://www.fisi.polimi.it/en/research/research_structures/laboratories/54063

Optical Frequency Comb

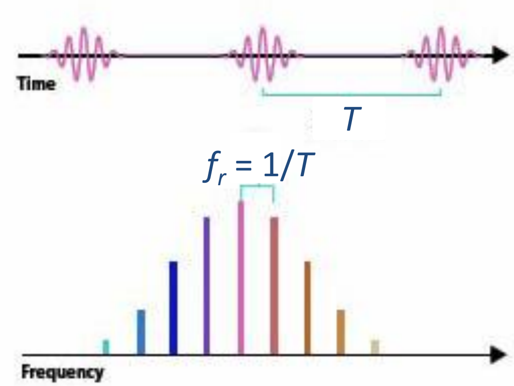
Continuous wave laser



Single pulse

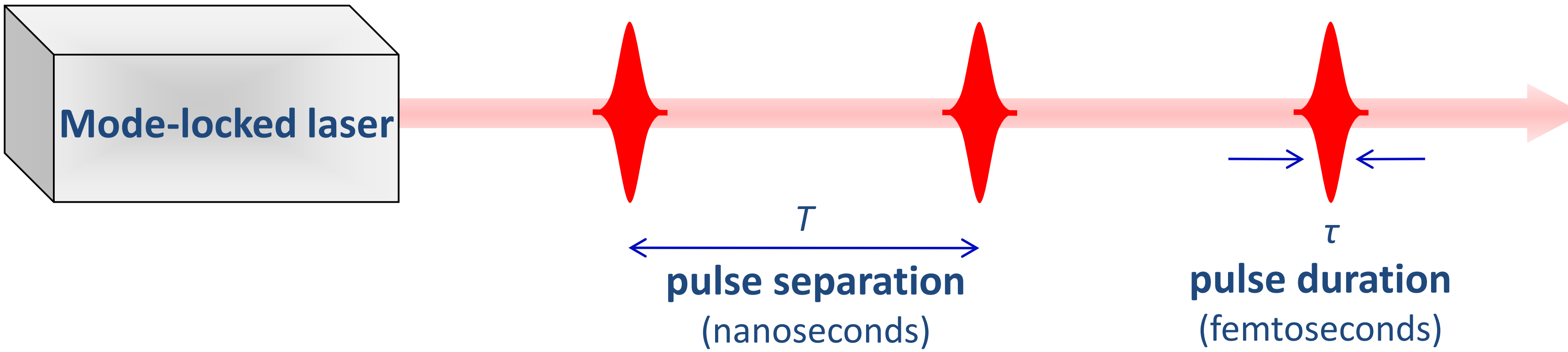


Train of mode-locked pulses



S. Cundiff *et al.*, Scientific American (2008)

Optical Frequency Comb



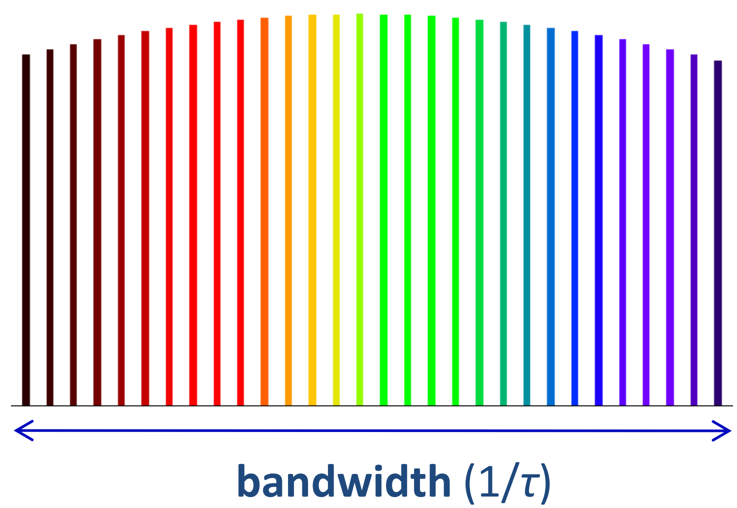
Frequency of the n^th comb mode

repetition rate ($f_r = 1/T$)

$$\nu_n = n f_r + f_0$$

mode number

offset frequency



Todd Johnson and Scott Diddams

Optical Frequency Comb

Two RF frequencies
exactly determine all optical frequencies:

- repetition frequency f_r
- carrier envelope offset frequency f_o

Control

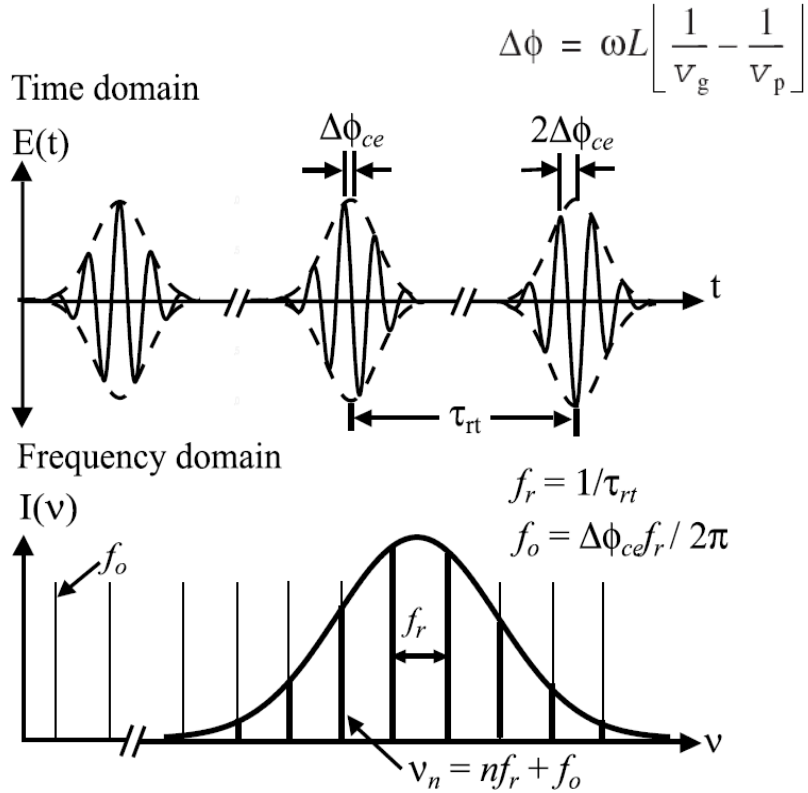
f_r - via laser cavity length

f_0 - via pump power and cavity phase shifts

Measurement

f_r - with fast detector

f_0 - via f-2f interferometry

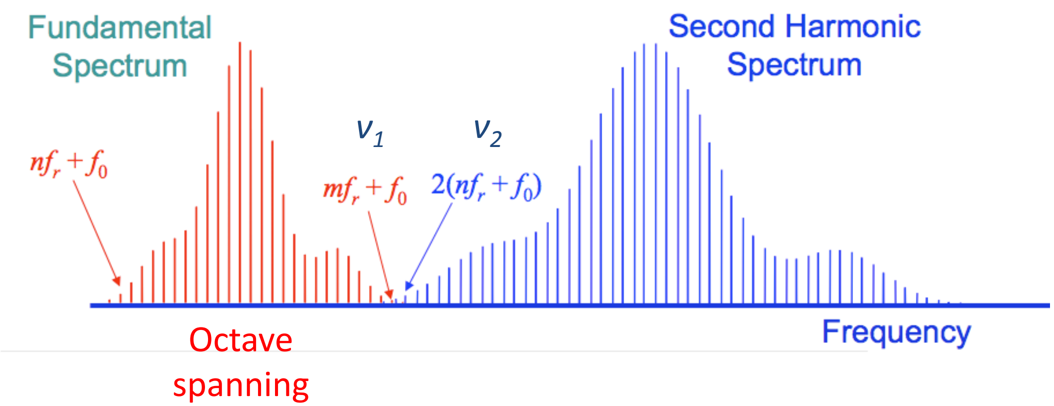


$$\nu_n = nf_r + f_o$$

S. Cundiff & J. Ye, Rev. Mod. Phys. **75**, 325 (2003)
M. J. Thorpe & J. Ye, Appl. Phys. B **91**, 397 (2008)

f-2f Interferometry

Measuring f_o via *f-2f*



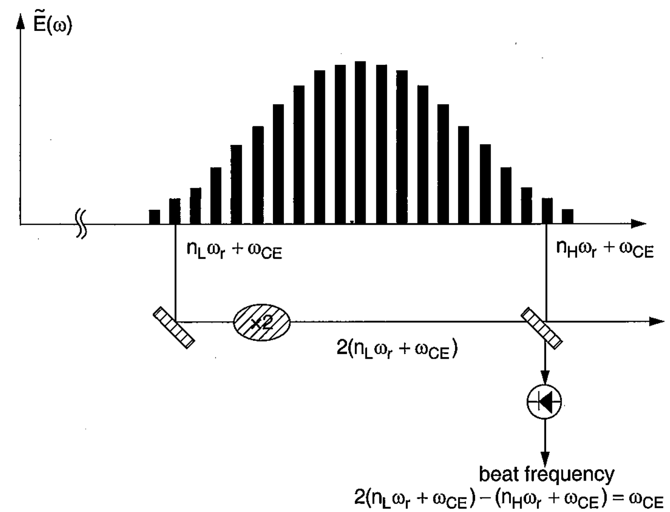


Fig. 9.71. Self-referencing of optical frequencies [9.174]

$$m=2n$$

$$\nu_2 - \nu_1 = 2(nf_r + f_0) - (mf_r + f_0) = f_0$$

W. Demtröder, Laser spectroscopy (2008)

Frequency Comb Sources

Ti:sapphire
- highest repetition rates
- solid state laser
  – free space components

Yb:fiber
- high-power capabilities
- fiber components

Er:fiber
- compact, easy to use
- fiber components

	Ti:Sapphire	Yb:Fiber	Er:Fiber
Wavelength range (directly)	0.7–0.9 μm	1.0–1.1 μm	1.4–1.6 μm
Wavelength range (supercontinuum)	0.4–1.2 μm	0.6–1.4 μm	1.0–2.2 μm
Maximum repetition rate	10 GHz	1 GHz	250 MHz
Output power (average)	<3 W	<50 W	<0.5 W
System design	Oscillator only	Oscillator and amplifier	Oscillator and amplifier
Octave spanning	With PCF or directly	With PCF	With HNF

[a]Abbreviations: HNF, highly nonlinear fiber; PCF, photonic crystal fiber.

F. Adler *et al.*, Annu. Rev. Anal. Chem. **3**, 175 (2010)

Frequency Comb Sources in the MIR

Direct comb sources

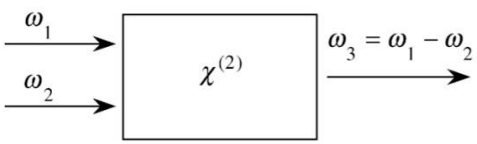
- Cr^{2+} ZnSe @ 2.4 µm
- Tm:fiber @ 2 µm

Other…

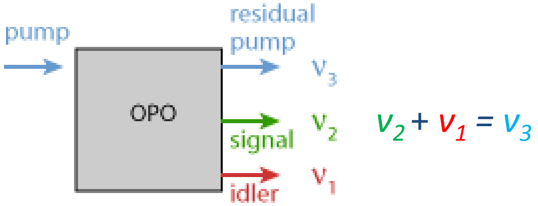
- *microresonators*
- *quantum cascade lasers*
- *towards XUV*
  – high harmonic generation

Nonlinear frequency conversion

- Difference Frequency Generation (DFG)



- Optical Parametric Oscillator (OPO)

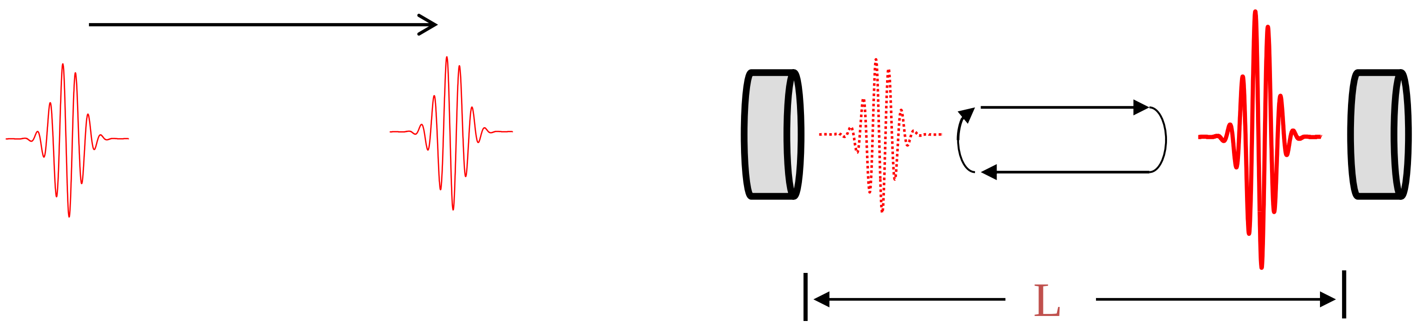


E. Sorokin *et al.*, Opt. Express **15**, 25 (2007)
J. Jiang *et al.*, CLEO Baltimore (2011)

P. Maddaloni *et al.*, New J. Phys. **8** (2006)
D. T. Reid *et al.*, Laser Phys. **18**, 2 (2008)
F. Adler *et al.*, Opt. Lett. **34**, 1330 (2009)
N. Leindecker *et al.*, Opt. Express **19**, 7 (2011)

Comb-Cavity Coupling

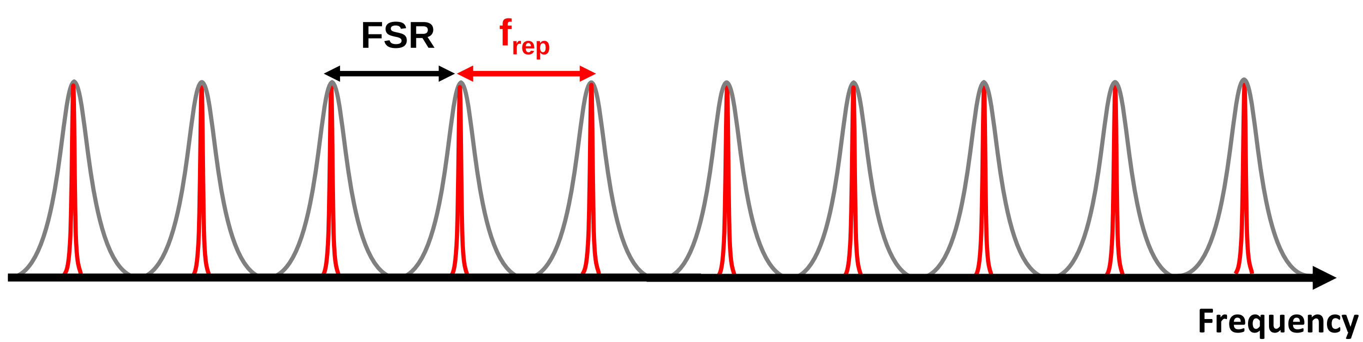
Time domain



Frequency domain

Cavity modes:

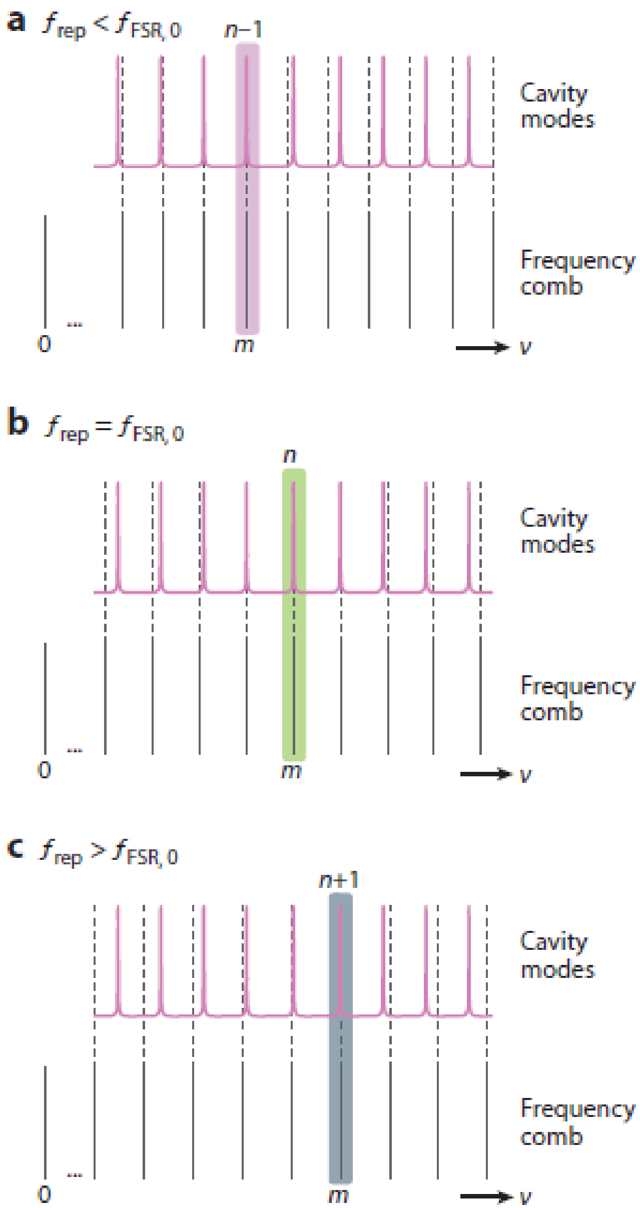
Comb modes:



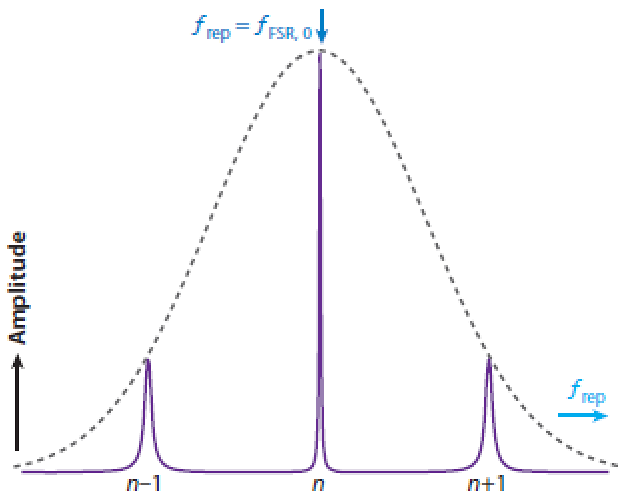
Dispersion…

$$FSR(\omega) = \frac{c}{2L + c\frac{d\phi}{d\omega}}$$

Comb-Cavity Coupling

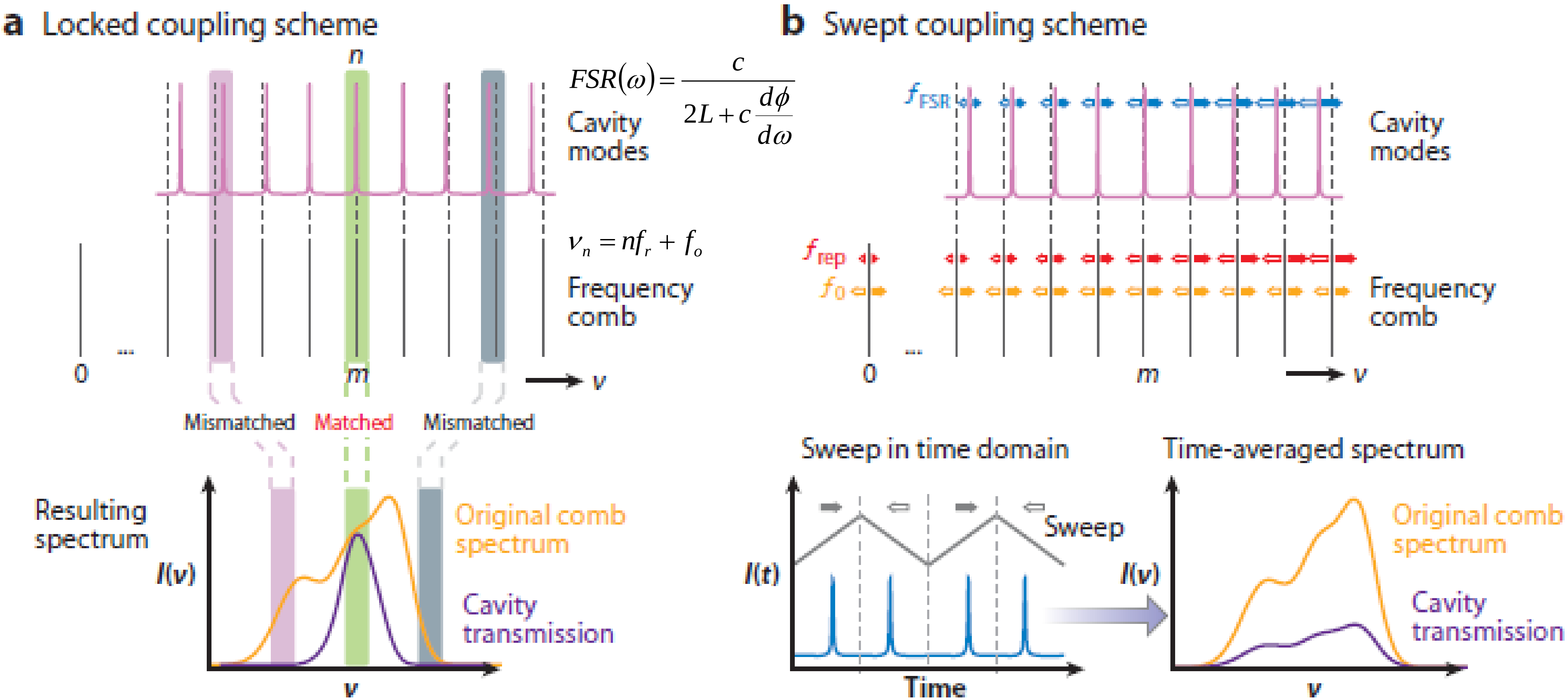


Cavity transmission - f_{rep} scan



F. Adler *et al*., Annu. Rev. Anal. Chem. **3**, 175 (2010)

Comb-Cavity Coupling

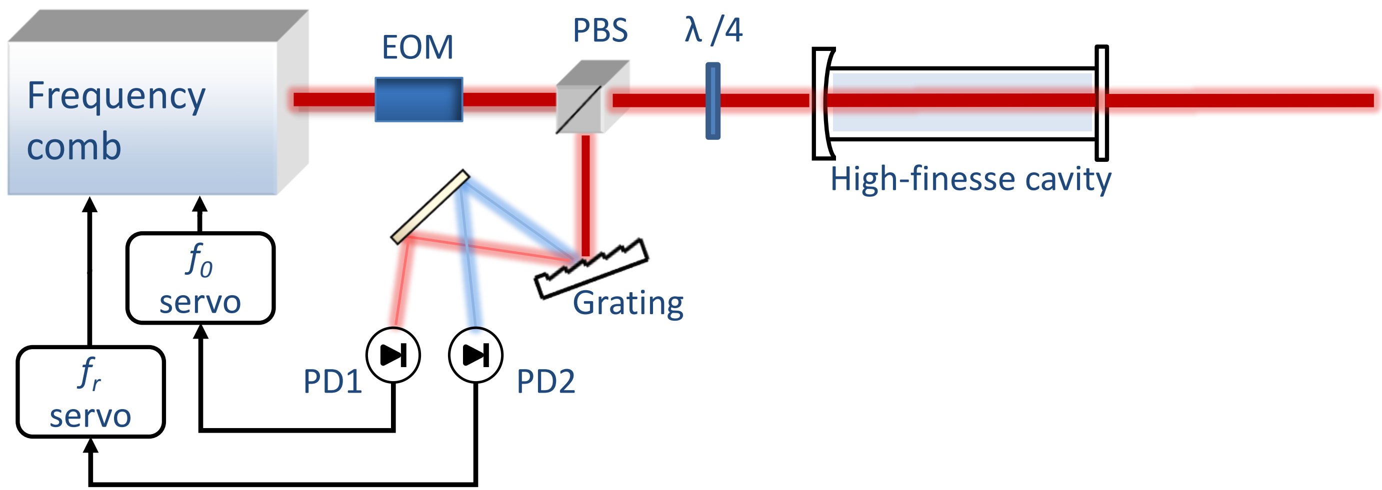


- Constant transmission
- Bandwidth limited by mirror dispersion
- Higher cavity enhancement
- Tight lock

- Lower transmitted power
- Entire bandwidth transmitted
- Lower cavity enhancement
- Dither lock

F. Adler *et al.*, Annu. Rev. Anal. Chem. **3**, 175 (2010)

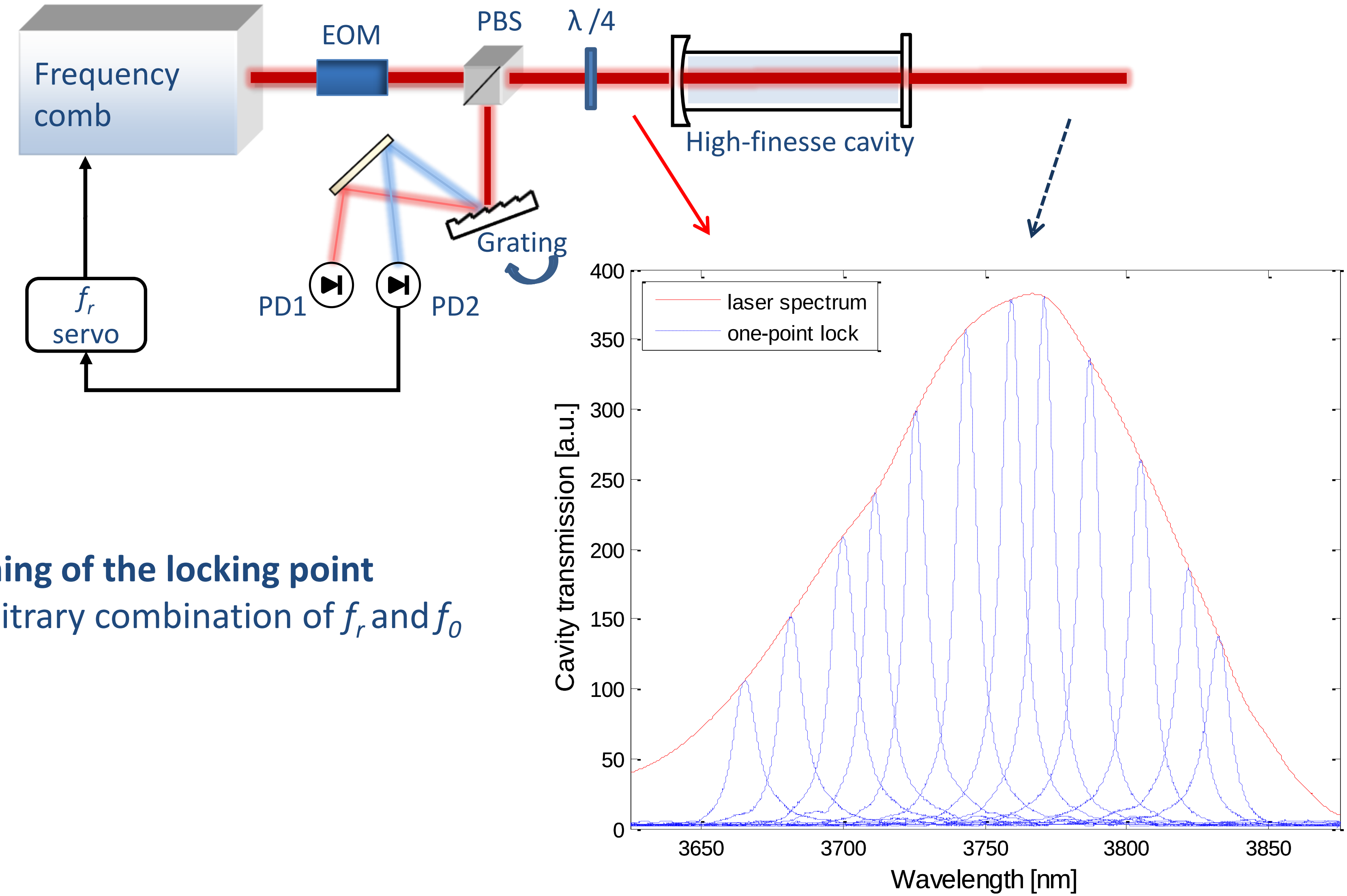
Two-Point Comb-Cavity Lock



Two-point Pound-Drever-Hall locking

Lock both comb degrees of freedom to the cavity

Two-Point Comb-Cavity Lock

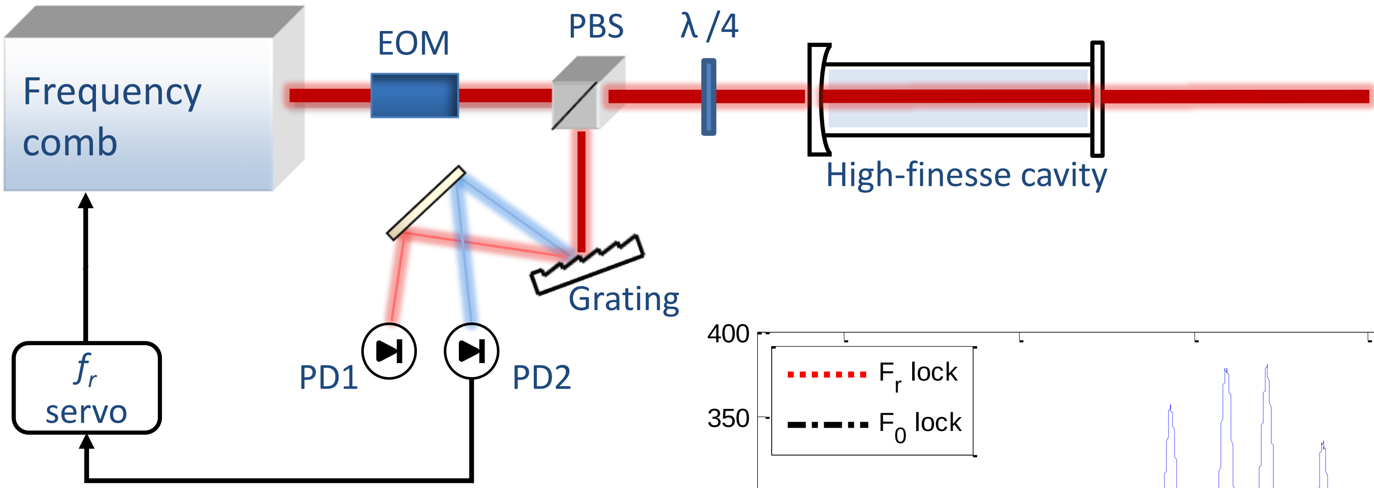


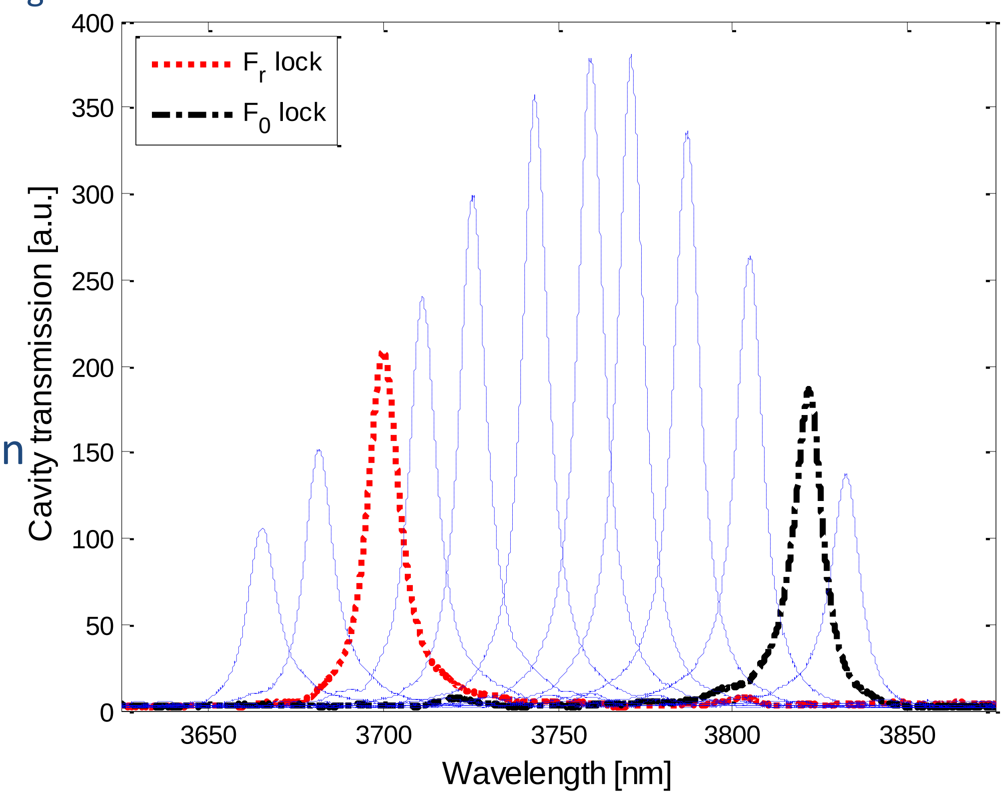
Tuning of the locking point

Arbitrary combination of f_r and f_0

A. Foltynowicz *et al.*, Appl. Phys. B **110**, 163 (2013)

Two-Point Comb-Cavity Lock



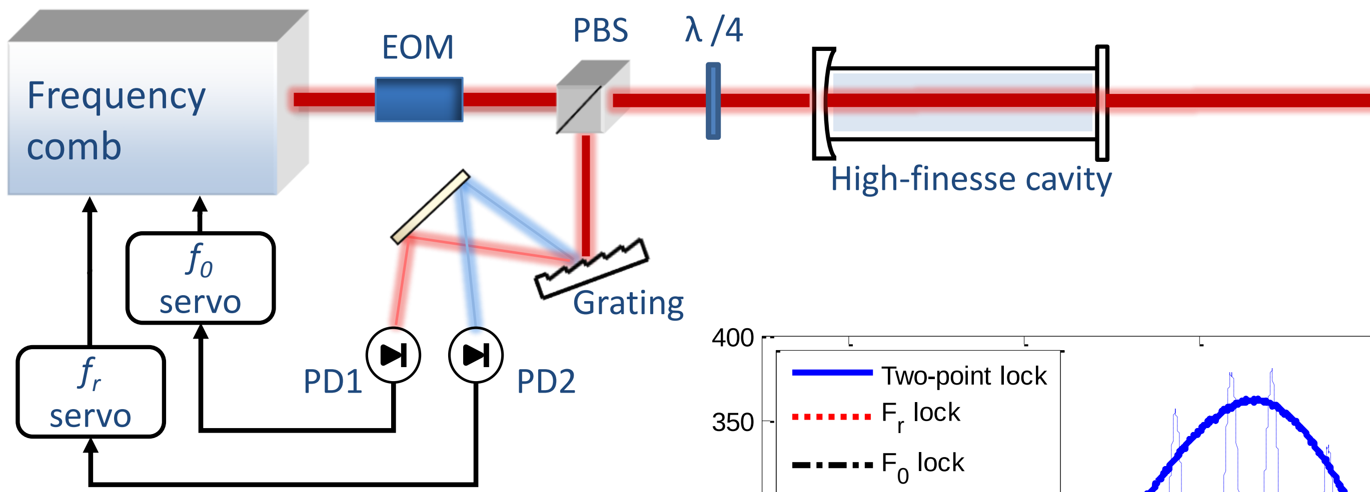


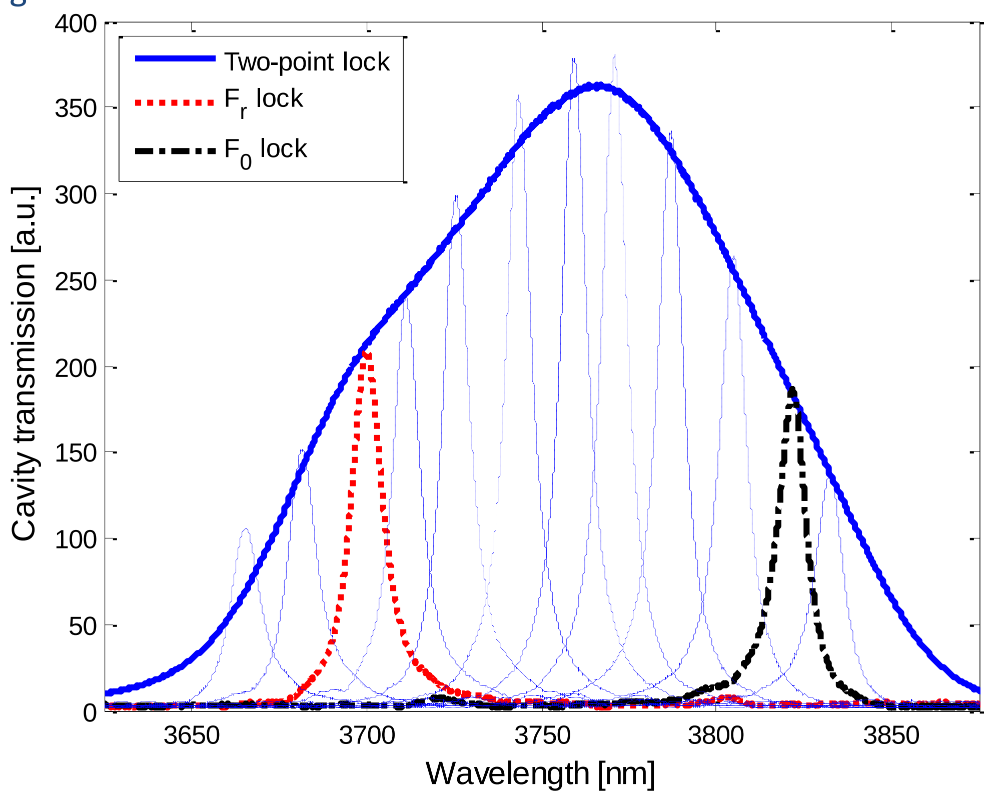
Two locking points

Separation limited by cavity dispersion

A. Foltynowicz *et al.*, Appl. Phys. B **110**, 163 (2013)

Two-Point Comb-Cavity Lock





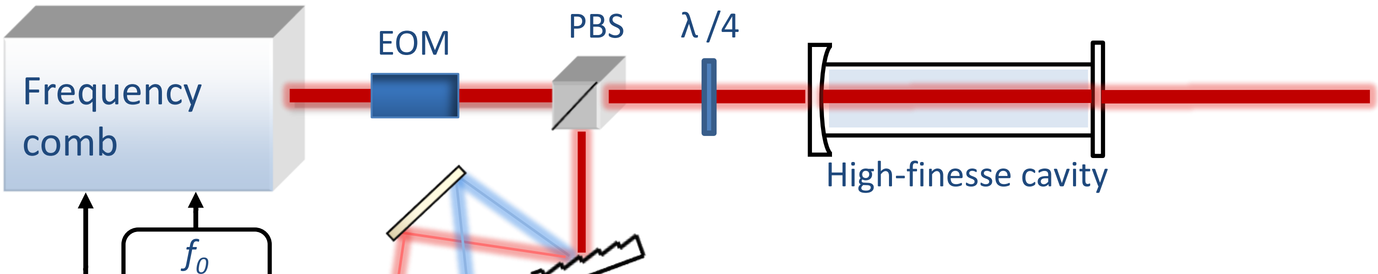
Two-point locking

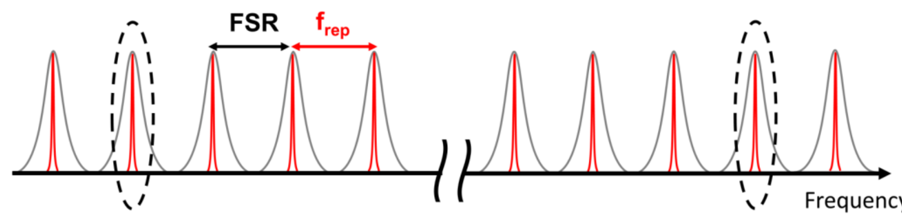
Stable transmission

at and between the locking points

A. Foltynowicz *et al.*, Appl. Phys. B **110**, 163 (2013)

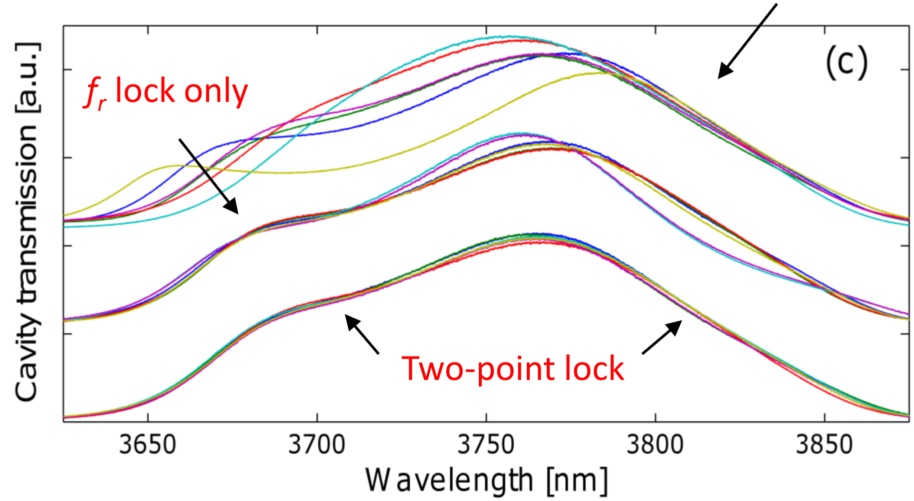
Two-Point Comb-Cavity Lock





Two-point Pound-Drever-Hall locking

Stability of 1-point vs 2-point lock



A. Foltynowicz *et al.*, Appl. Phys. B **110**, 163 (2013)

Detection Methods

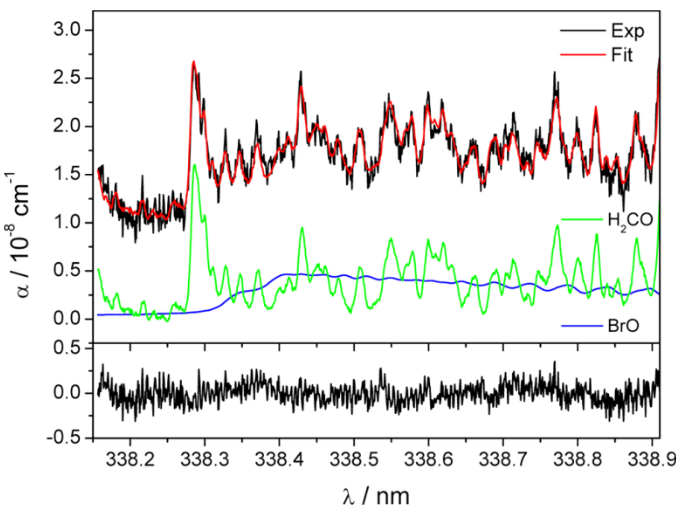
- Dispersion element
  - 1D - spectrograph
  - 2D - VIPA
- Comb-cavity filtering
  - Vernier spectroscopy
- Fourier transform spectroscopy
  - Mechanical FTS
  - Dual comb spsectroscopy

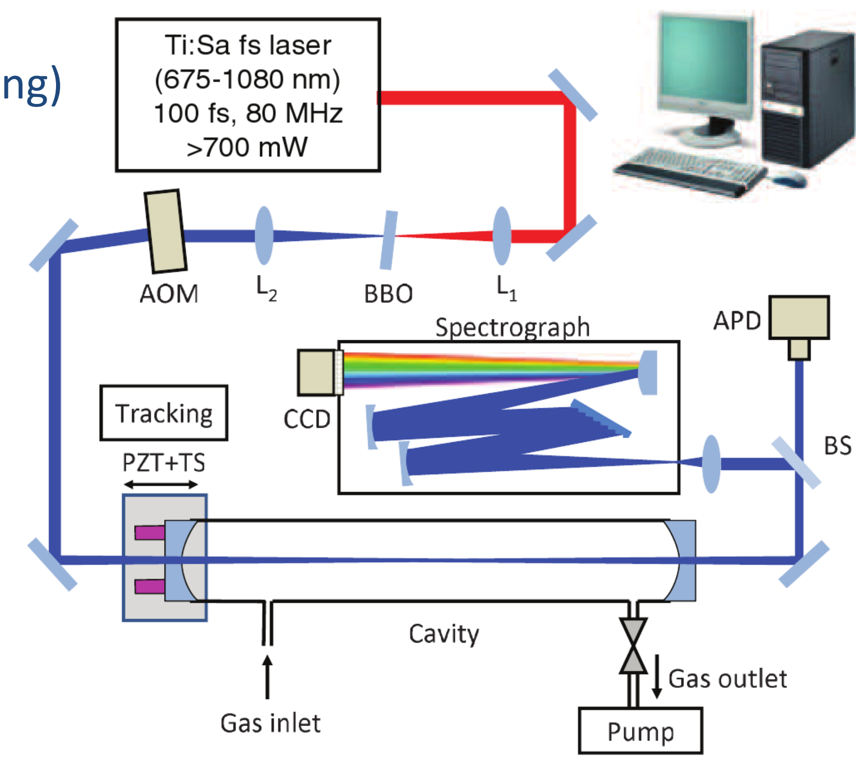
- Resolution
  - Comb lines resolved?
- Bandwidth
  - Entire comb bandwidth?
- Acquisition time
- Frequency calibration
  - Given by the comb?
- Sensitivity
- Stability
  robustness
  simplicity
  ….

Spectrograph

1D dispersion

- Grating spectrograph
- Dither lock
- Resolution - few GHz (limited by the grating)
- Frequency calibration needed
- Spectral coverage - few nm
  (limited by detector array size)
- Fast acquisition times – ms



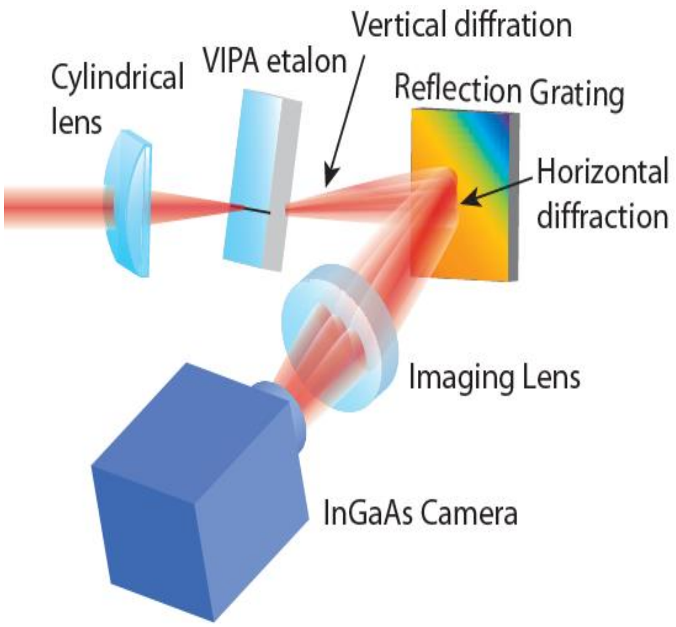


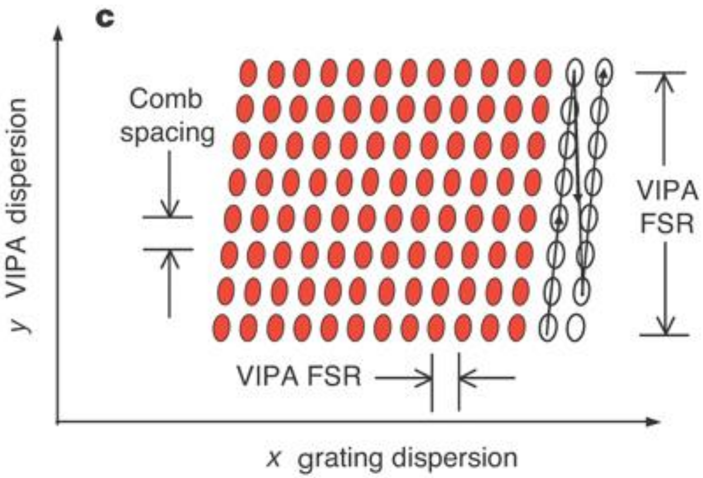
R. Grilli *et al.*, Phys. Rev. A **85**, 051804 (2012)
R. Grilli *et al.*, Environ. Sci. Technol. **46**, 10704–10710 (2012)

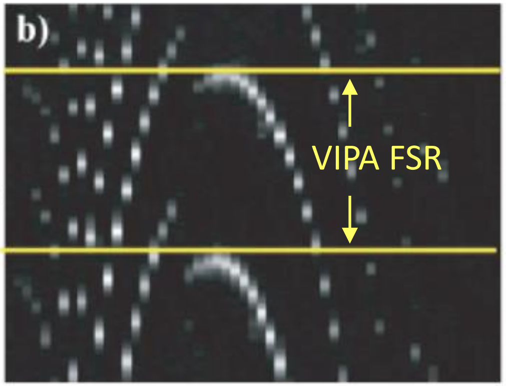
VIPA

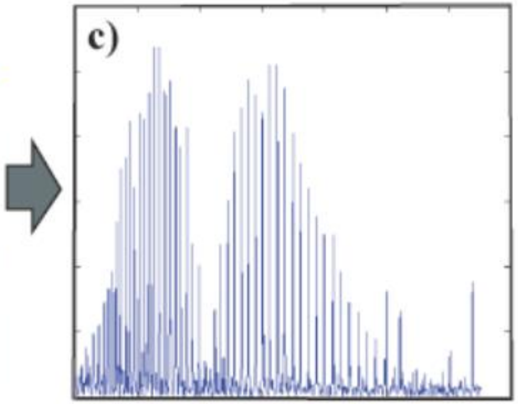
2D detection system

- Virtually imaged phased array (VIPA etalon)
  + grating cross-disperser (grating) + CCD camera
- Dither lock
- Resolution - sub-GHz (limited by the VIPA)
- Frequency calibration needed
  unless comb lines are resolved
- Spectral coverage - few tens of nm
  (limited by detector array size)
- Fast acquisition times – ms







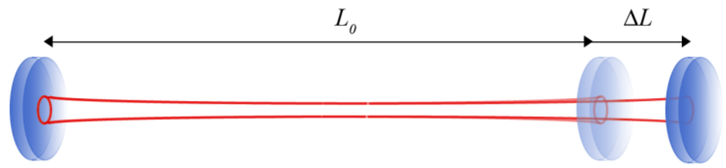


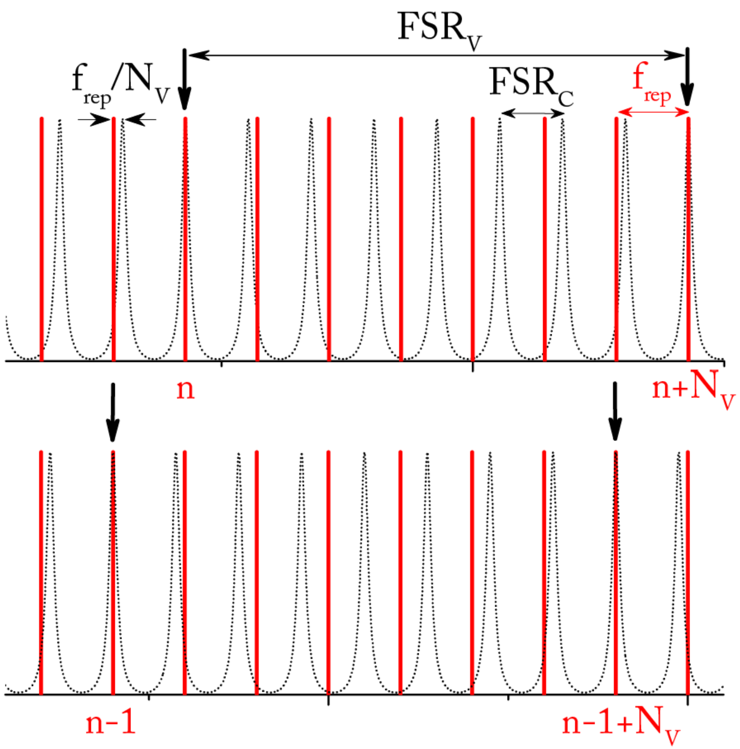
S. A. Diddams *et al.*, Nature **445**, 627 (2007)
M. J. Thorpe *et al.*, Opt. Express **16**, 2387 (2008)

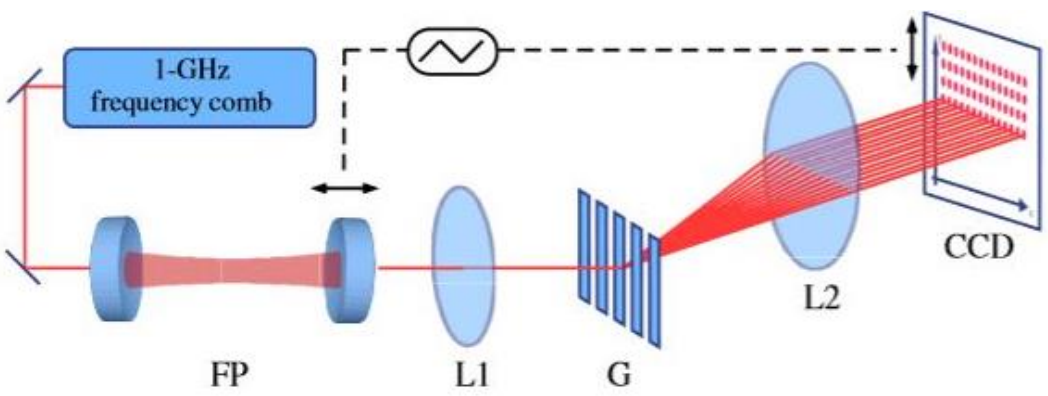
Vernier Coupling

High-resolution filter

- Comb-cavity filtering
  Mode-by mode measurement
- Separated by grating, recorded with CCD
- Resolution - comb lines resolved
- Spectral coverage - few nm
  (limited by detector array size)
- Acquisition time - ms
- Low sensitivity





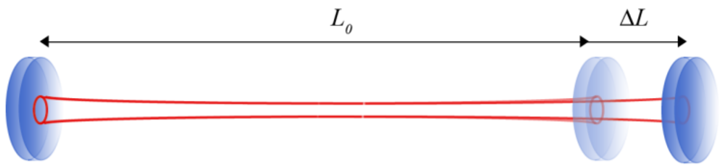


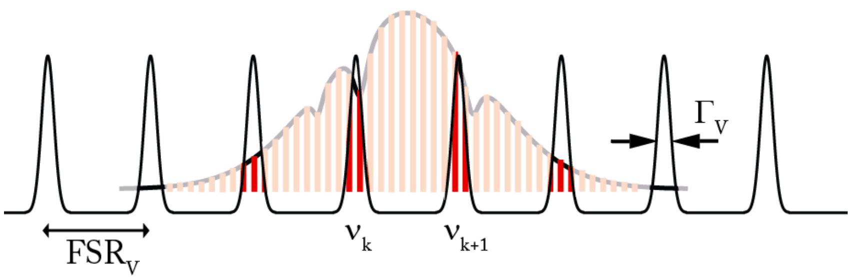
C. Gohle *et al*., Phys. Rev. Lett. **99**, 263902 (2007)
F. Zhu *et al*,. Opt. Express **22**, 23026 (2014)

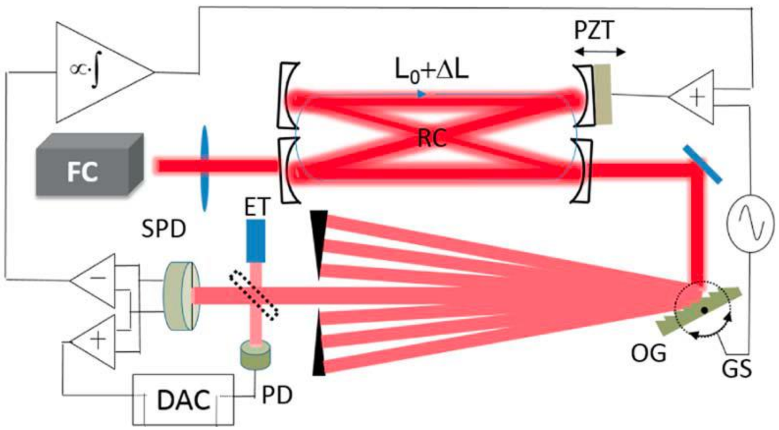
Vernier Coupling

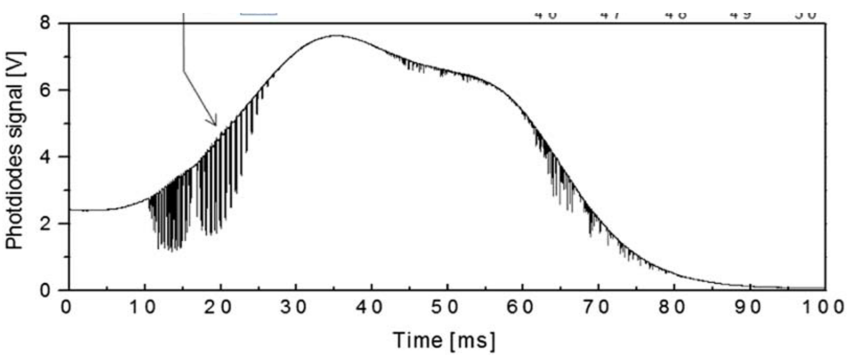
Low-resolution filter

- Comb-cavity filtering
  A few modes at a time
- Single detector
- Resolution - low GHz
- Frequency calibration needed
- Spectral coverage - entire comb bandwidth
- Acquisition time - hundreds of ms









L. Rutkowski and J. Morville, Opt. Lett. **39**, 6664 (2014)

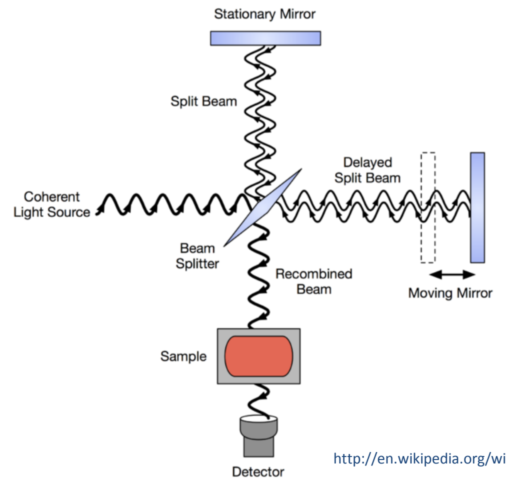
Fourier Transform Spectroscopy

Time-domain measurement

- Interferogram + FFT
- Single detector
- Spectral coverage
  - entire comb bandwidth
- Tight comb-cavity cavity lock

Mechanical FTS

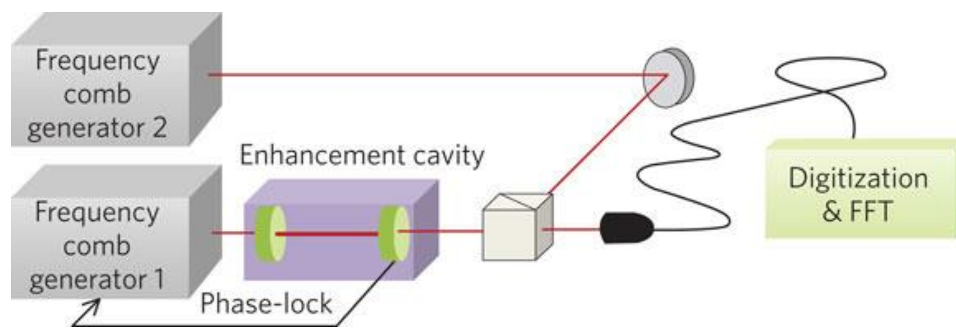
Michelson interferometer



http://en.wikipedia.org/wiki/File:FTIR_Interferometer.png

Dual comb spectroscopy

FTS without moving parts



B. Bernhardt *et al.*, Nat. Photonics **4**, 55 (2010)
A. Foltynowicz *et al.*, Phys. Rev. Lett. **107**, 233022 (2011)

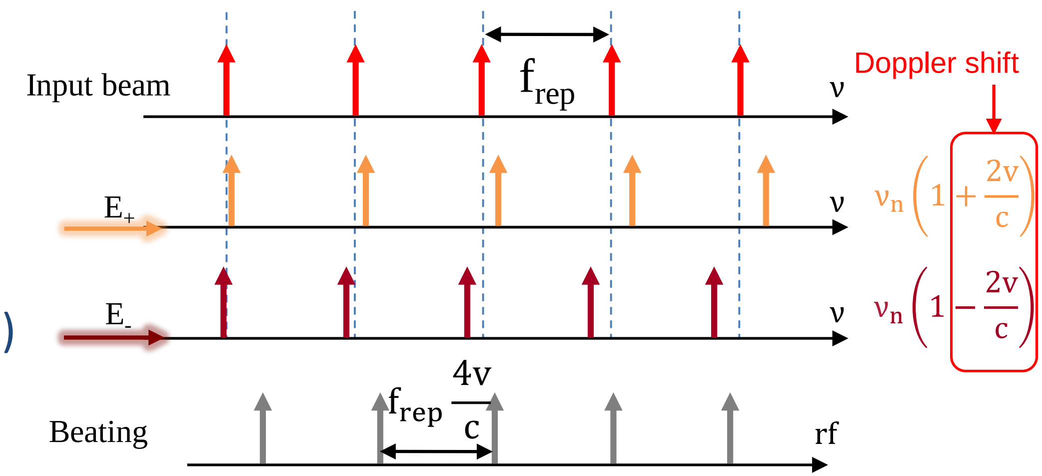
Fourier Transform Spectroscopy

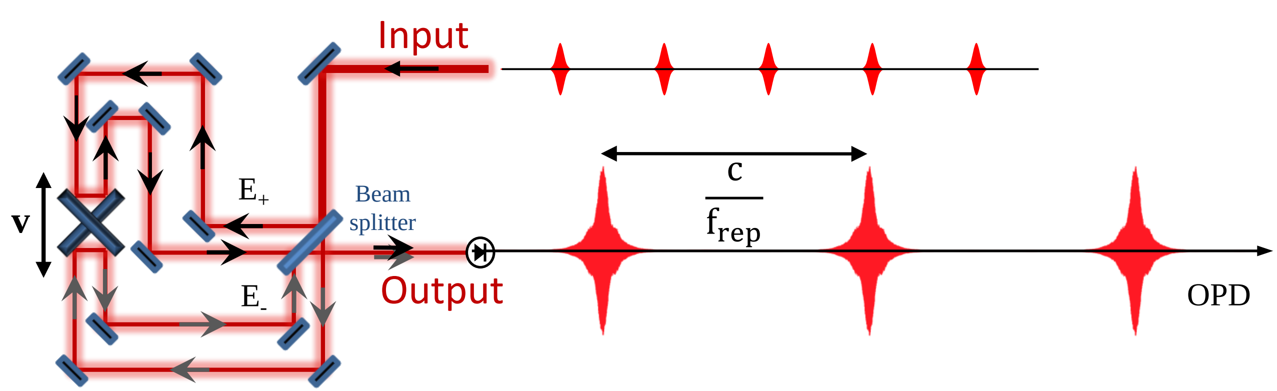
Time-domain measurement

- Interferogram + FFT
- Single detector
- Spectral coverage
  - entire comb bandwidth
- Tight comb-cavity cavity lock
- Resolution - hundreds of MHz (inverse of the optical path difference)
- Optical path difference calibrated by a cw laser
- Comb lines can be resolved
- Acquisition times - s

Mechanical FTS

Michelson interferometer





J. Mandon et al., Nat. Photonics **3**, 99 (2009)
A. Foltynowicz *et al.*, Phys. Rev. Lett. **107**, 233022 (2011)

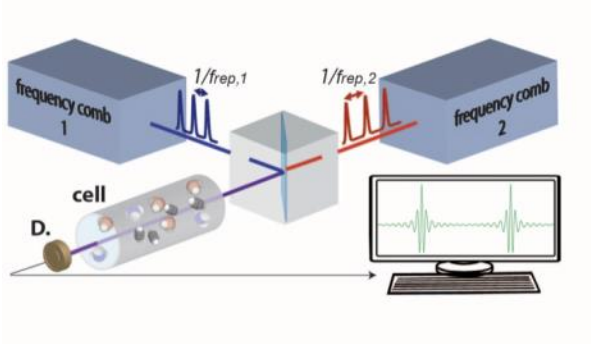
Fourier Transform Spectroscopy

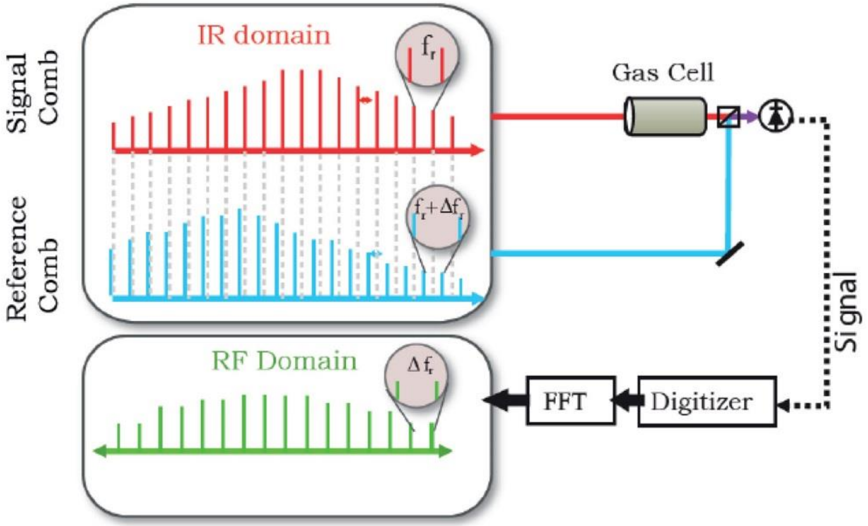
Time-domain measurement

- Interferogram + FFT
- Single detector
- Spectral coverage
  - entire comb bandwidth
- Tight comb-cavity cavity lock
- Two combs with different repetition rates
- Requires comb stabilization
  or adaptive sampling
  with reference cw lasers
- Comb lines can be resolved
  (not shown with a cavity)
- Acquisition times - µs-ms

Dual comb spectroscopy

FTS without moving parts





I. Coddington et al., Phys. Rev. Lett. **100**, 013902 (2008)
B. Bernhardt *et al.*, Nat. Photonics **4**, 55 (2010)

Sensitivity of CE-OFCS

Noise equivalent absorption (NEA)

- Standard deviation of the noise on the baseline, σ
- Cavity enhancement kF/π, k = 1…2, depending on comb-cavity coupling

$$\alpha_{\min} = \sigma\left(\frac{kFL}{\pi}\right)^{-1} T^{1/2}$$ [cm^{-1} Hz$^{-1/2}$]

or

$$\alpha_{\min} = \sigma\left(\frac{kFL}{\pi}\right)^{-1} @T$$ [cm^{-1}] in a given measurement time

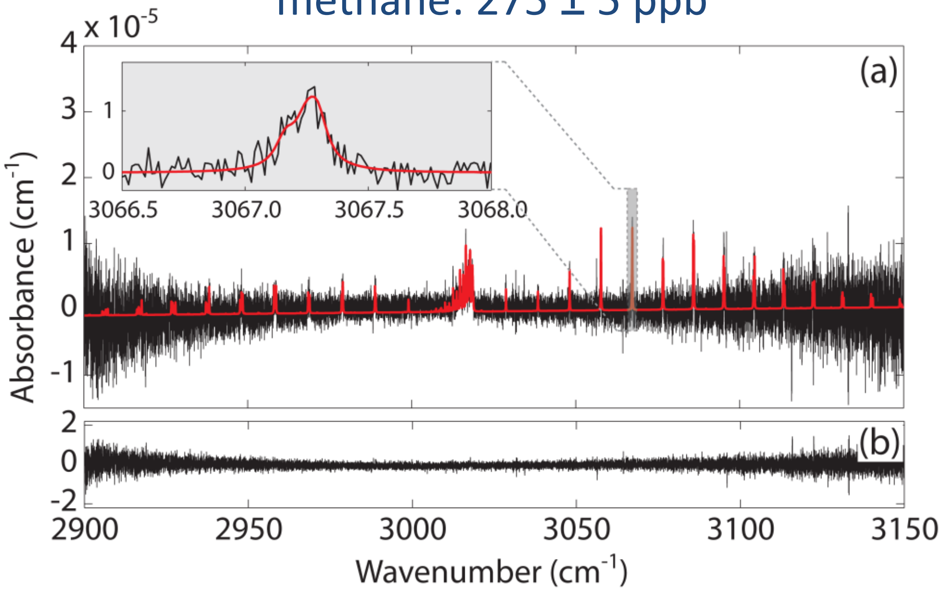
Sensitivity per spectral element (figure of merit)

- Normalized to the number of spectral element, M, to reflect the broadband advantage

$$sensitivity = \alpha_{\min} M^{-1/2}$$ [cm^{-1} Hz$^{-1/2}$]

Multiline Fitting

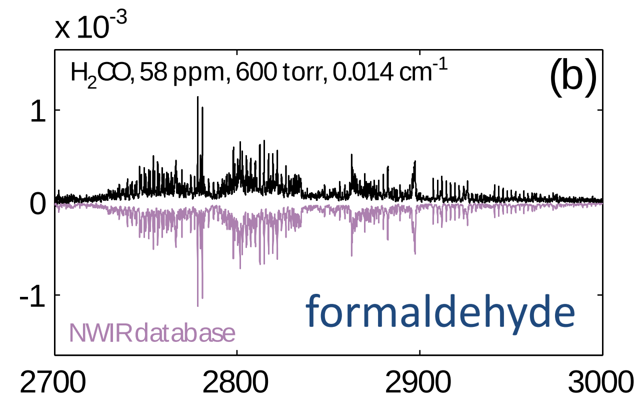
methane: 275 ± 5 ppb

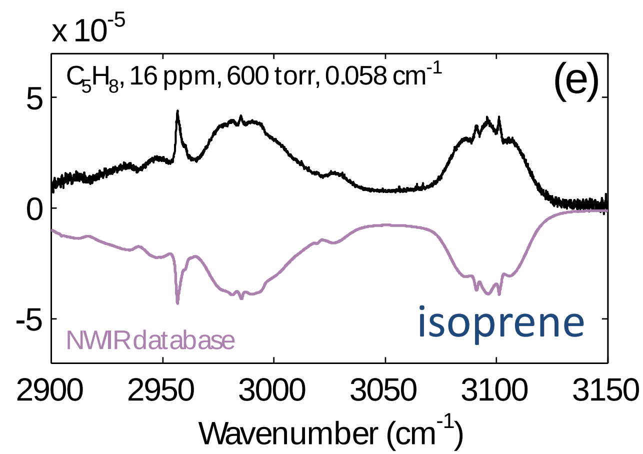


Sensitivity improvement

proportional to integrated absorption

$$S_N = \frac{\sigma_\alpha}{\left[\sum_{i=1}^{k} \alpha_T^{\,2}(\nu_i)\right]^{1/2}}$$





Molecule	Band center (cm^{-1})	Noise Equivalent Concentration (ppb)	Detection Limit (ppb)
CH_4	3020	22	5
H_2CO	2780	54	9
C_5H_8	3000	370	7

F. Adler *et al.*, Opt. Express **18**, 21861 (2010)

Technique Comparison

	FoM $[cm^{-1}\ Hz^{-1/2}]$	Finesse	α_{min} $[cm^{-1}]$	Acq. Time	Resolution	Spectral Coverage
CRDS M. J. Thorpe et al., Science, 311 (2006)	1.5×10^{-11} *	4 500	2.5×10^{-10}	1 s	25 GHz	15 nm detector
High-resolution Vernier C. Gohle et al. PRL, 99 (2007)	8×10^{-9} *	3 000	5×10^{-6}	10 ms	1 GHz *	10 nm detector
VIPA M. J. Thorpe et. al. Opt. Exp. 16 (2008)	7.4×10^{-11} *	28 000	8×10^{-10}	30 s	800 MHz *	25 nm detector
Dual comb B. Bernhardt et al. Nat. Phot. 4 (2010)	7×10^{-11} *	1 200	3×10^{-8}	18 µs	4.5 GHz	20 nm cavity
FT Spectrometer A. Foltynowicz et al. PRL 107 (2011)	3.4×10^{-11}	8 000	1.4×10^{-9}	6 s	380 MHz	30 nm cavity
Spectrograph R. Grilli et al. PRA 85 (2012)	1.5×10^{-11}	32 000	3×10^{-9}	12 ms	10 GHz	1.5 nm detector
Low-resolution Vernier L. Rutkowski et al. Opt. Lett. 39 (2014)	4×10^{-11}	3 000	7×10^{-9}	1.5 s	2 GHz	75 nm laser
NICE-OFCS A. Khodabakhsh et al. APB 119 (2015)	6.4×10^{-11}	9 000	4×10^{-9}	1 s	750 MHz	30 nm cavity

* not quoted
or quoted incorrectly

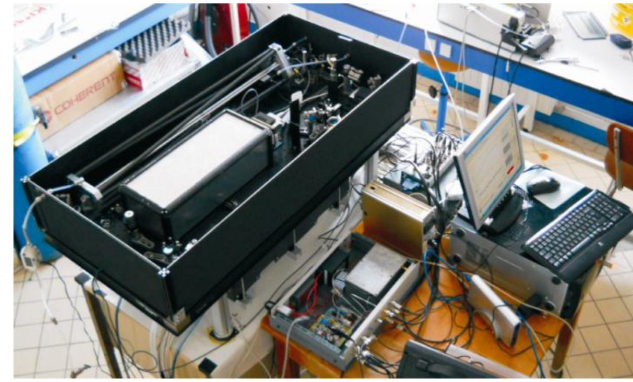
* comb lines resolved

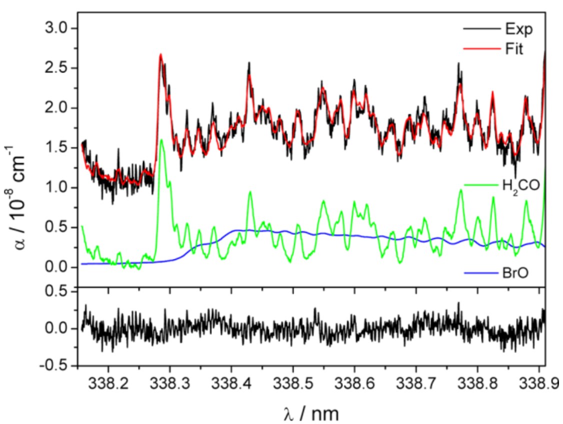
Environmental Monitoring

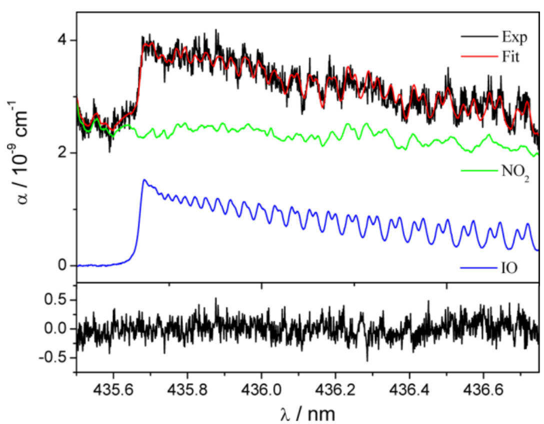
Detection of highly reactive halogenated radicals, formaldehyde and nitrogen dioxide

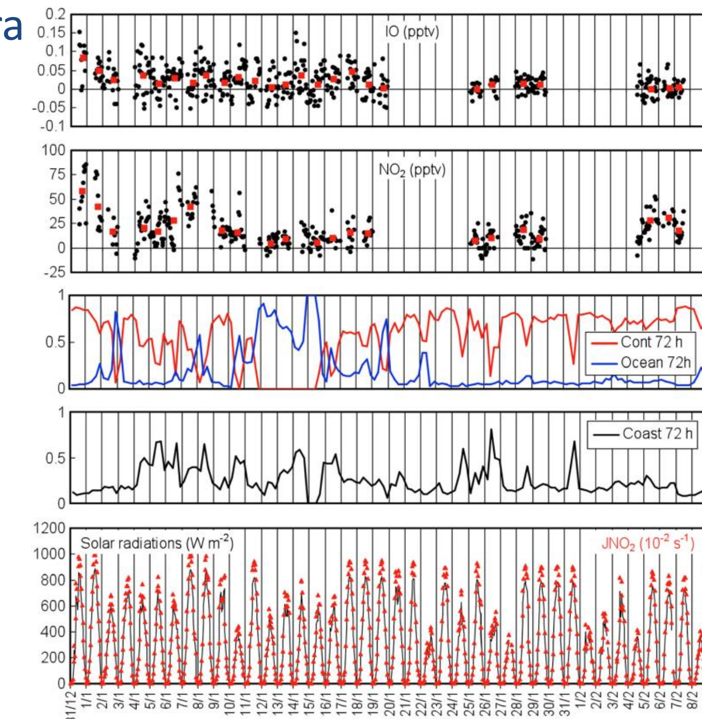
Field campaigns:
- at the Marine Boundary Layer in Roscoff (North West Atlantic coast of France)
- at Dumont d'Urville (East Antarctic coast)

- Frequency-doubled Ti:Sapph: 338 and 436 nm
- Compact spectrograph: echelle grating + CCD camera
- Two parallel cavities (BrO + H_2CO and IO + NO_2)
- Sensitivity 1.3×10^{-11} cm^{-1} $Hz^{-1/2}$ per sp. el.
- 20 ppq of IO in 5 min









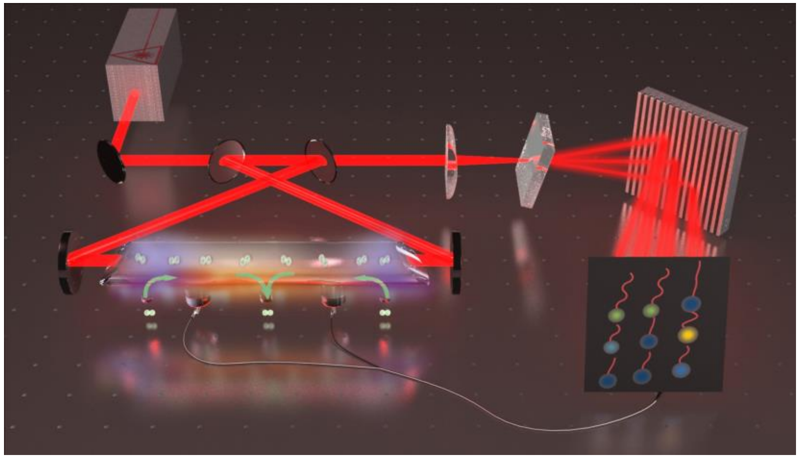
R. Grilli *et al.*, Env. Sci. Tech. **46**, 10704 (2012)
R. Grilli *et al.*, Geophys. Res. Lett. **40**, 791 (2013)

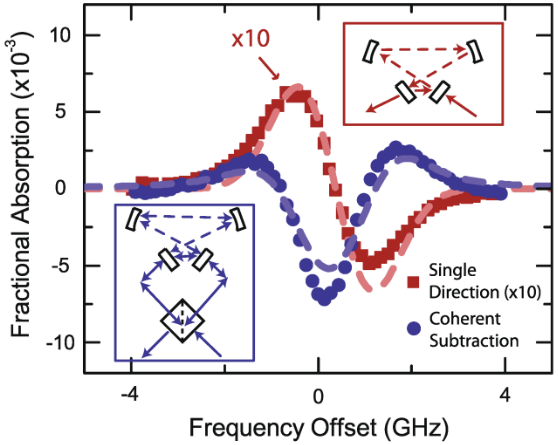
Ion Selective Detection

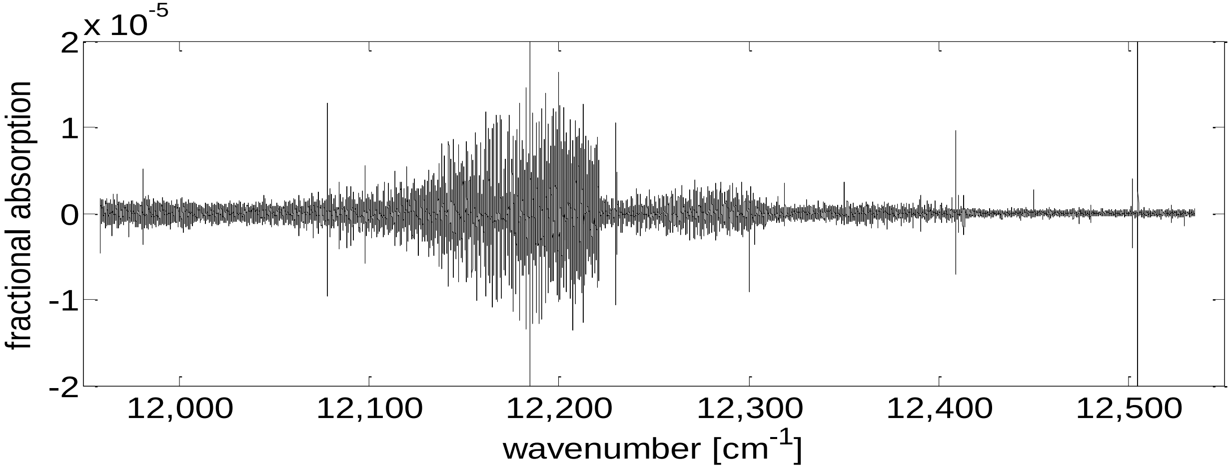
Spectroscopic data for $^3\Delta_1$ metastable state of HfF^+ and ThF^+
high sensitivity for electron electric dipole moment (eEDM) search

Frequency comb velocity modulation spectroscopy

- 3 GHz Ti:Sapph laser
- VIPA etalon + grating + Heliotis lock-in camera
- Alternating current discharge inside a ring cavity
  - produces and modulates the ions
  (Doppler shift)
- Lock-in detection on every comb tooth
- Sensitivity 4 x 10^{-8} $Hz^{-1/2}$ per sp. el.
- 150 cm^{-1} in under an hour







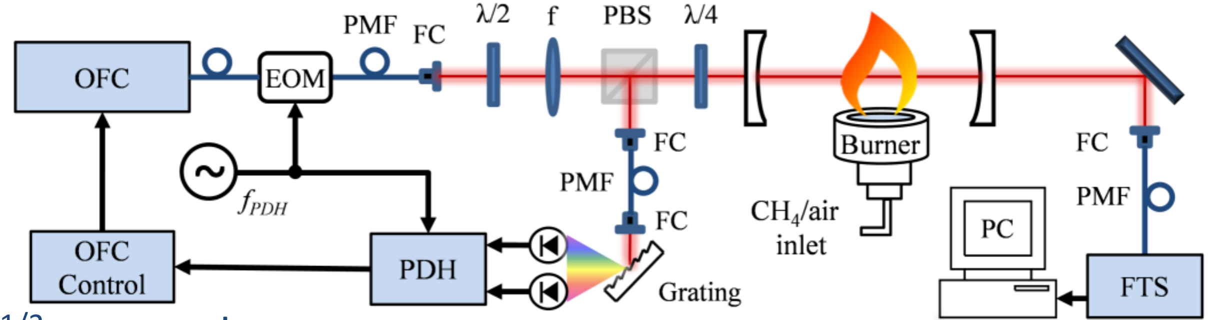
L. C. Sinclair *et al.*, Phys. Rev. Lett. **107**, 093002 (2011)
K. C. Cossel *et al.*, Chem. Phys. Lett. **546**, 1 (2012)

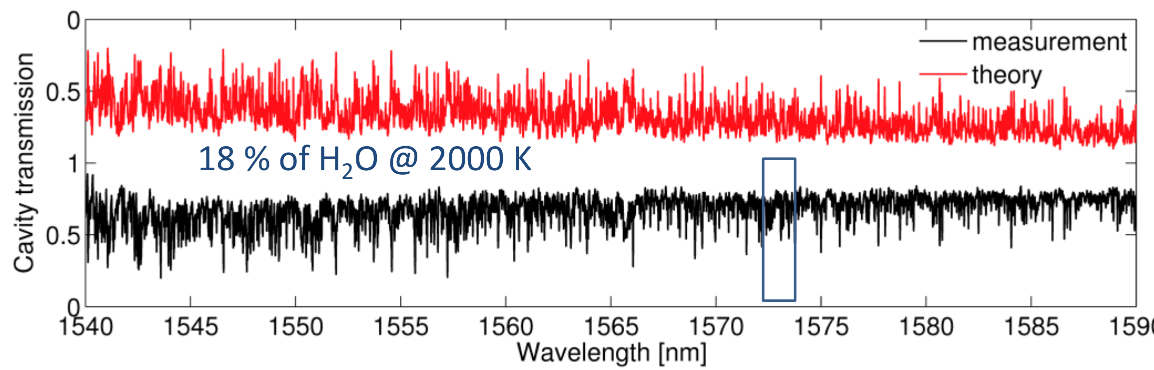
Combustion Analysis

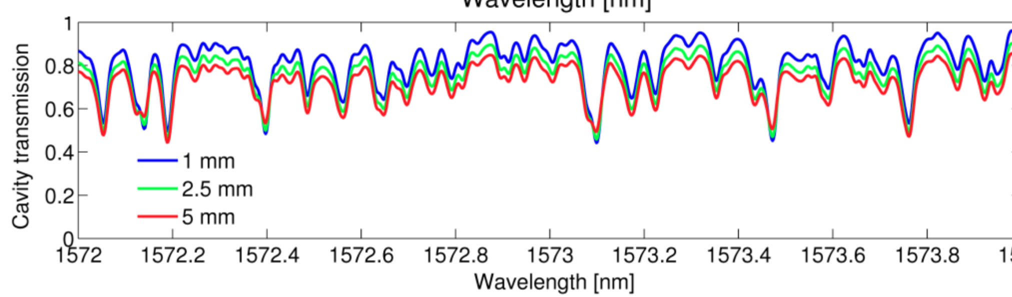
High-temperature water spectra in premixed methane/air flat flame

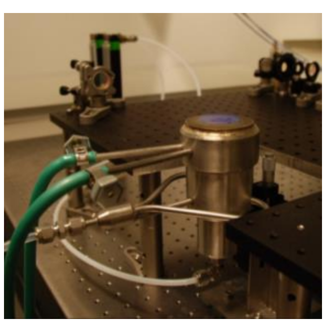
concentration and temperature characterization

- Er:fiber laser: 1.5 µm
- Fast-scanning FTS
  with autobalanced detection
- Two-point Pound-Drever-Hall
  comb-cavity lock
- Sensitivity 4.2×10^{-9} cm^{-1} Hz$^{-1/2}$ per sp. el.
- 50 nm of bandwidth with
  1 GHz resolution in 0.4 s
- Premixed methane/air
  flat flame burner, dia 3.8 cm







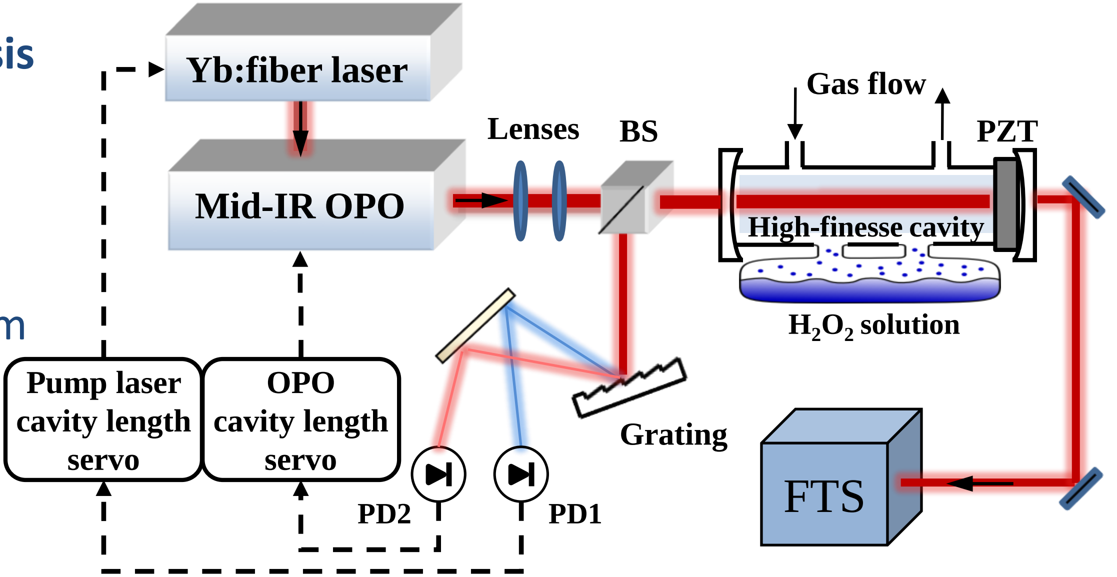


C. Abd Alrahman *et al.*, Opt. Express **22**, 13889 (2014)

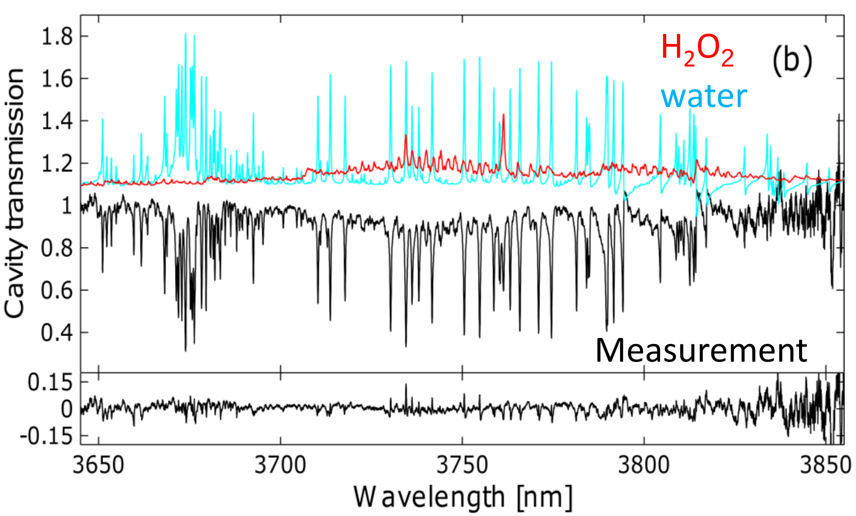
Hydrogen Peroxide Detection

Potential applications in breath analysis
marker for
oxidative stress in the lungs, asthma
chronic obstructive pulmonary disease
acute respiratory distress syndrome

- Yb:fiber pumped OPO: 2.8 to 4.8 µm
- Fast-scanning FTS with autobalanced detection
- Two-point Pound-Drever-Hall comb-cavity lock
- Sensitivity 6.9×10^{-11} cm^{-1} Hz$^{-1/2}$ per sp. el.
- Detection of H_2O_2 in the presence of % level of water
- Concentration detection limit 130 ppb of H_2O_2 in 3% of water in 1 s



5 ppm of H_2O_2 in 1.2 % of H_2O @ 37 °C

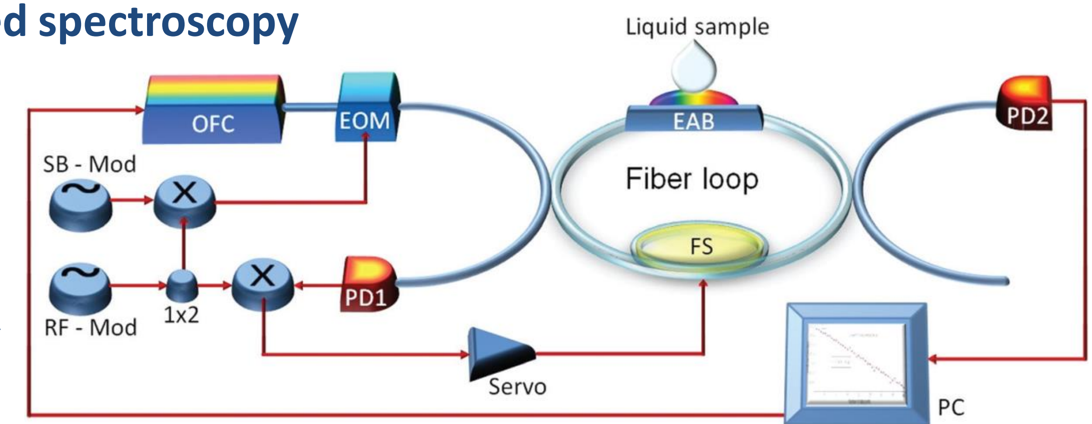


A. Foltynowicz *et al.*, Appl. Phys. B **110**, 163 (2013)

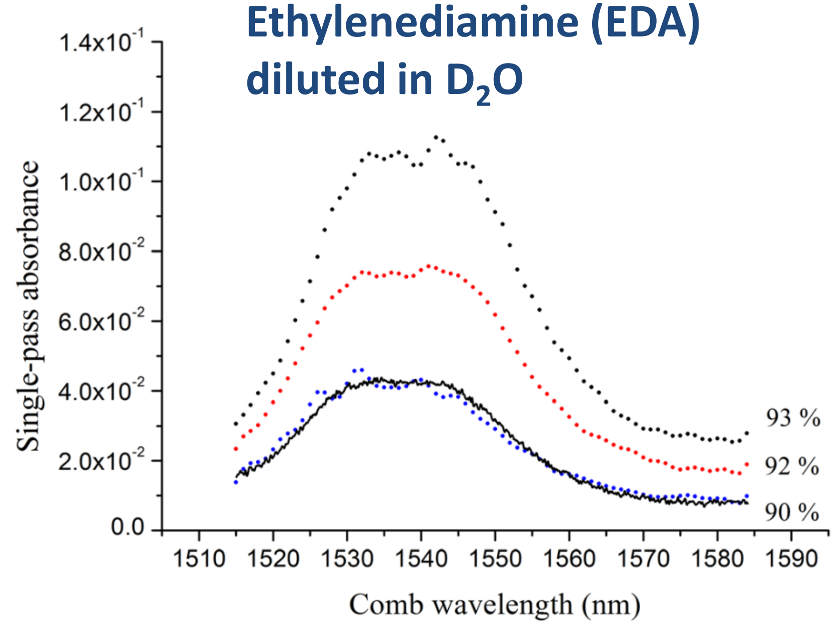
Fiber Sensing of Liquids

Evanescent-wave fiber cavity-enhanced spectroscopy sensing of liquids

- All-fiber setup
- Er:fiber laser: 1.5 μm
- fiber-loop cavity
  with evanescent-wave access block
- Cavity-comb PDH lock
- Comb filtering by cavity dispersion, f_{rep} sweep
- Cavity ringdown measurement
- Sensitivity 3 x 10^{-4} cm^{-1} $Hz^{-1/2}$ per sp. el.
- Detection of liquid polyamines
- Full spectrum recorded in 120 s



Ethylenediamine (EDA) diluted in D_2O

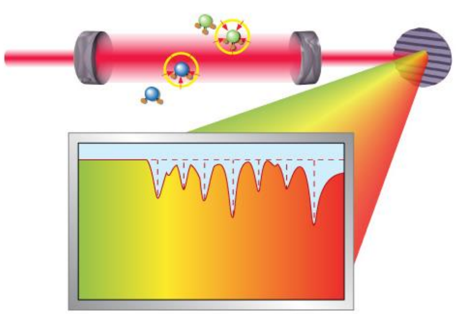


S. Avino *et al.*, Appl. Phys. Lett. **102**, 201116 (2013)

Summary

Simultaneous measurement with thousands of synchronized narrow laser lines

- Broad spectral bandwidth for multispecies detection and acquisition of entire absorption bands
- High resolution for identification and quantitative analysis of individual spectral features
- Fast acquisition for time dependence
- High sensitivity for trace gas detection



Questions?