



Noise-Immune Cavity-Enhanced Optical Frequency Comb Spectroscopy

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Laser-based Spectroscopic Technique



Expectations

- Broad spectral bandwidth
- High spectral resolution
- High absorption sensitivity
- Short measurement time with high SNR

Laser-based Spectroscopic Technique



Solutions

✓ Optical Frequency Comb

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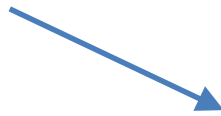


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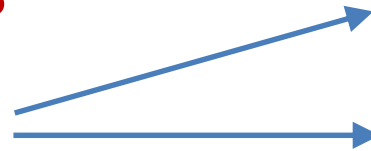


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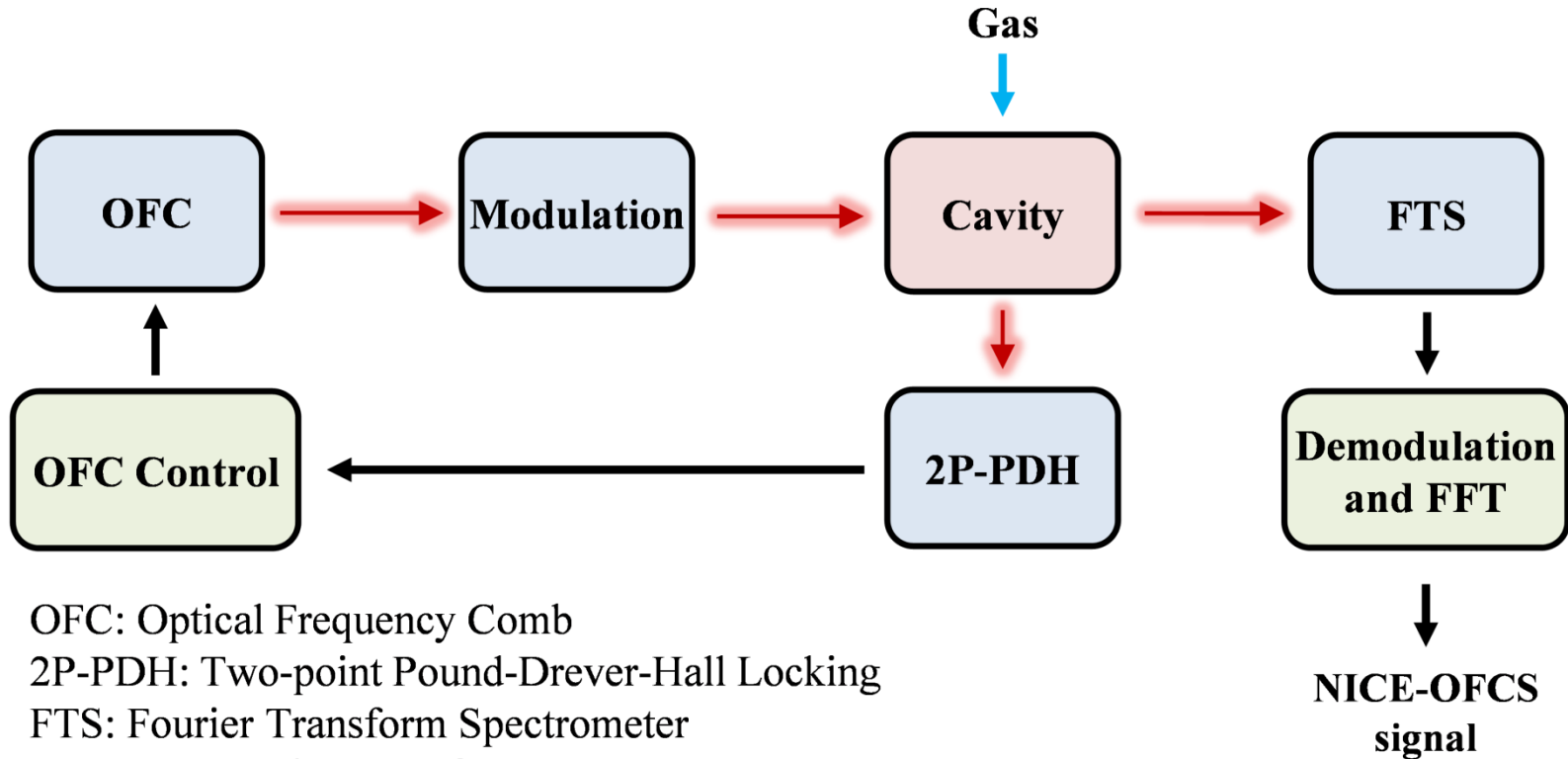
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Fourier-Transform-Based Noise-Immune
Cavity-Enhanced Optical Frequency Comb
Spectroscopy (FT-Based NICE-OFCS)

FT-based NICE-OFCS Principle



OFC: Optical Frequency Comb

2P-PDH: Two-point Pound-Drever-Hall Locking

FTS: Fourier Transform Spectrometer

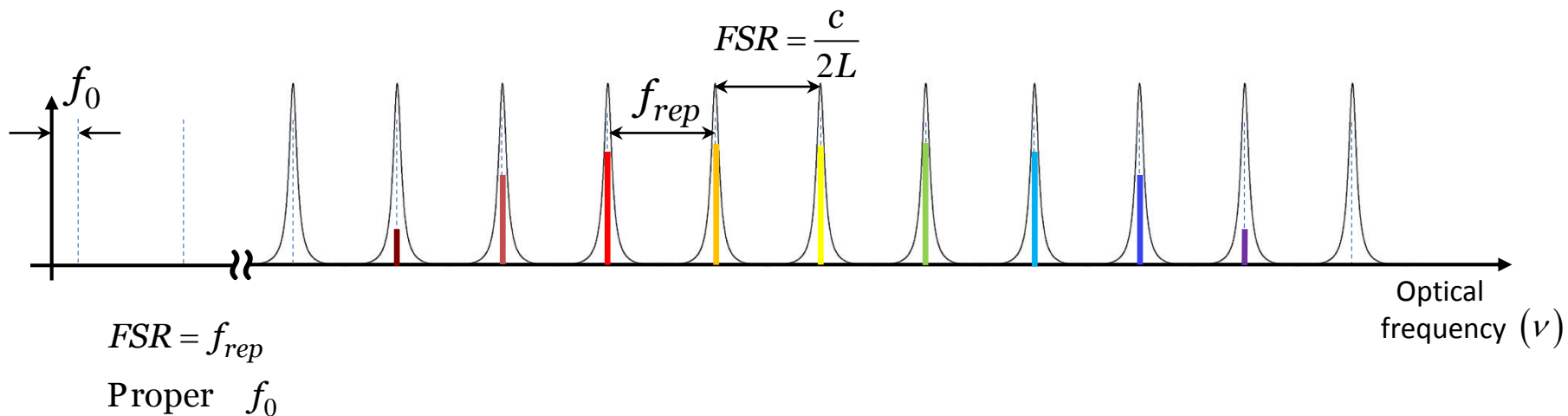
FFT: Fast Fourier Transform

→ Optical Connection

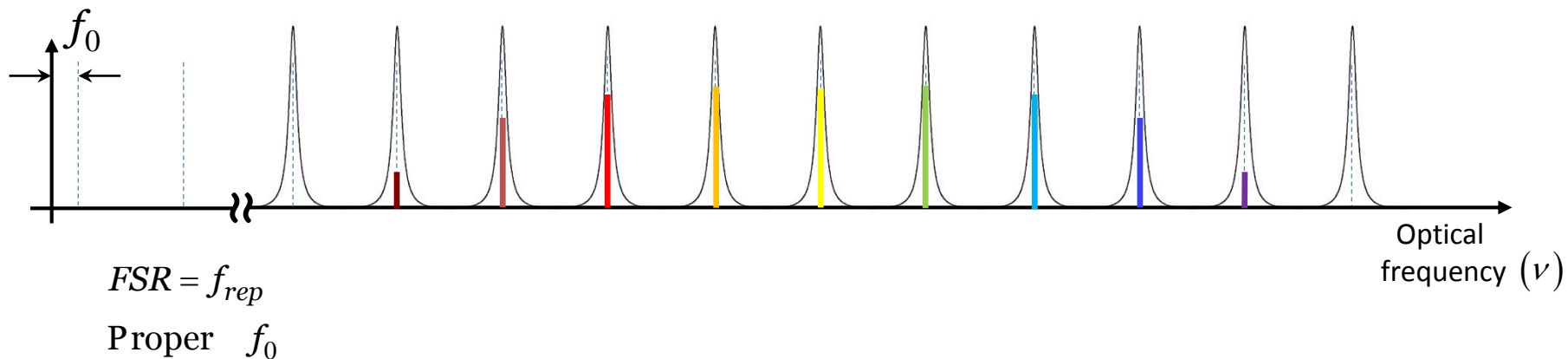
→ Electrical Connection

→ Pipeline Connection

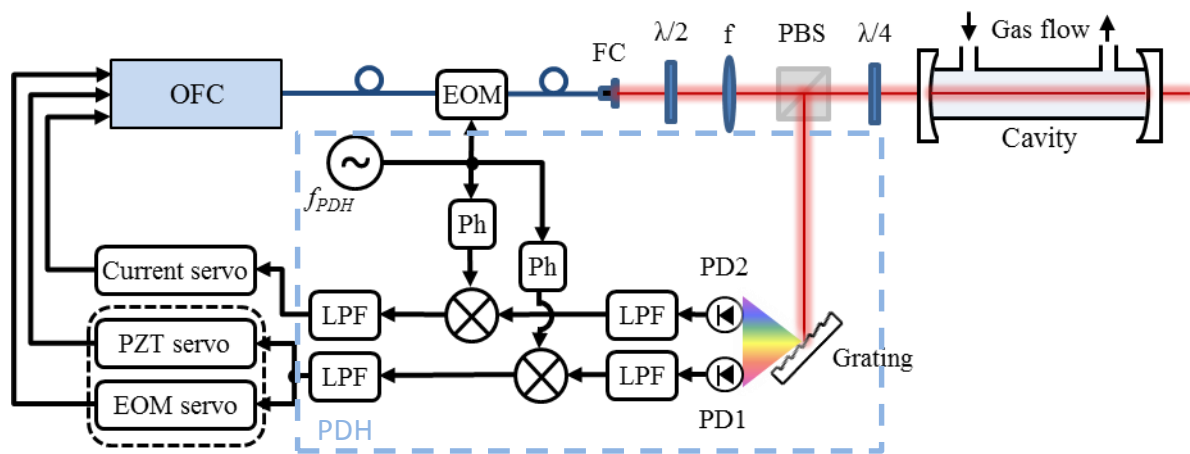
OFC and Enhancement Cavity



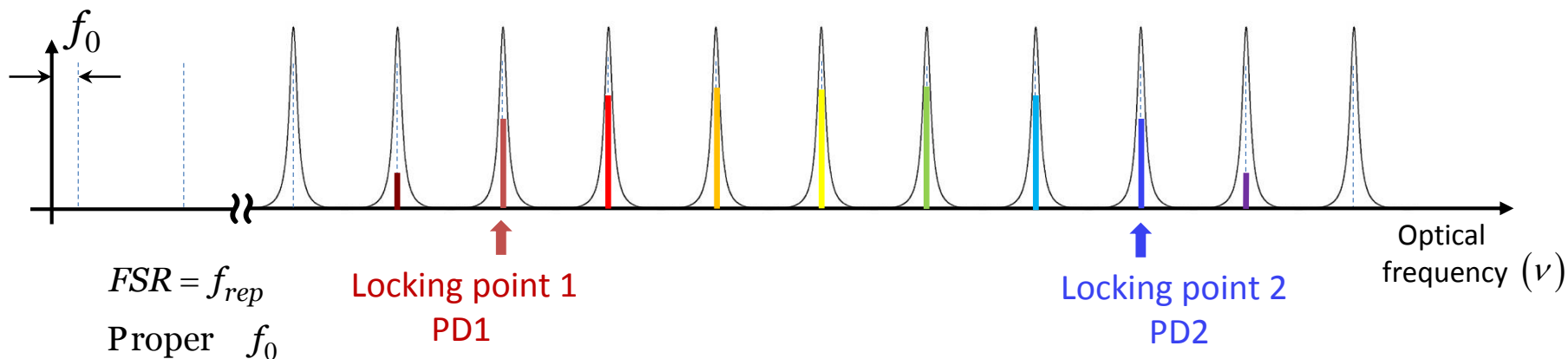
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Two point PDH locking:

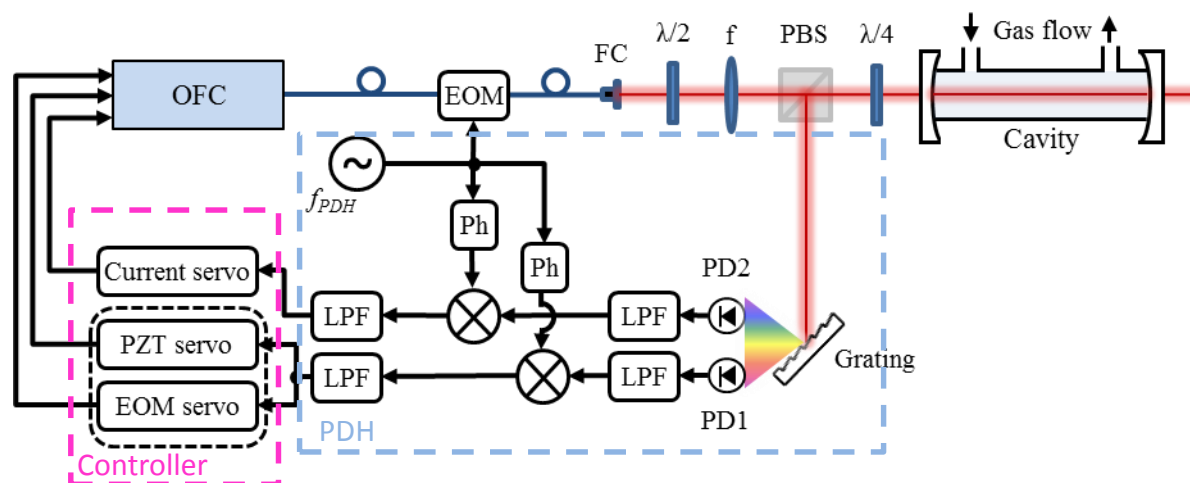


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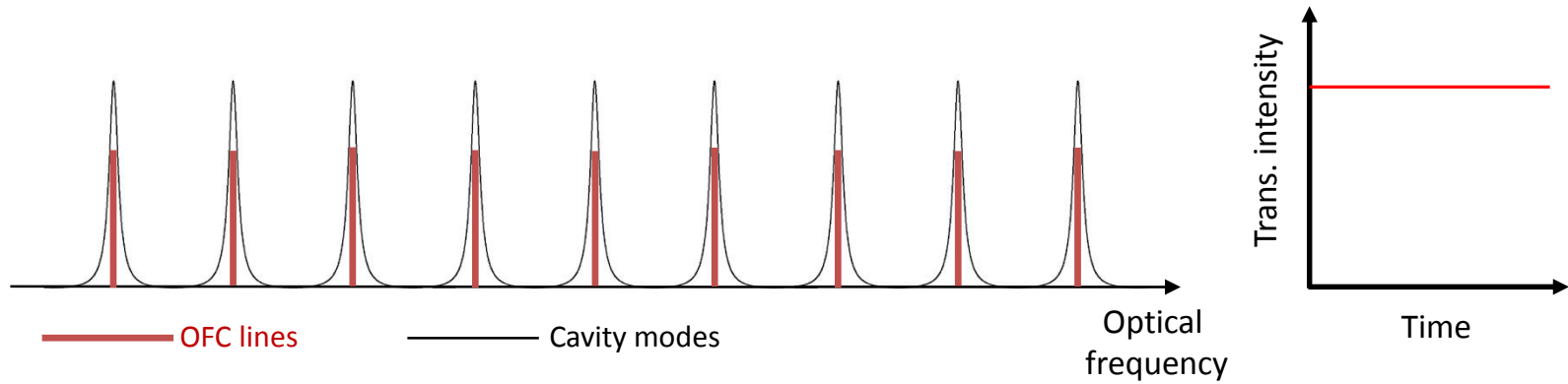
Two point PDH locking:

- Locking point 1 acts on f_0
- Locking point 2 acts on f_{rep}



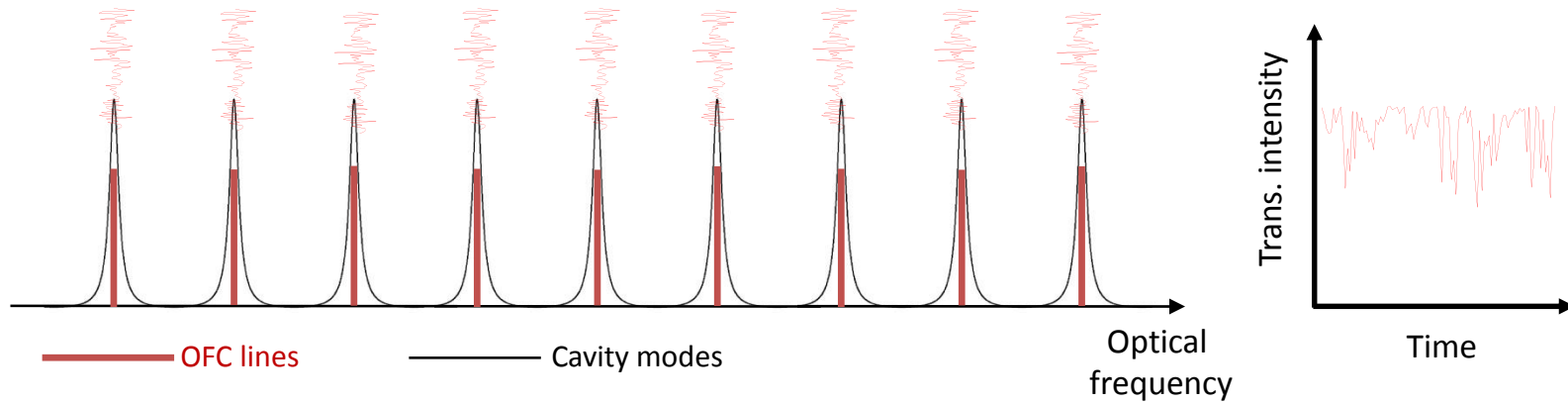
Noise Sources

Frequency to Amplitude Noise Conversion



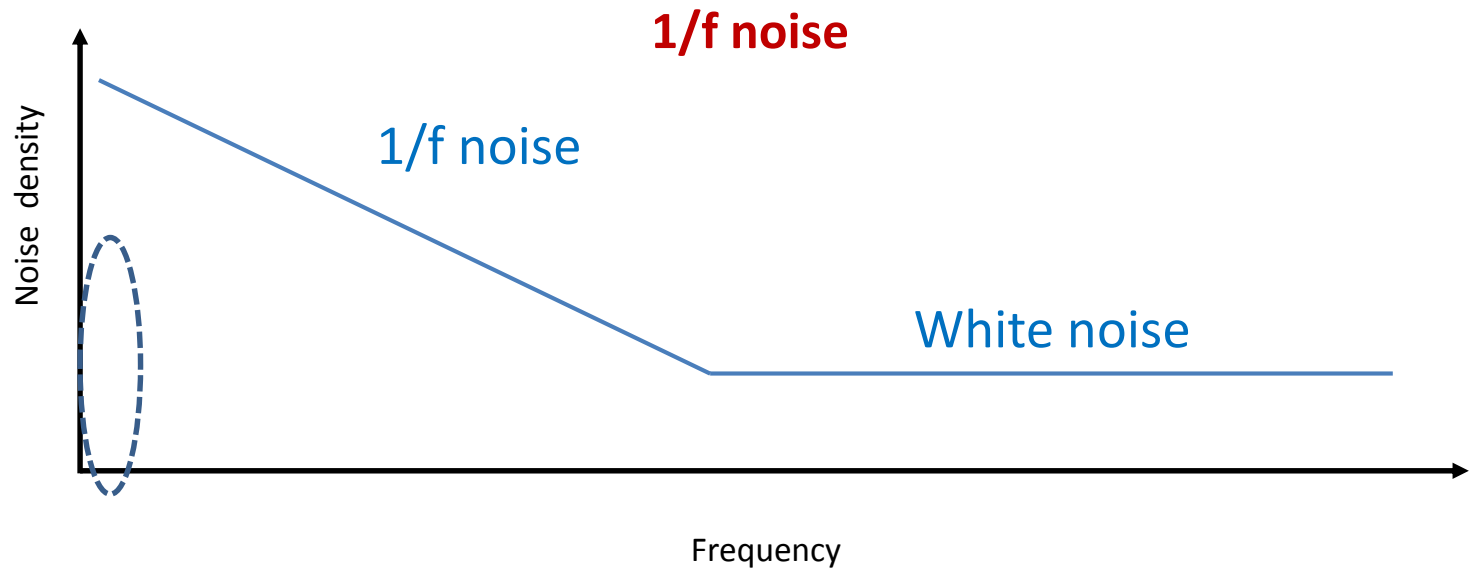
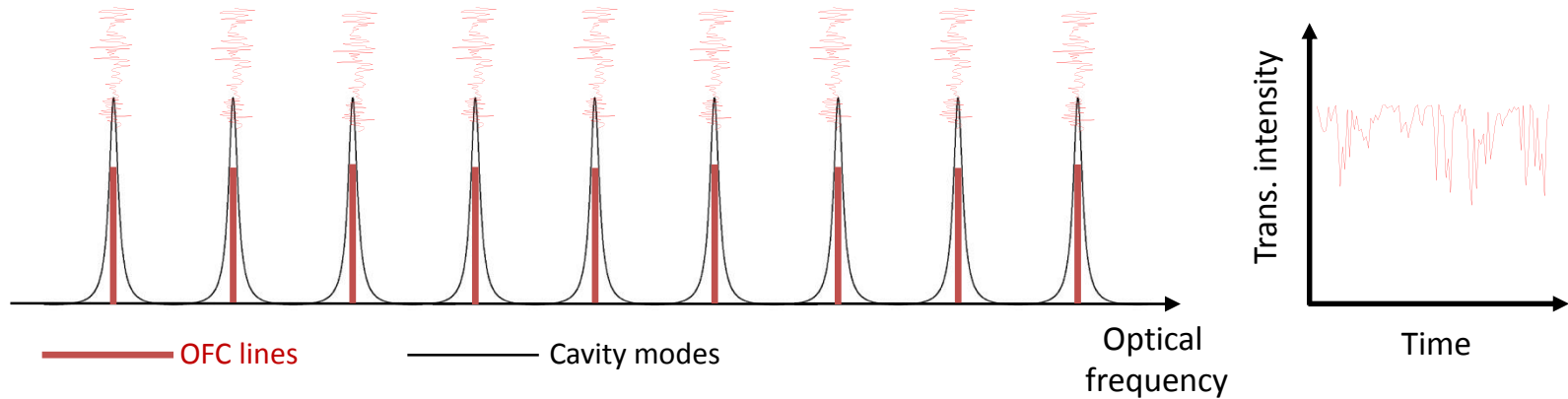
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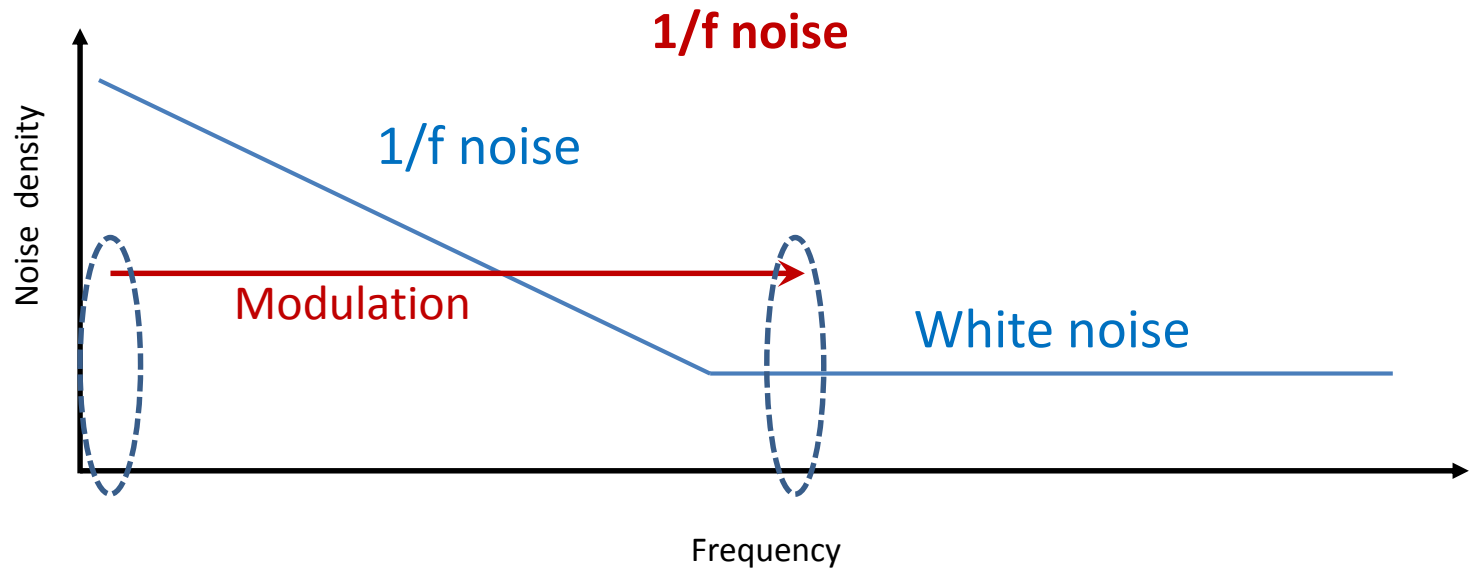
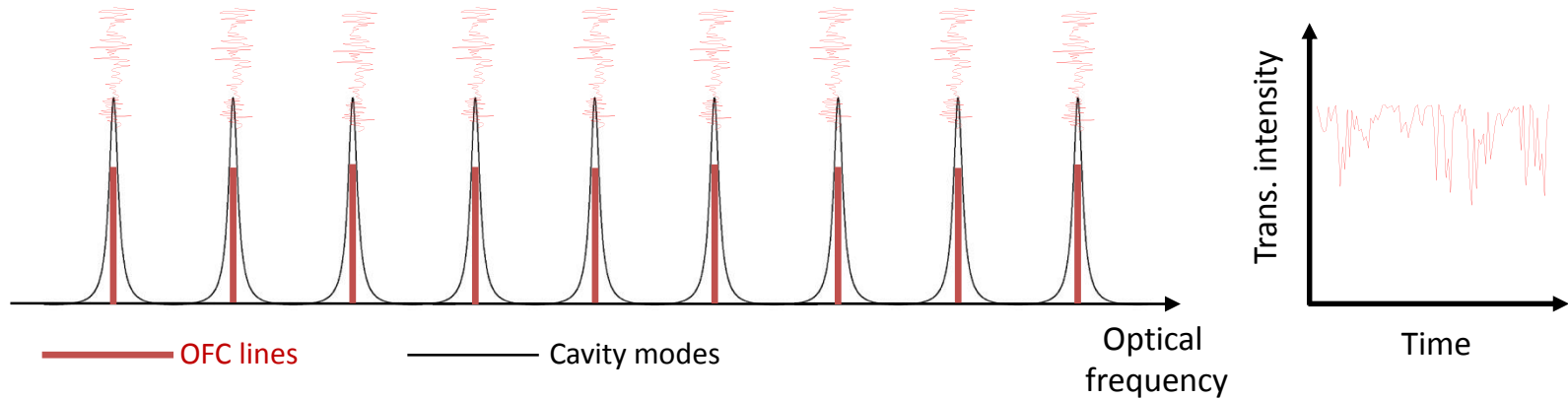
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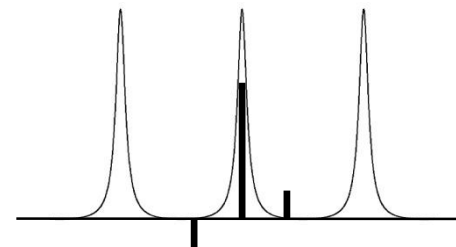
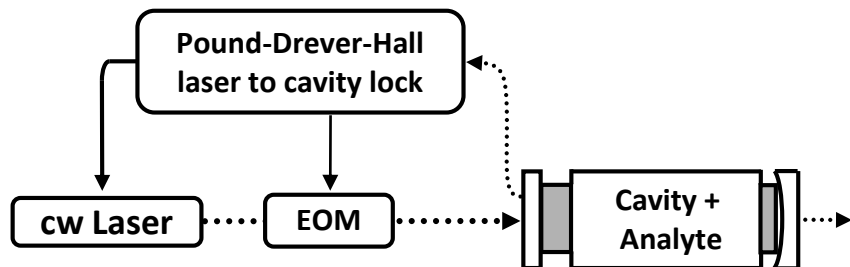
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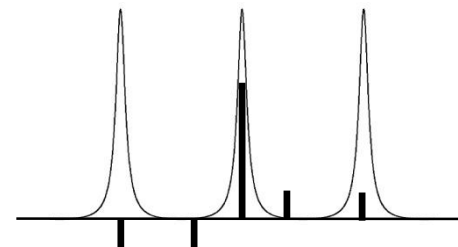
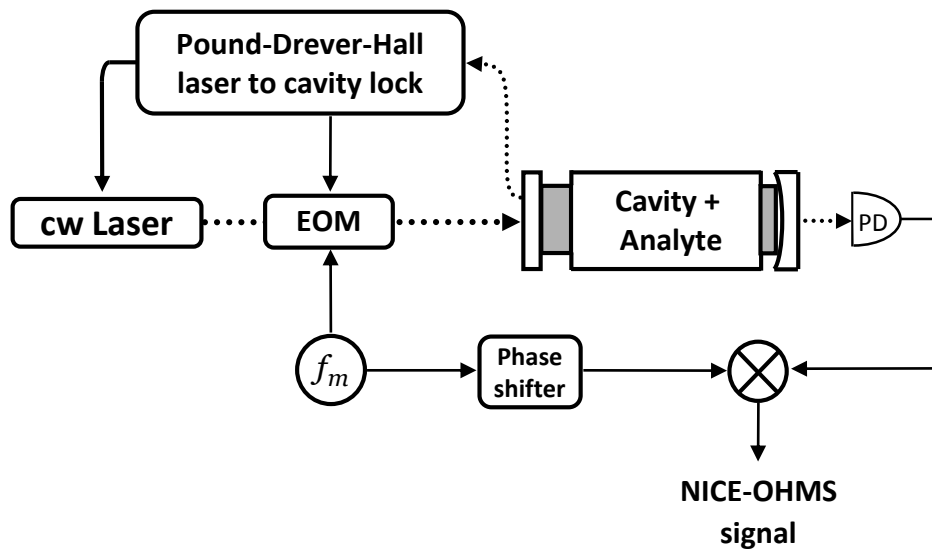
NICE-OHMS

Noise-Immune Cavity-Enhanced Optical Heterodyne Molecular Spectroscopy
 (Alternative name: Cavity-Enhanced Frequency Modulation Spectroscopy)



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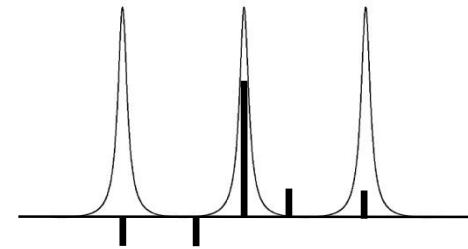
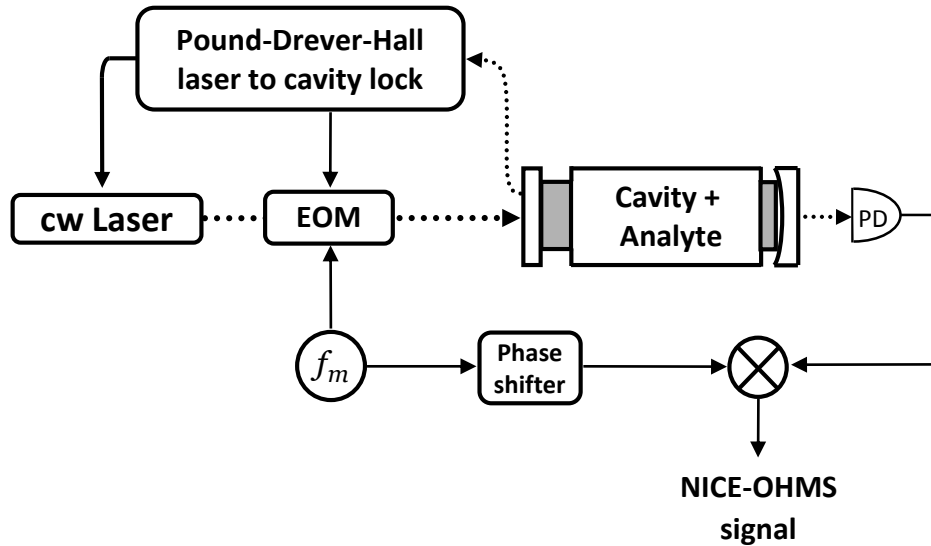
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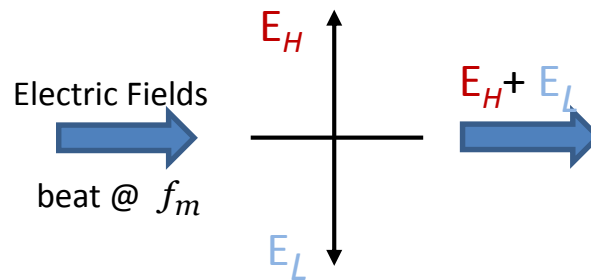
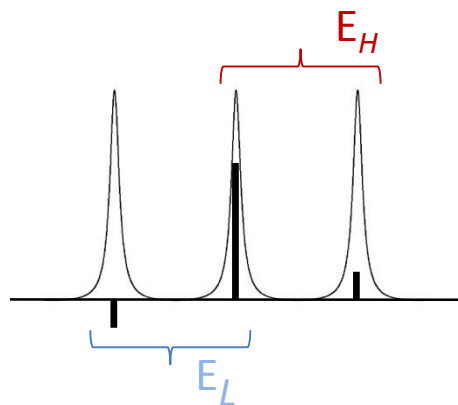
Noise Immunity: $f_m = qFSR$

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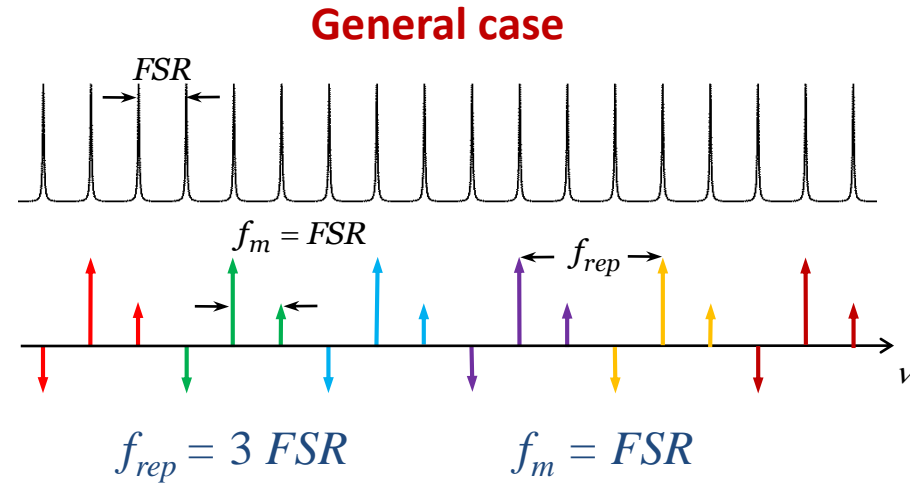
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Frequency to amplitude
noise immune signal

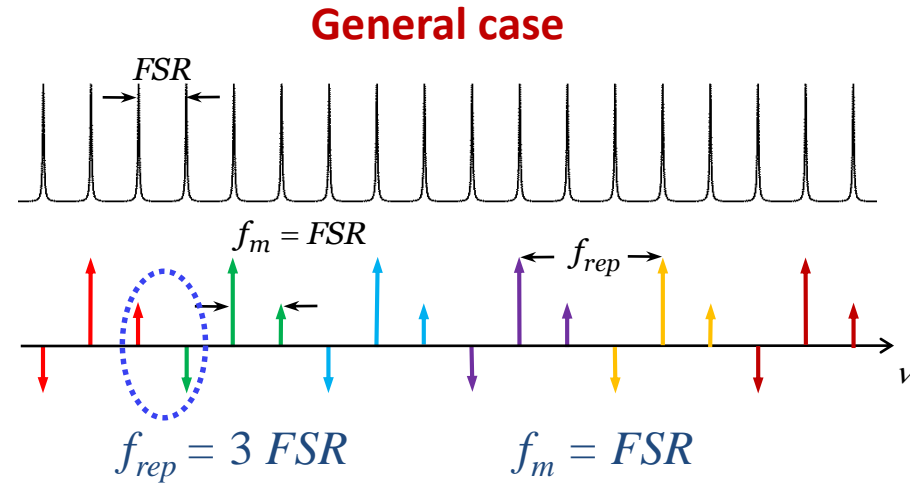
Comb-Cavity Matching

- Impractically long linear cavity for typical OFC sources
(e.g. 1.8 m for f_{rep} 250 MHz)



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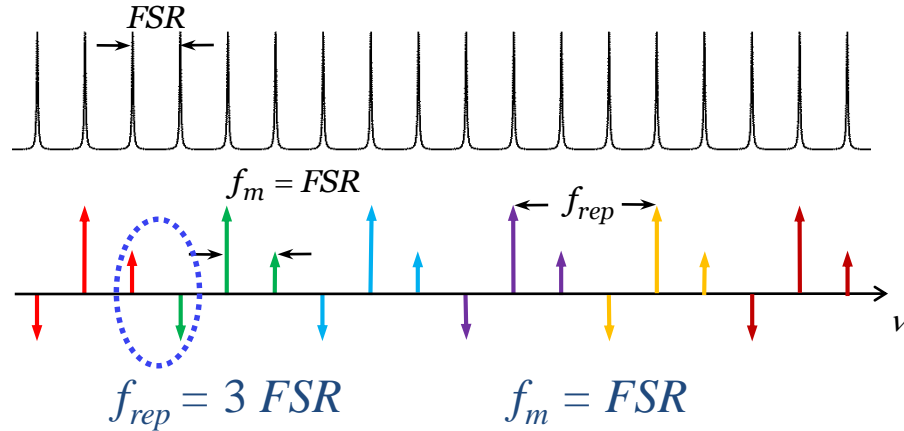
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- Instability/cross-talk from sideband-sideband beatings



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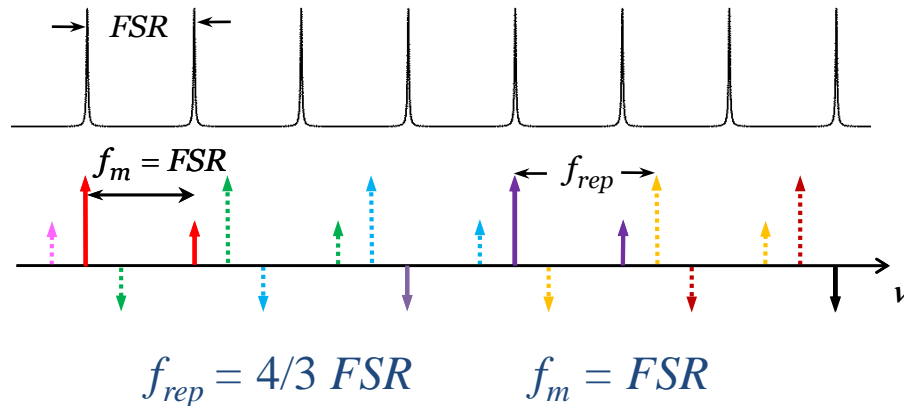
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General case



- Shorter linear cavity (80 cm)

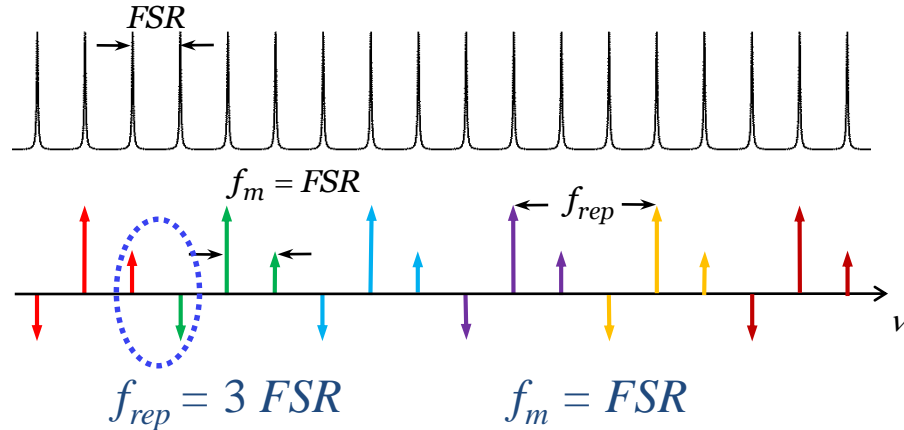
Filter solution



Comb-Cavity Matching

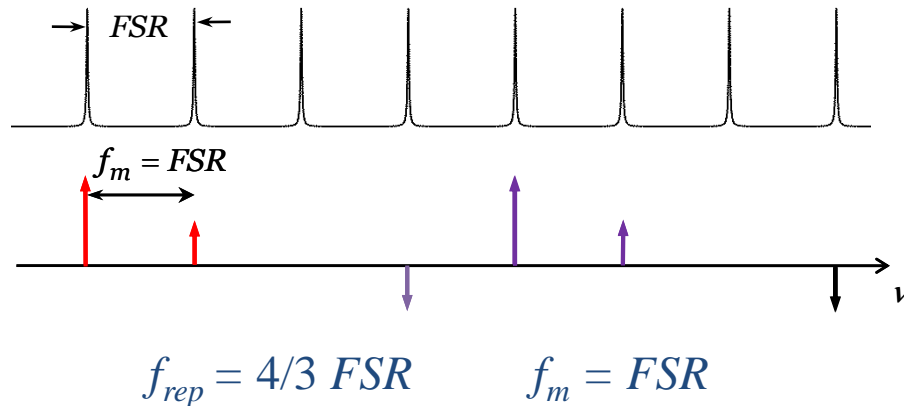
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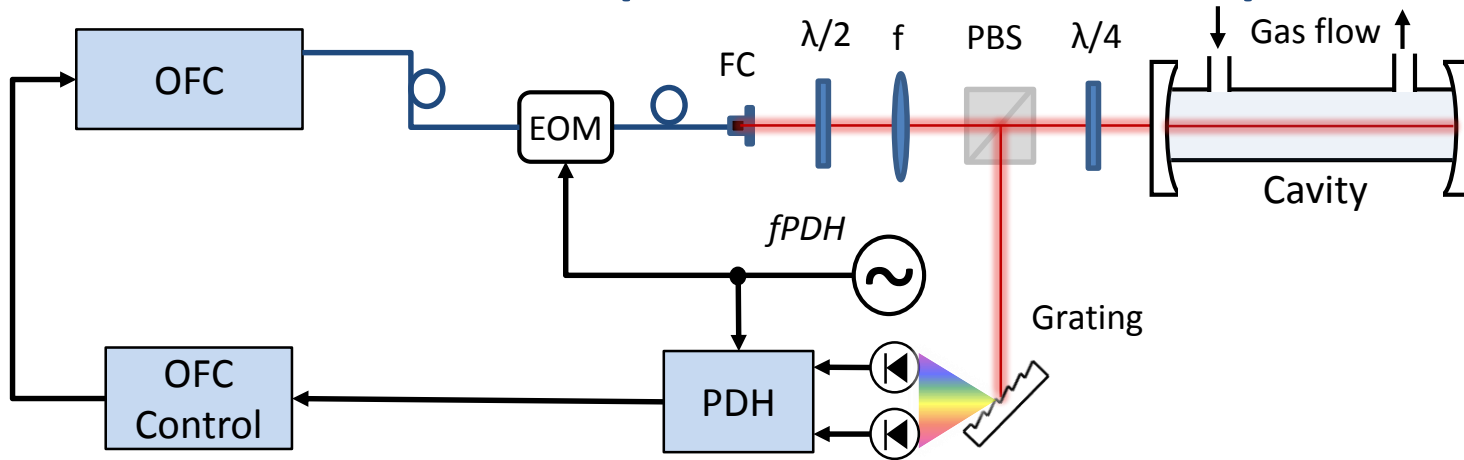


- Shorter linear cavity (80 cm)
- Lower transmitted power
- No sideband-sideband beatings – higher stability

Filter solution



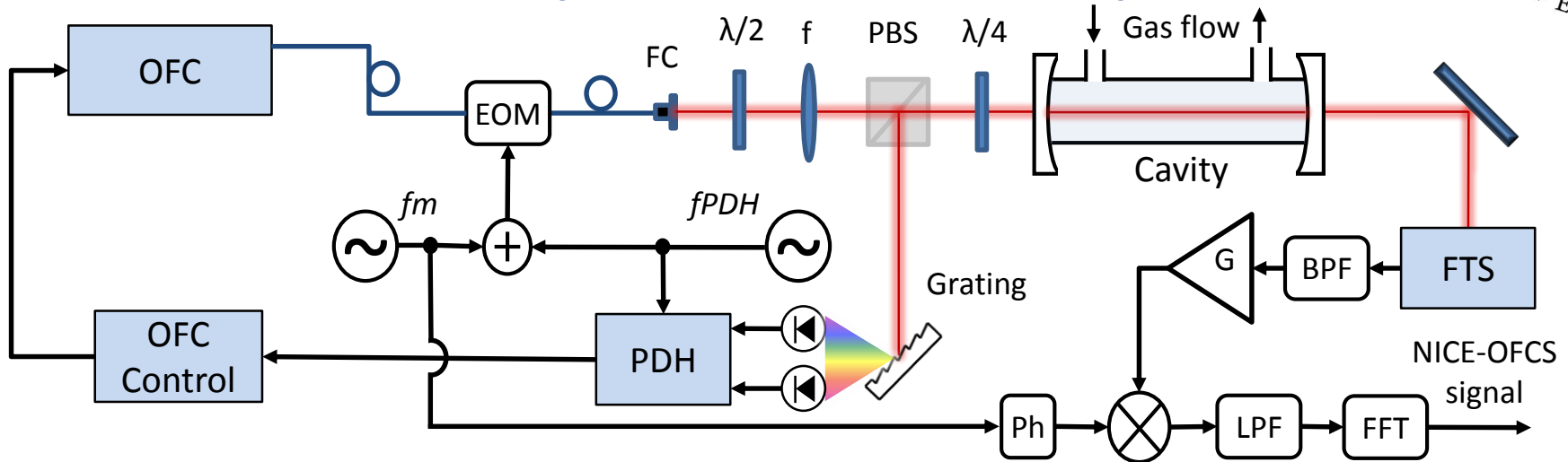
Experimental Setup



- Er:fiber femtosecond laser:
1.5-1.6 μm , 250 MHz repetition rate, 120 mW
- Cavities:
finesse ~ 1100 / ~ 9000 , length 80 cm, FSR 187 MHz
- Two-point Pound-Drever-Hall lock

OFC – optical frequency comb
 EOM – electro-optic modulator
 FC – fiber collimator
 PBS – polarizing beam splitter
 FTS – Fourier transform spectrometer
 BPF – band-pass filter
 LPF – low-pass filter
 FFT – fast Fourier transform
 Ph – phase shifter
 DDS – direct digital synthesizer
 PDH – Pound-Drever-Hall locking electronics
 f_{PDH} – PDH modulation frequency
 f_m – NICE-OFCS modulation frequency.

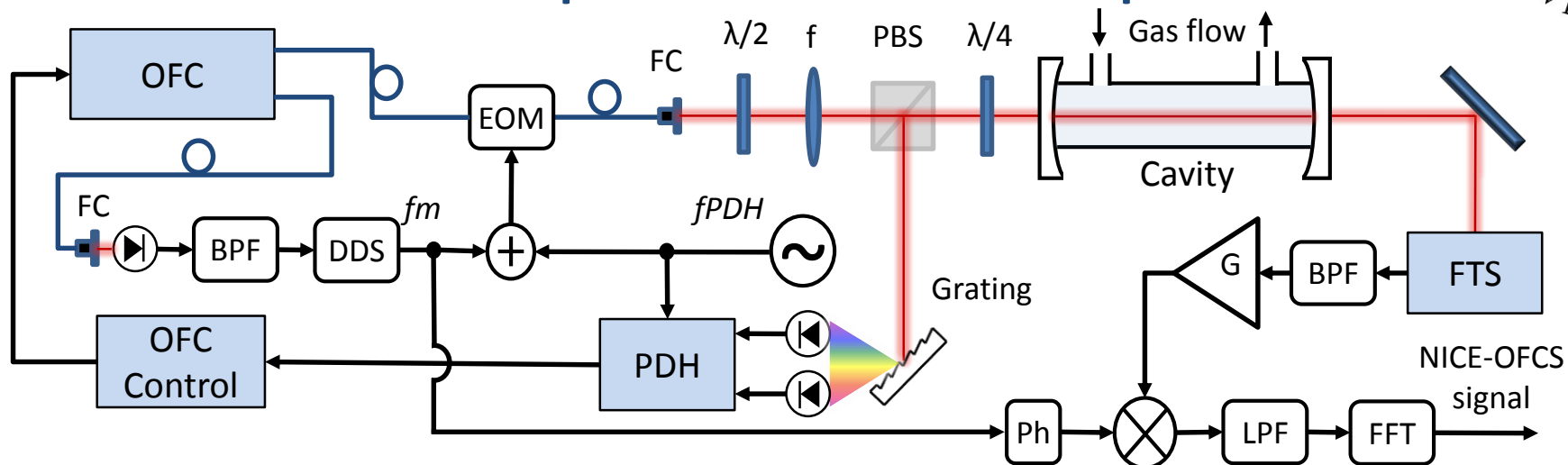
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0.8 m/s OPD Scan, ~ 0.5 s measurement time for 750 MHz resolution
- Synchronous demodulation and FFT

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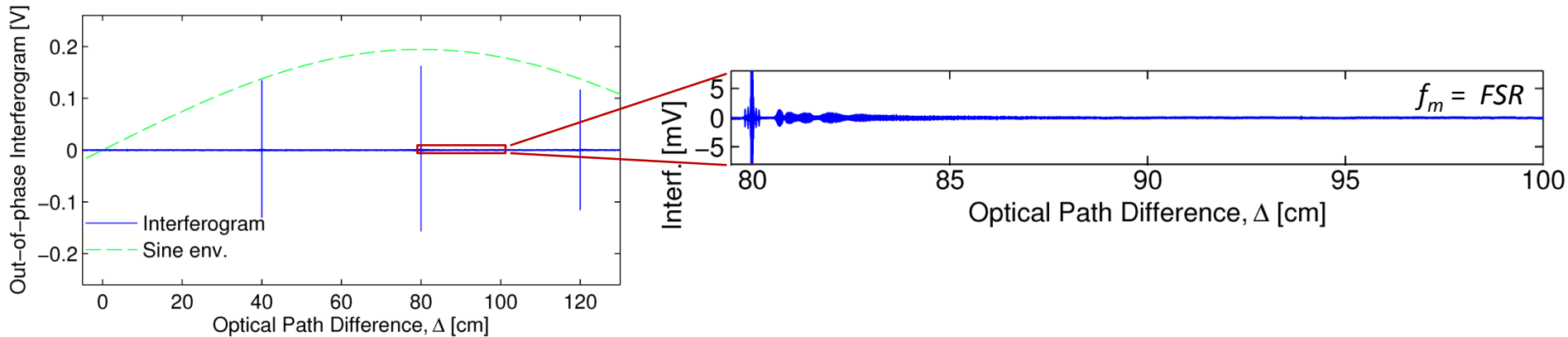
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- Passive lock of the f_m to the cavity FSR using f_{rep} clocked DDS

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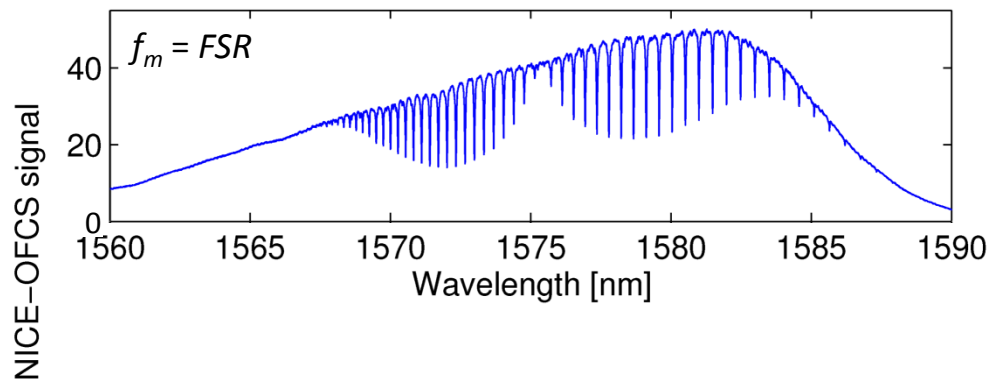
Spectra and Noise Immunity

OPD domain interferogram

- Absorption features clearly visible in the interferogram



After FFT

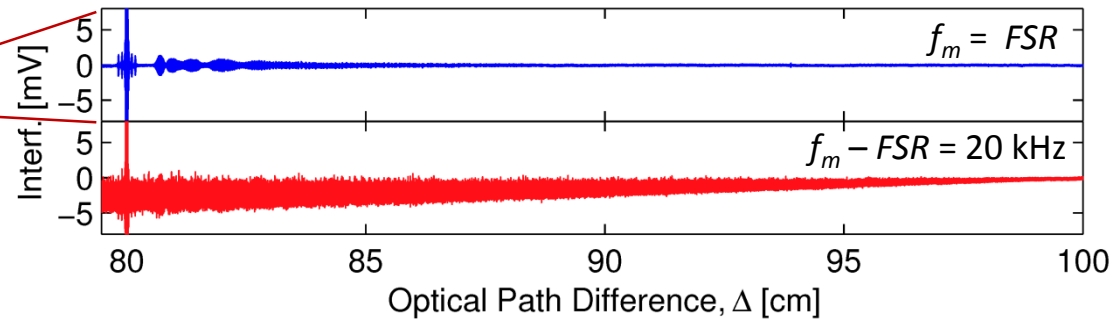
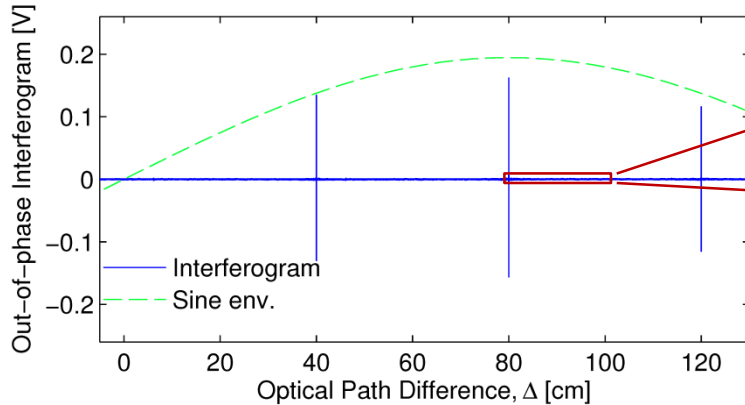


- 1% CO₂ in 500 Torr N₂
- Cavity finesse: **~1100**
- Spectral Bandwidth: **40 nm**
- Spectral resolution: **750 MHz**
- Acquisition time: **0.5 s**

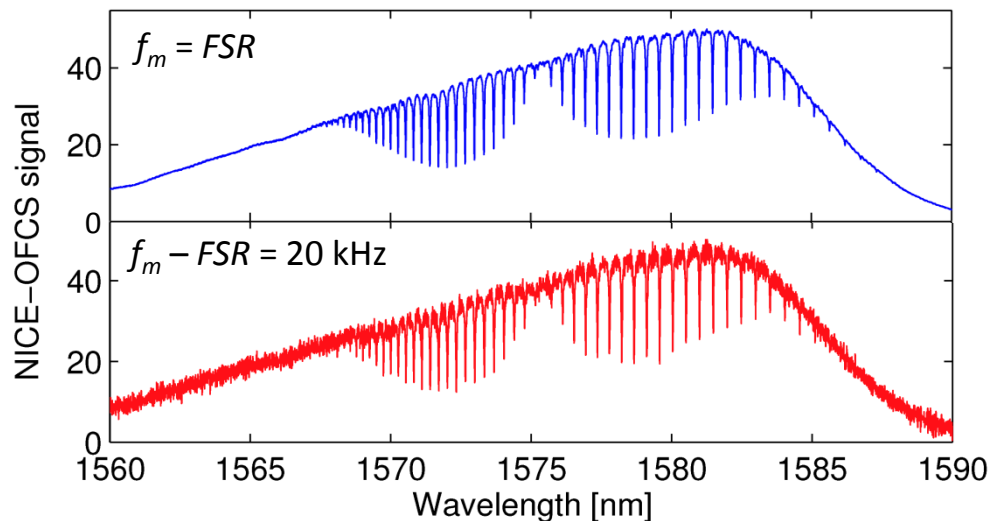
Spectra and Noise Immunity

OPD domain interferogram

- Absorption features clearly visible in the interferogram
- Mismatch of f_m and FSR declines the noise immunity and decreases the SNR



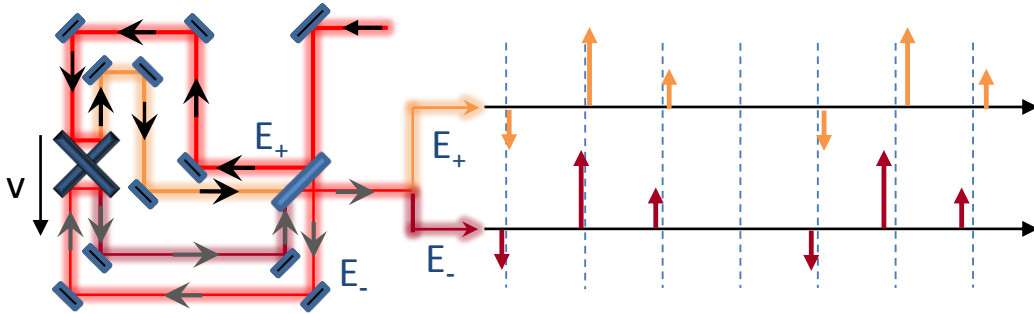
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Signal modelization

Electric field: comb modulated at f_m and Doppler shifted by v/c

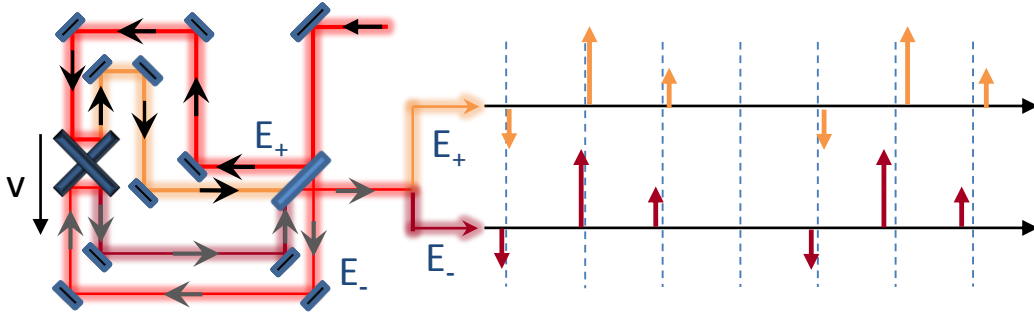


$$(\omega_n \pm \omega_m) \left(1 + \frac{2v}{c}\right)$$

$$(\omega_n \pm \omega_m) \left(1 - \frac{2v}{c}\right)$$

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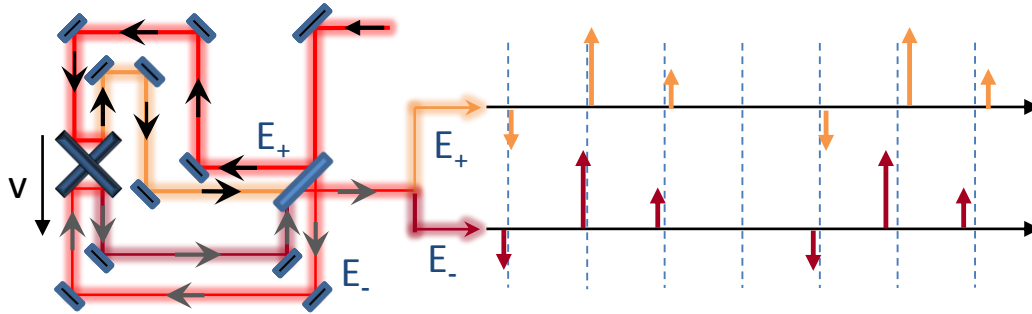
Doppler shift \downarrow

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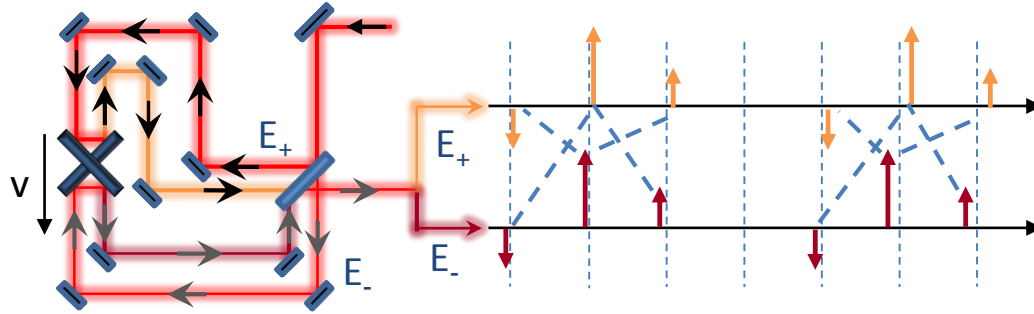
Electric field: comb modulated at f_m and Doppler shifted by v/c



$$E_{\pm} = \sum_n \sum_{k=-1,0,1} \frac{E_n}{4} J_k(\beta) T_{n,k} e^{i[(\omega_n + k\omega_m)(t \pm \frac{\Delta}{c})]} + \text{c. c.}$$

Signal modelization

Electric field: comb modulated at f_m and Doppler shifted by v/c



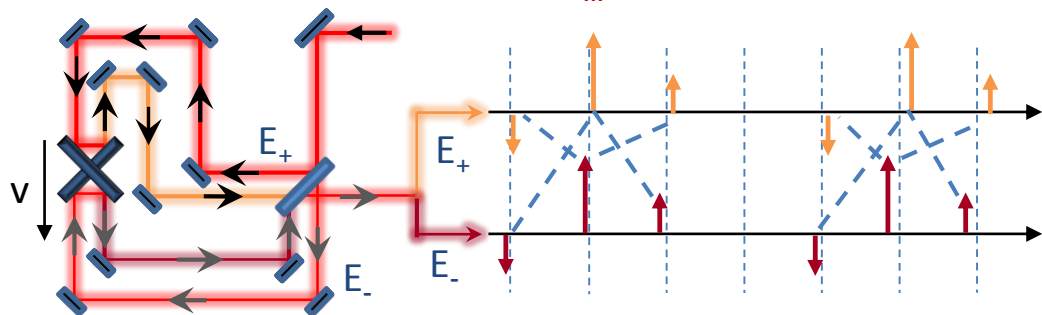
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Intensity: beating of the two combs demodulated at f_m and $\phi = \pi / 2$

$$I_{\omega_m} = J_0(\beta) J_1(\beta) \sum_n I_n \left\{ \begin{array}{l} \cos\left(\omega_n \frac{\Delta}{c}\right) \cos\left(\omega_m \frac{\Delta}{2c}\right) \text{Re}(T_{n,0} T_{n,-1}^* - T_{n,0}^* T_{n,+1}) \\ + \sin\left(\omega_n \frac{\Delta}{c}\right) \sin\left(\omega_m \frac{\Delta}{2c}\right) \text{Re}(T_{n,0} T_{n,-1}^* + T_{n,0}^* T_{n,+1}) \end{array} \right\}$$

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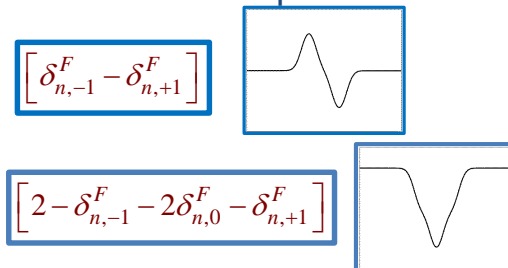


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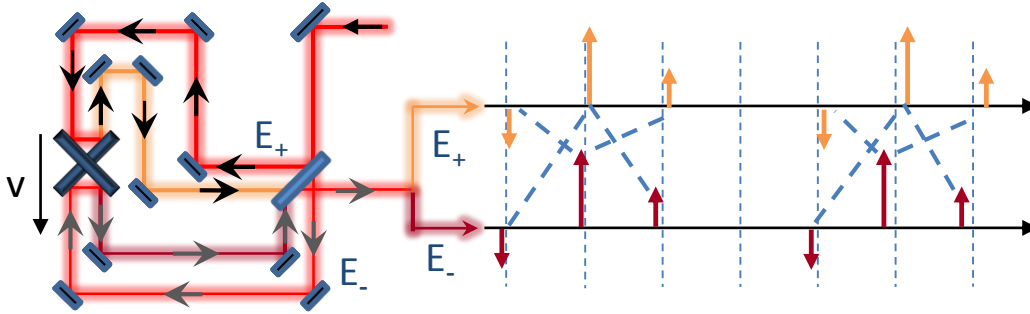
For weak absorption lines:



- Direct cavity-enhanced absorption like signal which makes NICE-OFCS calibration-free

Signal modelization

Electric field: comb modulated at f_m and Doppler shifted by v/c

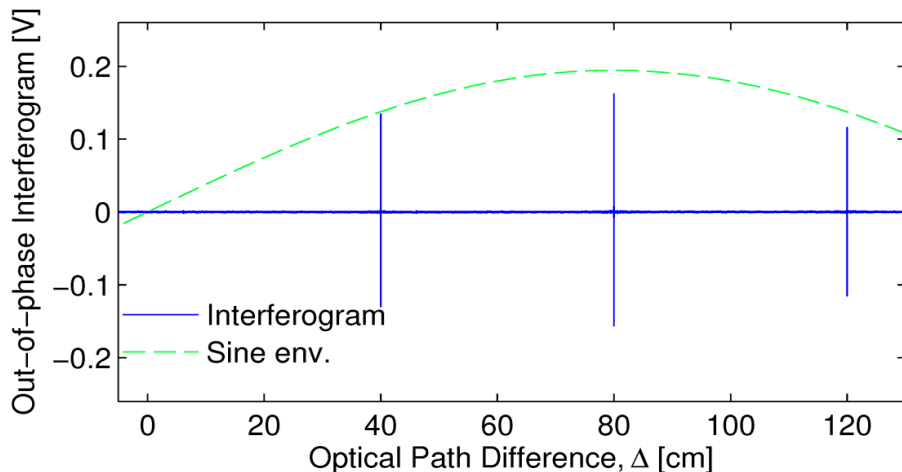
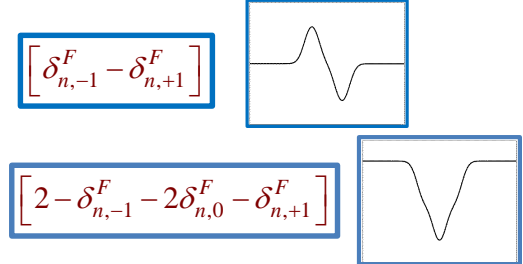


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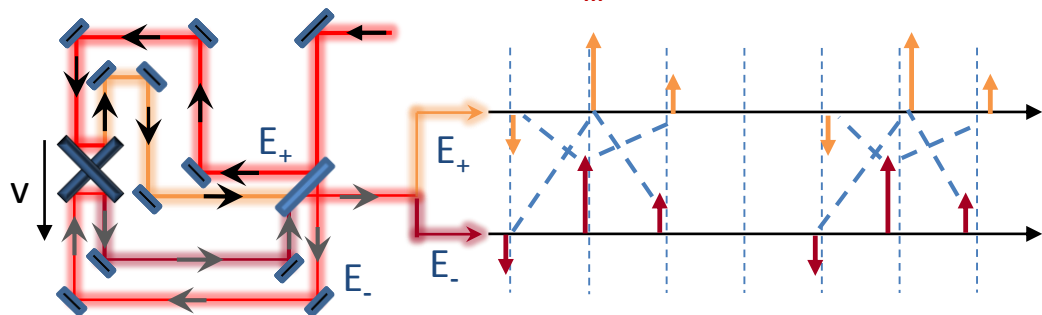
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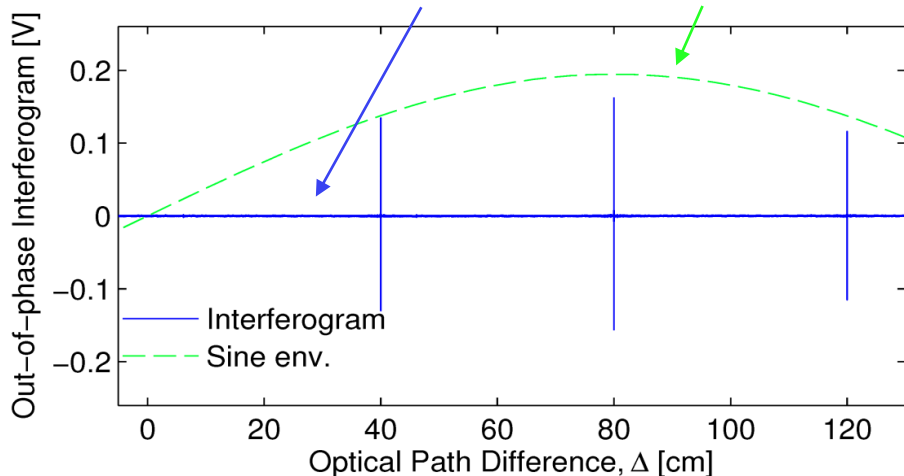
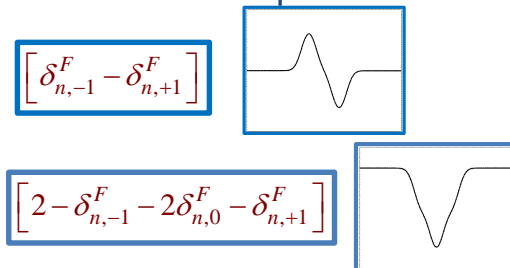


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$$I_{\omega_m} = J_0(\beta) J_1(\beta) \sum_n I_n \left\{ \begin{array}{l} \text{Cosine envelope} \\ \cos\left(\omega_n \frac{\Delta}{c}\right) \cos\left(\omega_m \frac{\Delta}{2c}\right) \text{Re}(T_{n,0} T_{n,-1}^* - T_{n,0}^* T_{n,+1}) \\ \text{Interferogram} \\ + \sin\left(\omega_n \frac{\Delta}{c}\right) \sin\left(\omega_m \frac{\Delta}{2c}\right) \text{Re}(T_{n,0} T_{n,-1}^* + T_{n,0}^* T_{n,+1}) \\ \text{Sine envelope} \end{array} \right\}$$

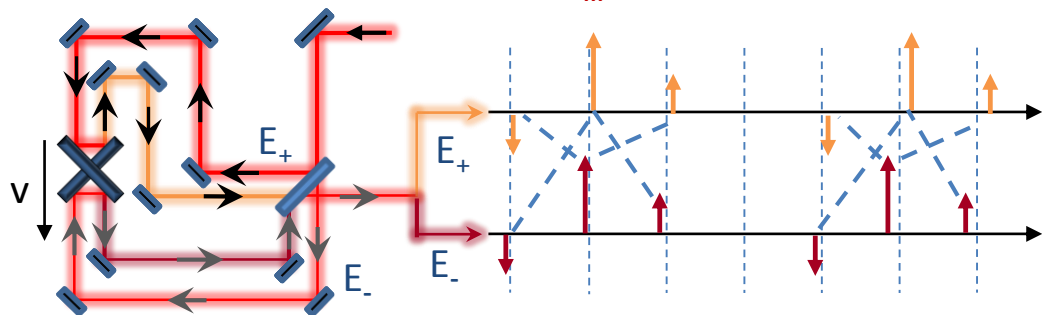
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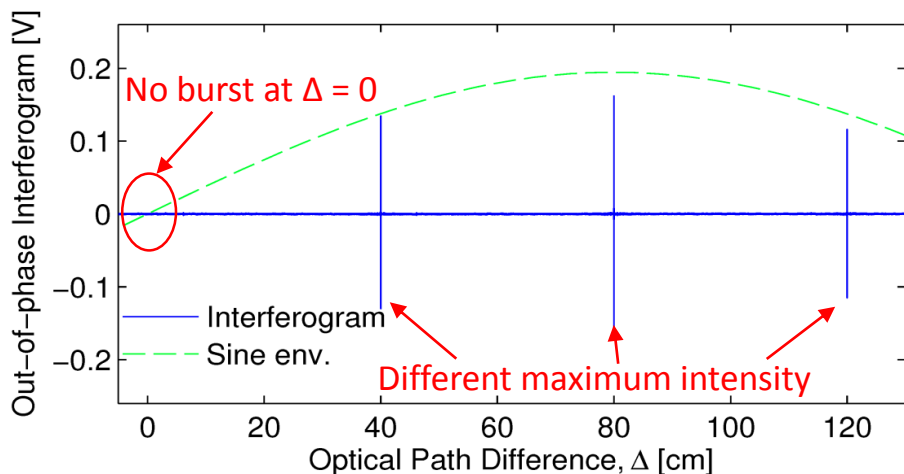
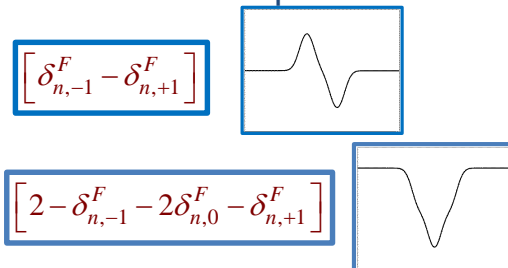


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$$I_{\omega_m} = J_0(\beta) J_1(\beta) \sum_n I_n \left\{ \begin{array}{l} \text{Cosine envelope} \\ \cos\left(\omega_n \frac{\Delta}{c}\right) \cos\left(\omega_m \frac{\Delta}{2c}\right) \text{Re}(T_{n,0} T_{n,-1}^* - T_{n,0}^* T_{n,+1}) \\ \text{Interferogram} \quad \text{Green envelope} \\ + \sin\left(\omega_n \frac{\Delta}{c}\right) \sin\left(\omega_m \frac{\Delta}{2c}\right) \text{Re}(T_{n,0} T_{n,-1}^* + T_{n,0}^* T_{n,+1}) \end{array} \right\}$$

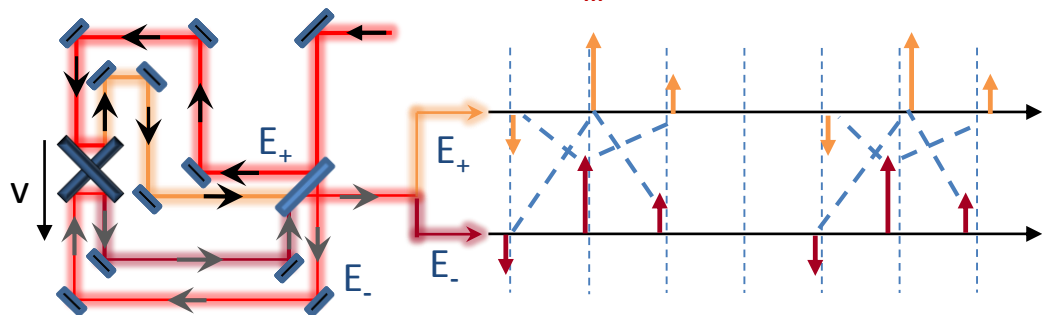
For weak absorption lines:



- Direct cavity-enhanced absorption like signal which makes NICE-OFCS calibration-free
- Interferogram intensity ponderated by the envelopes induced by the modulation frequency

Signal modelization

Electric field: comb modulated at f_m and Doppler shifted by v/c

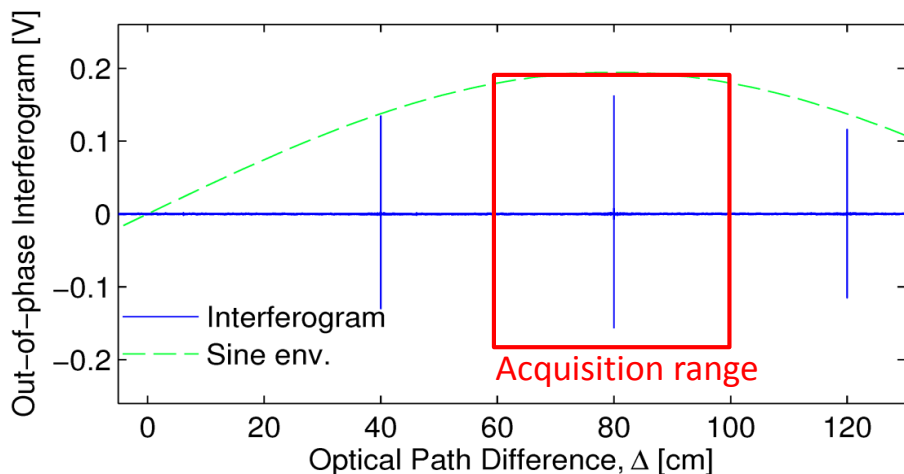
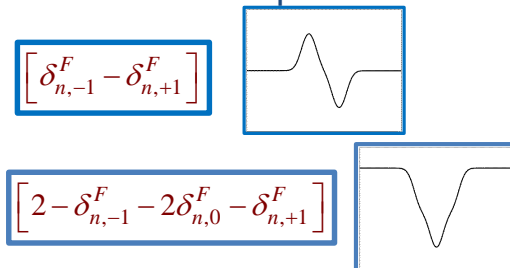


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Absorption lineshape model

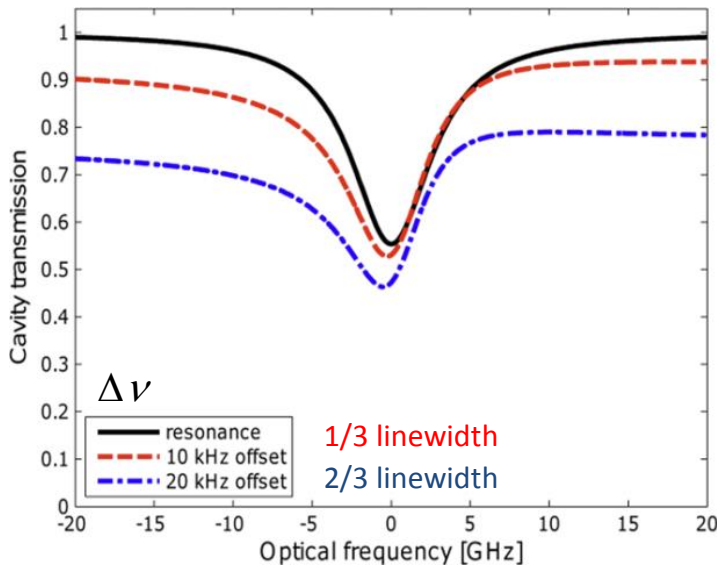
Transmitted intensity

$$T_{n,k}(\nu) = \frac{T^2(\nu)e^{-\alpha(\nu)L}}{1 + R^2(\nu)e^{-2\alpha(\nu)L} - 2R(\nu)e^{-\alpha(\nu)L}\cos[\phi(\nu)L + \varphi(\nu)]}$$

Molecular absorption

Molecular phase shift

Round trip intracavity phase shift

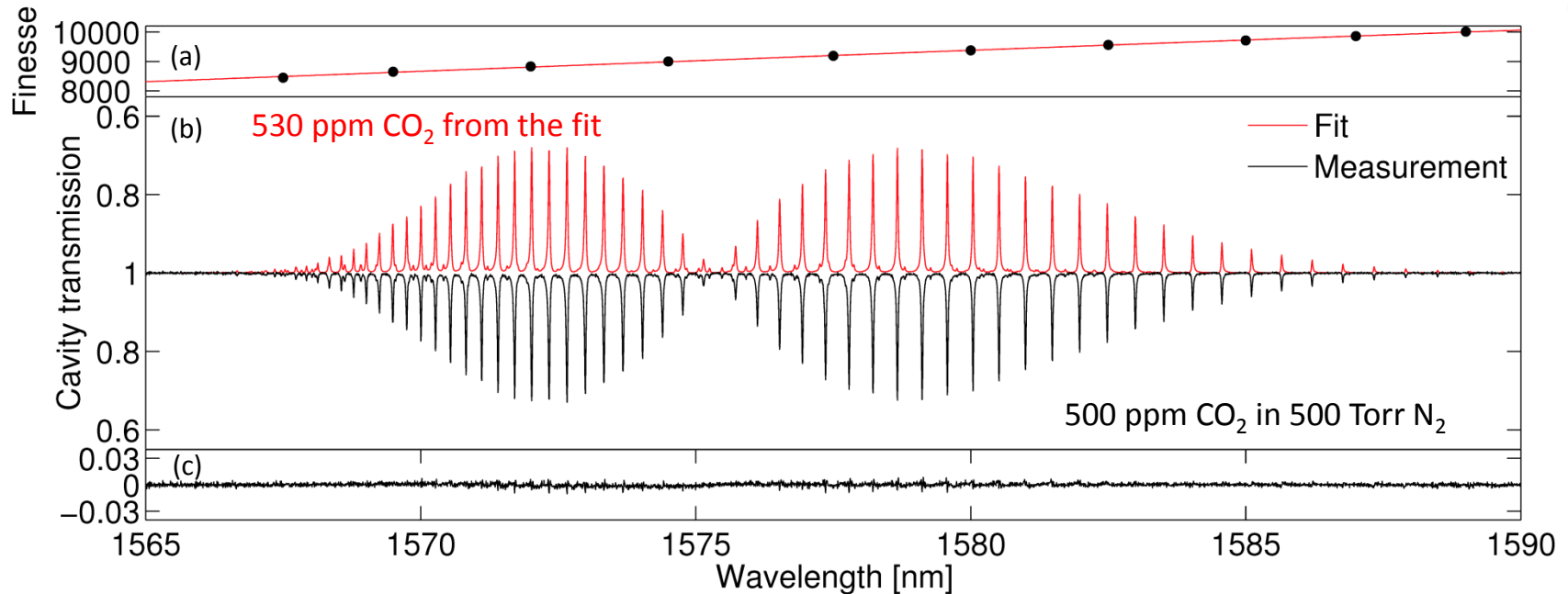


$$\varphi(\nu) = 4\pi \nu \frac{L}{c} = \frac{2\pi\nu}{FSR}$$

$$\varphi(\Delta\nu) = 2n\pi + 2\pi \frac{\Delta\nu}{FSR}$$

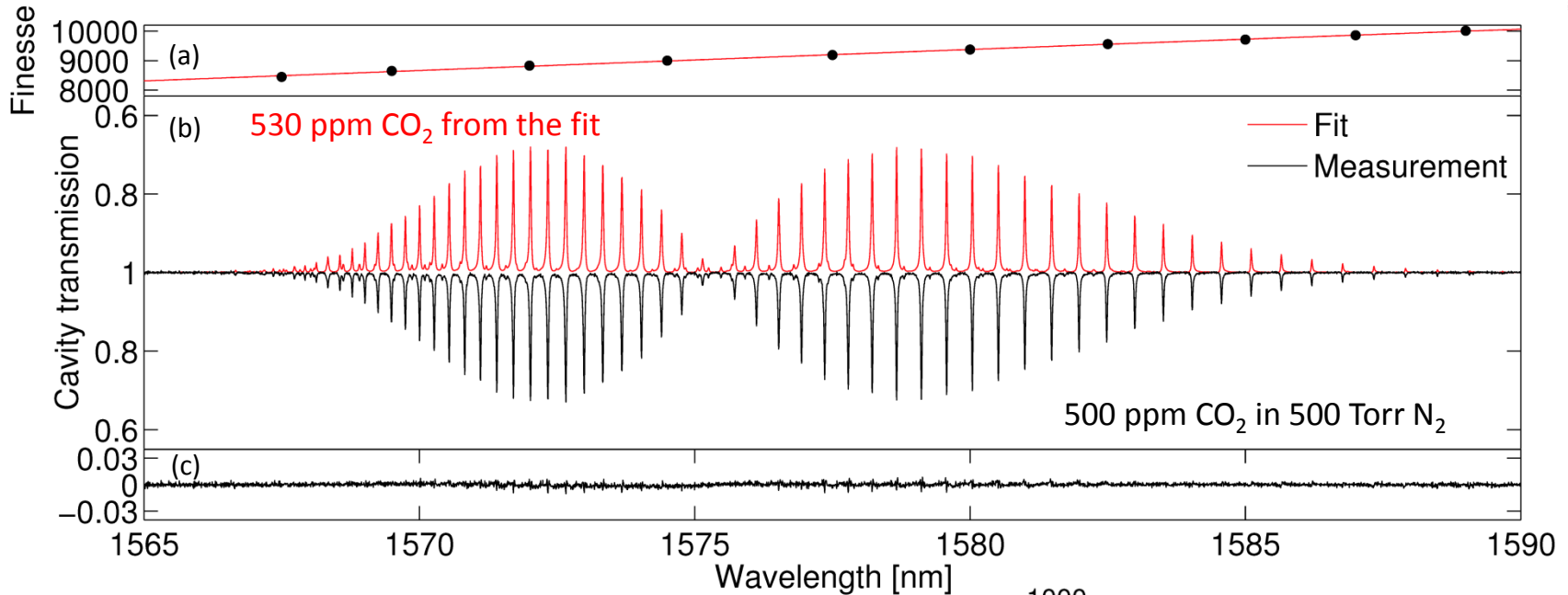
T – mirror transmission
 R – mirror reflection
 L – cavity length

Sensitivity and Detection Limit

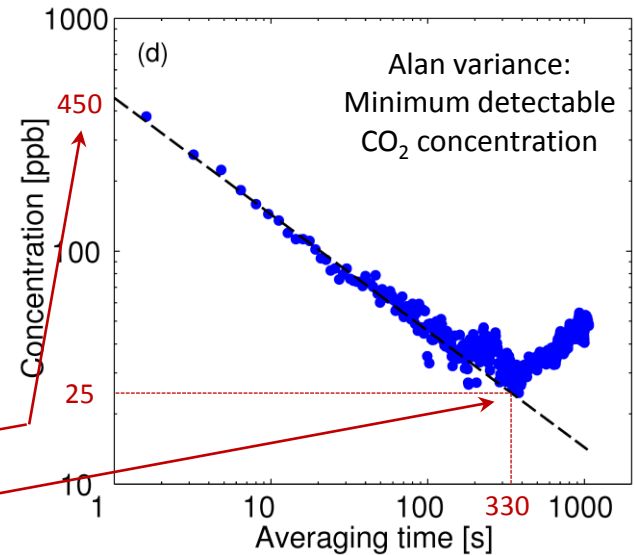


- 500 ppm CO₂ in 500 Torr N₂
- Cavity finesse: ~ 9000
- Spectral Bandwidth: 40 nm
- Spectral resolution: 750 MHz
- Acquisition time: 0.5 s
- Noise equivalent absorption sensitivity:
 $6.4 \times 10^{-11} \text{ cm}^{-1} \text{ Hz}^{-1/2}$ per spectral element

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- CO₂ detection limit (multiline fitting):
 $450 \text{ ppb Hz}^{-1/2}$
 25 ppb after 330 s



Conclusions

- FT-based NICE-OFCS : broadband, highly sensitive, high resolution technique with a short acquisition time
- Calibration-free technique due to the existence of signal background (for a known cavity finesse)
- Stable, long term noise immune operation achieved with a simple passive lock
- Compatible with commercial FTIR instruments using a high bandwidth detector
- Standard and commercially available components
- Outlook: Improved model of the spectrum to decrease the concentration discrepancy



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Thank you for your attention!