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

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✓ Optical Frequency Comb

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## Laser-based Spectroscopic Technique

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- ✓ Cavity Enhancement
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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

OFC Control

Modulation

Cavity

FTS

Gas

2P-PDH

Demodulation and FFT

NICE-OFCS signal

Optical Connection
Electrical Connection
Pipeline Connection
OFC and Enhancement Cavity

\[ FSR = \frac{c}{2L} \]

\[ FSR = f_{\text{rep}} \]

Proper \( f_0 \)

OFC and Enhancement Cavity

Two point PDH locking:

FSR = \( f_{rep} \)
Proper \( f_0 \)

\[ f_{PDH} \]

OFC and Enhancement Cavity

Two point PDH locking:
- Locking point 1 acts on $f_0$
- Locking point 2 acts on $f_{rep}$

$FSR = f_{rep}$

Proper $f_0$

Noise Sources

Frequency to Amplitude Noise Conversion

- OFC lines
- Cavity modes
- Optical frequency
- Trans. intensity
- Time
Noise Sources

Frequency to Amplitude Noise Conversion

- OFC lines
- Cavity modes
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Frequency to Amplitude Noise Conversion

1/f noise

White noise
Noise Sources

Frequency to Amplitude Noise Conversion

OFC lines | Cavity modes
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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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Noise Immunity: \[ f_m = q \text{FSR} \]

NICE-OHMS

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(Alternative name: Cavity-Enhanced Frequency Modulation Spectroscopy)

Noise Immunity: \( f_m = q \text{FSR} \)

Electric Fields beat @ \( f_m \)

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)

General case

\[ f_{rep} = 3 \text{ FSR} \quad \text{and} \quad f_m = \text{FSR} \]
Comb-Cavity Matching

- Impractically long linear cavity for typical OFC sources (e.g. 1.8 m for $f_{\text{rep}}$ 250 MHz)
- Instability/cross-talk from sideband-sideband beatings

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- $f_m = \text{ FSR}$

Filter solution

- $f_{rep} = 4/3 \text{ FSR}$
- $f_m = \text{ FSR}$

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Filter solution

\[ f_{\text{rep}} = \frac{4}{3} \text{ FSR} \quad f_m = \text{FSR} \]

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

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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- **Synchronous demodulation and FFT**

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

**Diagram:**
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Spectra and Noise Immunity

**OPD domain interferogram**
- Absorption features clearly visible in the interferogram

After FFT
- 1% CO$_2$ in 500 Torr N$_2$
- Cavity finesse: $\sim 1100$
- 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 $v/c$

\[ \omega_n \pm \omega_m \left(1 \pm \frac{2v}{c}\right) \]
Signal modelization

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

$$\text{Doppler shift} = (\omega_n \pm \omega_m) \left(1 \pm \frac{2v}{c}\right)$$
Signal modelization

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

\[
E_{\pm} = \sum_{n} \sum_{k=-1,0,1} \frac{E_n}{4} J_k(\beta) T_{n,k} e^{i[(\omega_n+k\omega_m)(t\pm\Delta t/c)]} + c.c.
\]
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Intensity: beating of the two combs demodulated at $f_m$ and $\phi = \pi / 2$

$$I_{\omega_m} = J_0(\beta) J_1(\beta) \sum_n I_n \left\{ \cos\left(\omega_n \frac{\Delta}{c}\right) \cos\left(\omega_m \frac{\Delta}{2c}\right) \text{Re}(T_{n,0} T^*_{n,-1} - T^*_{n,0} T_{n,+1}) + \sin\left(\omega_n \frac{\Delta}{c}\right) \sin\left(\omega_m \frac{\Delta}{2c}\right) \text{Re}(T_{n,0} T^*_{n,-1} + T^*_{n,0} T_{n,+1}) \right\}$$
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For weak absorption lines:

$$\begin{bmatrix} \delta^F_{n,-1} - \delta^F_{n,+1} \\ 2 - \delta^F_{n,-1} - 2\delta^F_{n,0} - \delta^F_{n,+1} \end{bmatrix}$$

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

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- Interferogram intensity ponderated by the envelopes induced by the modulation frequency

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Absorption lineshape model

**Transmitted intensity**

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

- **Molecular absorption**
- **Molecular phase shift**
- **Round trip intracavity phase shift**

\[ \phi(\nu) = 4\pi \frac{\nu L}{c} = \frac{2\pi \nu}{FSR} \]

\[ \phi(\Delta \nu) = 2n\pi + 2\pi \frac{\Delta \nu}{FSR} \]

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

Sensitivity and Detection Limit

- 500 ppm CO\textsubscript{2} in 500 Torr N\textsubscript{2}
- Cavity finesse: \( \sim 9000 \)
- 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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- CO\textsubscript{2} detection limit (multiline fitting): 450 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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