

# Quantitative Molecular Spectroscopy in Cavity

Presented by:





## **Technical Group Leadership:**

**Krishnan Parameswaran, Analog Devices Inc., USA**

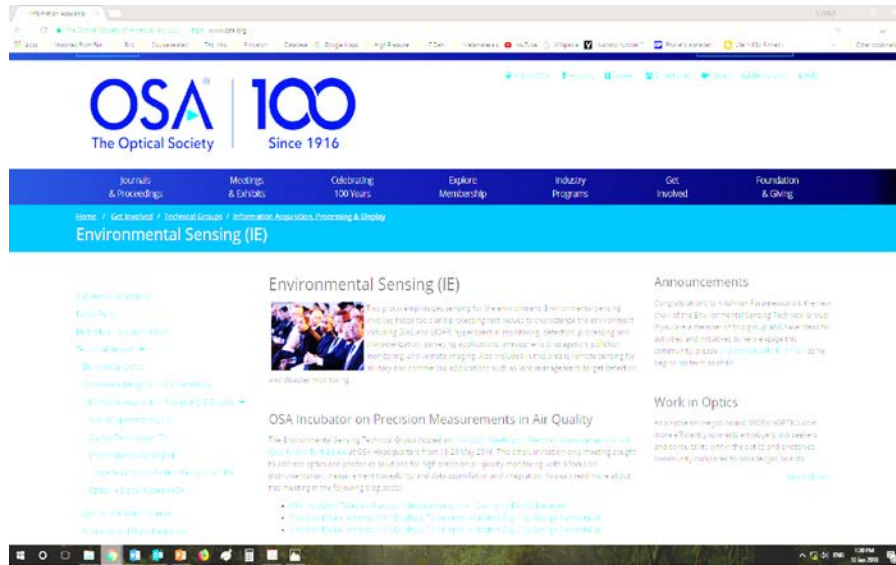
**Joachim Sacher, Sacher Lasertechnik GmbH, Germany**

**Amartya Sengupta, Indian Institute of Technology Delhi, India**



## Technical Group Website:

[www.osa.org/EnvironmentalSensingTG](http://www.osa.org/EnvironmentalSensingTG)



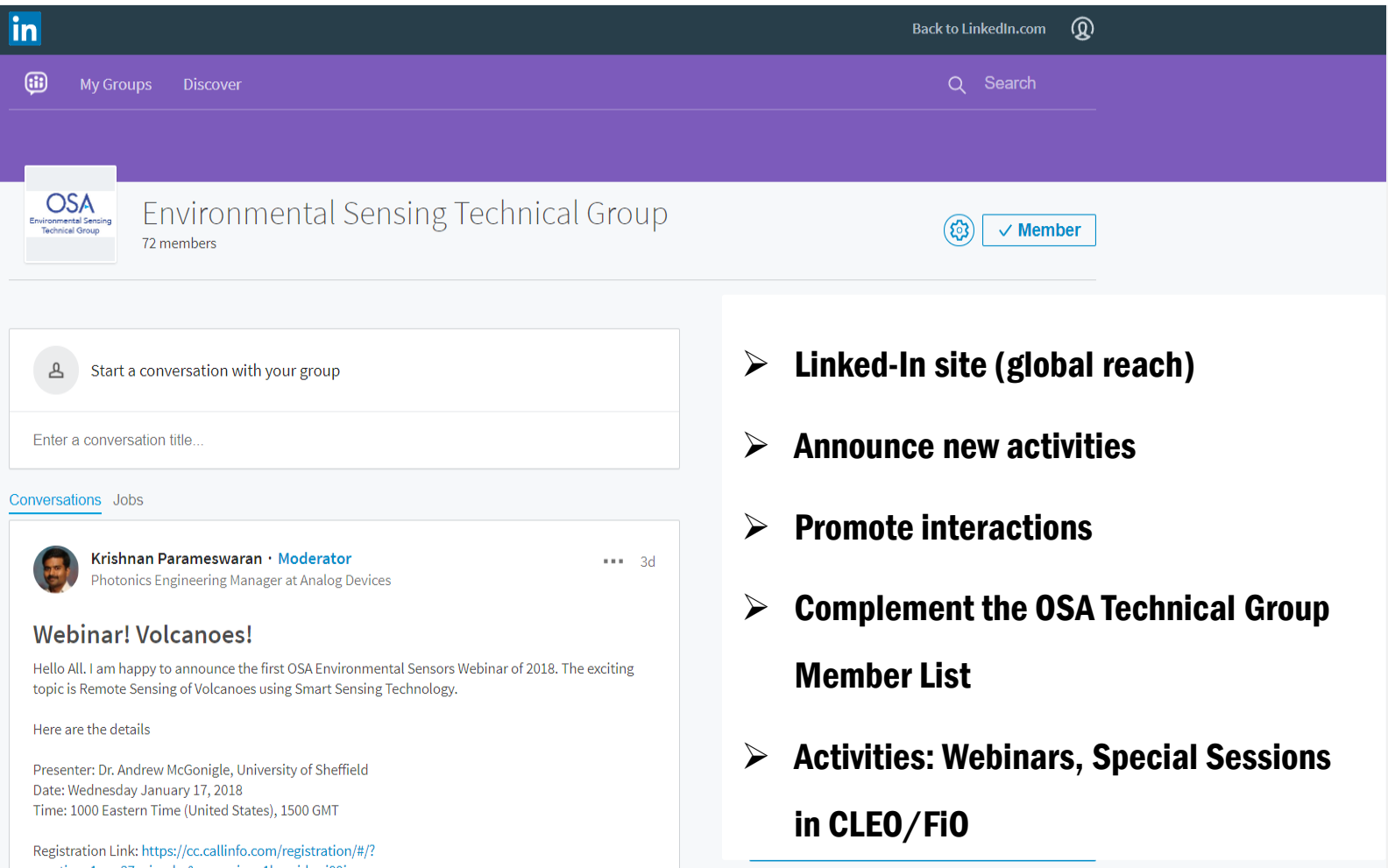
Over 1,100 Total Members

## Scope:

This technical group covers optical tools and techniques used in environmental sensing, including DIAL and LIDAR, hyperspectral monitoring, detection, processing and characterization, surveying applications, atmospheric propagation, pollution monitoring, and remote imaging. Also included in this area is remote sensing for military and commercial applications such as land management, target detection, and disaster monitoring.

# Contact your Technical Group and Get Involved!

[www.linkedin.com/groups/12055528](http://www.linkedin.com/groups/12055528)



The screenshot shows the LinkedIn profile for the Environmental Sensing Technical Group. The page header includes the LinkedIn logo, navigation links for 'My Groups' and 'Discover', and a search bar. The group's name and member count (72) are displayed, along with a 'Member' button. A conversation prompt is visible, followed by a post from Krishnan Parameswaran, Moderator, announcing a webinar on 'Volcanoes!'. The post includes details about the presenter, date, time, and registration link.

**OSA** Environmental Sensing Technical Group  
72 members

Start a conversation with your group

Enter a conversation title...

[Conversations](#) [Jobs](#)

**Krishnan Parameswaran** · Moderator  
Photonics Engineering Manager at Analog Devices

**Webinar! Volcanoes!**

Hello All. I am happy to announce the first OSA Environmental Sensors Webinar of 2018. The exciting topic is Remote Sensing of Volcanoes using Smart Sensing Technology.

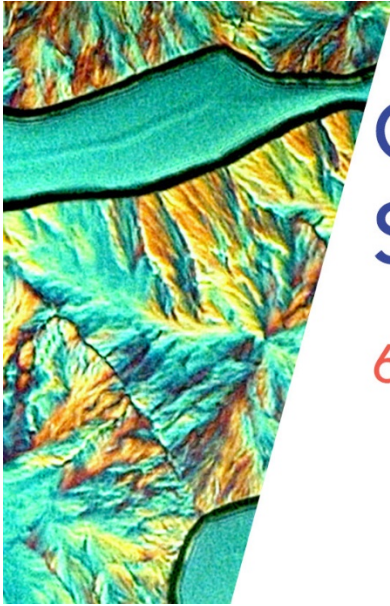
Here are the details

Presenter: Dr. Andrew McGonigle, University of Sheffield  
Date: Wednesday January 17, 2018  
Time: 1000 Eastern Time (United States), 1500 GMT

Registration Link: <https://cc.callinfo.com/registration/#/?meeting=1ee27e6c1e6e&campaign=1b7e6d6e1e6e>

- **Linked-In site (global reach)**
- **Announce new activities**
- **Promote interactions**
- **Complement the OSA Technical Group Member List**
- **Activities: Webinars, Special Sessions in CLEO/FiO**

Welcome to Today's Webinar!



# QUANTITATIVE MOLECULAR SPECTROSCOPY IN CAVITY WEBINAR

6 December 2018 • 10:00 EST

## **Dr. Patrick Dupré, Université du Littoral Côte d'Opale**

Patrick Dupré is a recognized expert in molecular high resolution and quantitative laser Spectroscopy. His career has included work in physics and chemistry laboratories in France, the United States, the United Kingdom and Germany. His interests include experimental spectroscopy and modeling. He is presently involved in developing Noise-Immune Cavity-Enhanced Optical Heterodyne Molecular Spectrometry (NICE-OHMS) for metrology applications and for trace gas detection in the Mid-InfraRed. Spectroscopy with high finesse cavity is an ideal tool for saturated absorption, i.e. under sub-Doppler conditions.

# Quantitative Molecular Spectroscopy in Cavity

Patrick DUPRÉ

Laboratoire de Physico-Chimie de l'Atmosphère, ULCO  
Dunkerque, France  
December 2018



- 1 Motivations
- 2 Introduction to CEAS
- 3 Absorption in Cavity
- 4 Saturated Absorption: Modeling and Simulations
- 5 NICE-OHMS: Principles and Implementation
- 6  $C_2H_2$  in the NIR
- 7 HD in the NIR (Forbidden Transition)
- 8 Bibliography
- 9 Conclusions

- 1 Motivations
- 2 Introduction to CEAS
- 3 Absorption in Cavity
- 4 Saturated Absorption: Modeling and Simulations
- 5 NICE-OHMS: Principles and Implementation
- 6  $C_2H_2$  in the NIR
- 7 HD in the NIR (Forbidden Transition)
- 8 Bibliography
- 9 Conclusions



- Ultrasensitivity, i.e., Trace Detection

# Motivations

- Ultrasensitivity, i.e., Trace Detection
- Quantitative Spectroscopy of Gas (aerosol?)

# Motivations

- Ultrasensitivity, i.e., Trace Detection
- Quantitative Spectroscopy of Gas (aerosol?)
- Gas Metrology (OFC)

# Motivations

- Ultrasensitivity, i.e., Trace Detection
- Quantitative Spectroscopy of Gas (aerosol?)
- Gas Metrology (OFC)
- **Line Shape Analysis**

# Motivations

- Ultrasensitivity, i.e., Trace Detection
- Quantitative Spectroscopy of Gas (aerosol?)
- Gas Metrology (OFC)
- Line Shape Analysis
- Compact Setups

# Motivations

- Ultrasensitivity, i.e., Trace Detection
- Quantitative Spectroscopy of Gas (aerosol?)
- Gas Metrology (OFC)
- Line Shape Analysis
- Compact Setups
- Beyond the linear Absorption (Doppler-Free)

# Motivations

- Ultrasensitivity, i.e., Trace Detection
- Quantitative Spectroscopy of Gas (aerosol?)
- Gas Metrology (OFC)
- Line Shape Analysis
- Compact Setups
- Beyond the linear Absorption (Doppler-Free)
- **Ab-initio Molecular Calculation Benchmarking**

- Ultrasensitivity, i.e., Trace Detection
- Quantitative Spectroscopy of Gas (aerosol?)
- Gas Metrology (OFC)
- Line Shape Analysis
- Compact Setups
- Beyond the linear Absorption (Doppler-Free)
- Ab-initio Molecular Calculation Benchmarking
- Molecule Internal Couplings (like Hyperfine Couplings)



- Ultrasensitivity, i.e., Trace Detection
- Quantitative Spectroscopy of Gas (aerosol?)
- Gas Metrology (OFC)
- Line Shape Analysis
- Compact Setups
- Beyond the linear Absorption (Doppler-Free)
- Ab-initio Molecular Calculation Benchmarking
- Molecule Internal Couplings (like Hyperfine Couplings)
- **Physics Beyond the Standard Model (i.e., QED)**

- Ultrasensitivity, i.e., Trace Detection
- Quantitative Spectroscopy of Gas (aerosol?)
- Gas Metrology (OFC)
- Line Shape Analysis
- Compact Setups
- Beyond the linear Absorption (Doppler-Free)
- Ab-initio Molecular Calculation Benchmarking
- Molecule Internal Couplings (like Hyperfine Couplings)
- Physics Beyond the Standard Model (i.e., QED)
- Challenging the Photon-Shot-Noise

- Ultrasensitivity, i.e., Trace Detection
- Quantitative Spectroscopy of Gas (aerosol?)
- Gas Metrology (OFC)
- Line Shape Analysis
- Compact Setups
- Beyond the linear Absorption (Doppler-Free)
- Ab-initio Molecular Calculation Benchmarking
- Molecule Internal Couplings (like Hyperfine Couplings)
- Physics Beyond the Standard Model (i.e., QED)
- Challenging the Photon-Shot-Noise
-

# Outline

- 1 Motivations
- 2 Introduction to CEAS**
- 3 Absorption in Cavity
- 4 Saturated Absorption: Modeling and Simulations
- 5 NICE-OHMS: Principles and Implementation
- 6  $C_2H_2$  in the NIR
- 7 HD in the NIR (Forbidden Transition)
- 8 Bibliography
- 9 Conclusions

# Cavity Enhanced Absorption Spectroscopy (CEAS)

Basic Idea: **Enhancing the Absorption Length**, i.e., the length of interaction between light and analyte. How?

# Cavity Enhanced Absorption Spectroscopy (CEAS)

Basic Idea: **Enhancing the Absorption Length**, i.e., the length of interaction between light and analyte. How?

- Multipass Cell

# Cavity Enhanced Absorption Spectroscopy (CEAS)

Basic Idea: **Enhancing the Absorption Length**, i.e., the length of interaction between light and analyte. How?

- Multipass Cell
  - White Cell

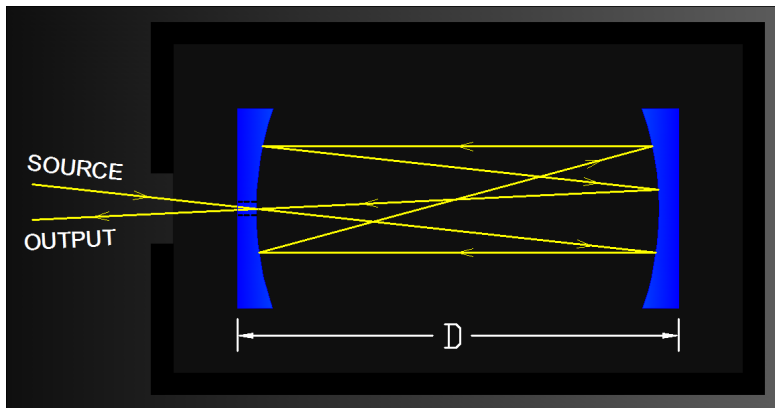
# Cavity Enhanced Absorption Spectroscopy (CEAS)

Basic Idea: **Enhancing the Absorption Length**, i.e., the length of interaction between light and analyte. How?

- Multipass Cell
  - White Cell
  - Herriot Cell



# Cavity Enhanced Absorption Spectroscopy (CEAS)



Large Metallic Mirrors

# Cavity Enhanced Absorption Spectroscopy (CEAS)

Basic Idea: **Enhancing the Absorption Length**, i.e., the length of interaction between light and analyte. How?

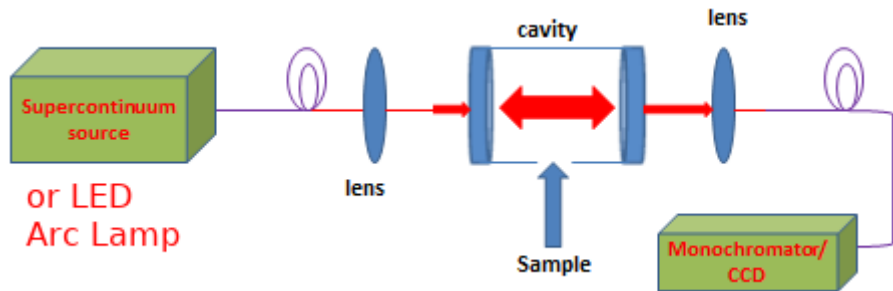
- Multipass Cell
  - White Cell
  - Herriot Cell
- **Alternative: Resonators** (using small Dichroic Mirrors, 1984)

# Cavity Enhanced Absorption Spectroscopy (CEAS)

Basic Idea: **Enhancing the Absorption Length**, i.e., the length of interaction between light and analyte. How?

- Multipass Cell
  - White Cell
  - Herriot Cell
- Alternative: **Resonators** (using small Dichroic Mirrors, 1984)
- **BBCEAS** (Broad-Band Cavity-Enhanced Absorption Spectroscopy) based on Broad Band sources (coherent or not): Arc Lamps, Supercontinuum, LED, **OFCS**. It requires a **Dispersive Detection**

# Cavity Enhanced Absorption Spectroscopy (CEAS)

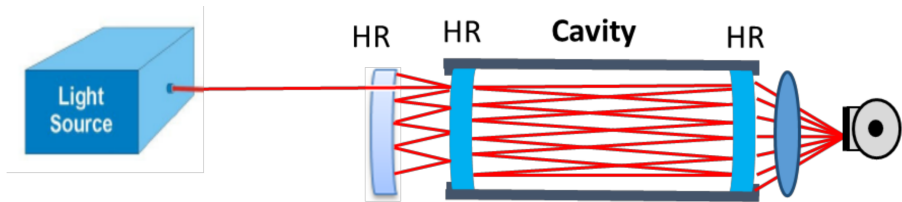


# Cavity Enhanced Absorption Spectroscopy (CEAS)

Basic Idea: **Enhancing the Absorption Length**, i.e., the length of interaction between light and analyte. How?

- Multipass Cell
  - White Cell
  - Herriot Cell
- Alternative: **Resonators** (using small Dichroic Mirrors, 1984)
- BBCEAS (Broad-Band Cavity-Enhanced Absorption Spectroscopy) based on Broad Band sources (coherent or not): Arc Lamps, Supercontinuum, LED, **OFCS**. It requires a **Dispersive Detection**
- **ICOS** (Integrated Cavity Output Spectroscopy), On-Axis, vs. Off-Axis

# Cavity Enhanced Absorption Spectroscopy (CEAS)



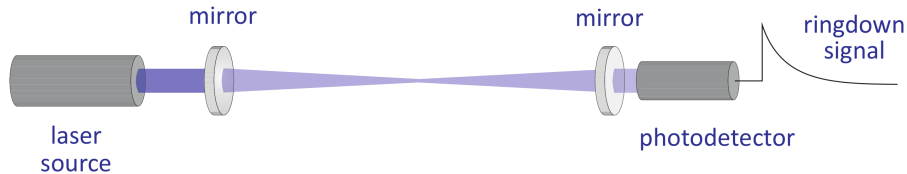
Off-Axis ICOS with recycling mirror to recover the leaking input power  
(with the permission of J. Mandon)

# Cavity Enhanced Absorption Spectroscopy (CEAS)

Basic Idea: **Enhancing the Absorption Length**, i.e., the length of interaction between light and analyte. How?

- Multipass Cell
  - White Cell
  - Herriot Cell
- Alternative: **Resonators** (using small Dichroic Mirrors, 1984)
- BBCEAS (Broad-Band Cavity-Enhanced Absorption Spectroscopy) based on Broad Band sources (coherent or not): Arc Lamps, Supercontinuum, LED, **OFCS**. It requires a **Dispersive Detection**
- ICOS (Integrated Cavity Output Spectroscopy), On-Axis, vs. Off-Axis
- CRDS (Cavity Ring-Down Spectroscopy), Continuous or Pulsed Wave, Broad-Band vs. Narrow-Band Source (see O'Keefe 1988)

# Cavity Enhanced Absorption Spectroscopy (CEAS)



With the permission of C. Vallance



# Cavity Enhanced Absorption Spectroscopy (CEAS)

Basic Idea: **Enhancing the Absorption Length**, i.e., the length of interaction between light and analyte. How?

- Multipass Cell
  - White Cell
  - Herriot Cell
- Alternative: **Resonators** (using small Dichroic Mirrors, 1984)
- BBCEAS (Broad-Band Cavity-Enhanced Absorption Spectroscopy) based on Broad Band sources (coherent or not): Arc Lamps, Supercontinuum, LED, **OFCS**. It requires a **Dispersive Detection**
- ICOS (Integrated Cavity Output Spectroscopy), On-Axis, vs. Off-Axis
- CRDS (Cavity Ring-Down Spectroscopy), Continuous or Pulsed Wave, Broad-Band vs. Narrow-Band Source (see O'Keefe 1988)
- Cavity Finesse measurement (in Frequency)

# Cavity Enhanced Absorption Spectroscopy (CEAS)

Basic Idea: **Enhancing the Absorption Length**, i.e., the length of interaction between light and analyte. How?

- Multipass Cell
  - White Cell
  - Herriot Cell
- Alternative: **Resonators** (using small Dichroic Mirrors, 1984)
- BBCEAS (Broad-Band Cavity-Enhanced Absorption Spectroscopy) based on Broad Band sources (coherent or not): Arc Lamps, Supercontinuum, LED, **OFCS**. It requires a **Dispersive Detection**
- ICOS (Integrated Cavity Output Spectroscopy), On-Axis, vs. Off-Axis
- CRDS (Cavity Ring-Down Spectroscopy), Continuous or Pulsed Wave, Broad-Band vs. Narrow-Band Source (see O'Keefe 1988)
- Cavity Finesse measurement (in Frequency)
- Cavity Impedance Mismatch (Ring)

# Cavity Enhanced Absorption Spectroscopy (CEAS)

Basic Idea: **Enhancing the Absorption Length**, i.e., the length of interaction between light and analyte. How?

- Multipass Cell
  - White Cell
  - Herriot Cell
- Alternative: **Resonators** (using small Dichroic Mirrors, 1984)
- BBCEAS (Broad-Band Cavity-Enhanced Absorption Spectroscopy) based on Broad Band sources (coherent or not): Arc Lamps, Supercontinuum, LED, **OFCS**. It requires a **Dispersive Detection**
- ICOS (Integrated Cavity Output Spectroscopy), On-Axis, vs. Off-Axis
- CRDS (Cavity Ring-Down Spectroscopy), Continuous or Pulsed Wave, Broad-Band vs. Narrow-Band Source (see O'Keefe 1988)
- Cavity Finesse measurement (in Frequency)
- Cavity Impedance Mismatch (Ring)
- **FMS** (Frequency Modulation Spectroscopy)

# Cavity Enhanced Absorption Spectroscopy (CEAS)

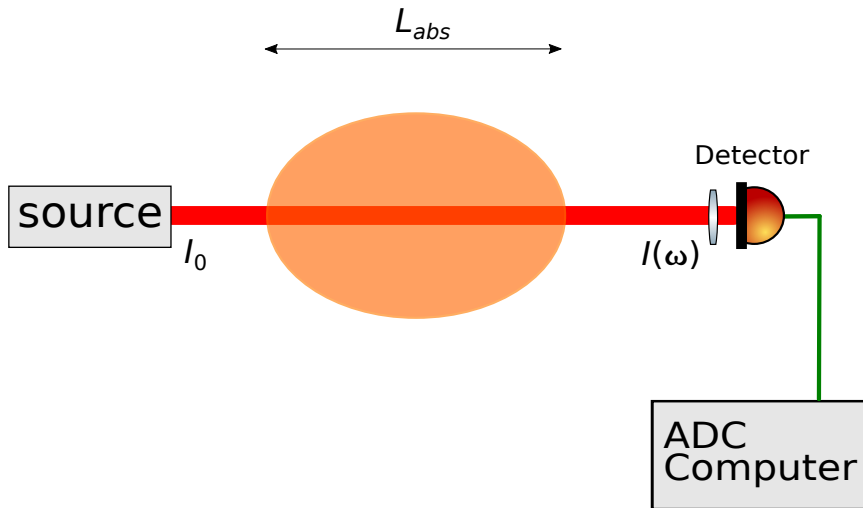
Basic Idea: **Enhancing the Absorption Length**, i.e., the length of interaction between light and analyte. How?

- Multipass Cell
  - White Cell
  - Herriot Cell
- Alternative: **Resonators** (using small Dichroic Mirrors, 1984)
- BBCEAS (Broad-Band Cavity-Enhanced Absorption Spectroscopy) based on Broad Band sources (coherent or not): Arc Lamps, Supercontinuum, LED, **OFCS**. It requires a **Dispersive Detection**
- ICOS (Integrated Cavity Output Spectroscopy), On-Axis, vs. Off-Axis
- CRDS (Cavity Ring-Down Spectroscopy), Continuous or Pulsed Wave, Broad-Band vs. Narrow-Band Source (see O'Keefe 1988)
- Cavity Finesse measurement (in Frequency)
- Cavity Impedance Mismatch (Ring)
- FMS (Frequency Modulation Spectroscopy)
- **NICE-OHMS** (Noise-Immune Cavity-Enhanced Optical Heterodyne Molecular Spectroscopy)

# Outline

- 1 Motivations
- 2 Introduction to CEAS
- 3 Absorption in Cavity**
- 4 Saturated Absorption: Modeling and Simulations
- 5 NICE-OHMS: Principles and Implementation
- 6 C<sub>2</sub>H<sub>2</sub> in the NIR
- 7 HD in the NIR (Forbidden Transition)
- 8 Bibliography
- 9 Conclusions

# Absorption



# Background

- The Absorption Beer-Lambert Law:

$$I(\omega) = I_0 e^{-\alpha(\omega) L_{abs}}$$

$\alpha(\omega) = \mathcal{N} \sigma(\omega)$  is the frequency depending absorption coef. (in  $\text{cm}^{-1}$ ).

# Background

- The Absorption Beer-Lambert Law:

$$I(\omega) = I_0 e^{-\alpha(\omega) L_{abs}}$$

$\alpha(\omega) = \mathcal{N} \sigma(\omega)$  is the frequency depending absorption coef. (in  $\text{cm}^{-1}$ ).

- Approximation of the Optically Thin Medium [ $\alpha(\omega) L_{abs} \ll 1$ ]:

$$\Delta I(\omega) = \frac{I_0 - I(\omega)}{I_0} = \alpha(\omega) L_{abs}$$



# Background

- The Absorption Beer-Lambert Law:

$$I(\omega) = I_0 e^{-\alpha(\omega) L_{abs}}$$

$\alpha(\omega) = \mathcal{N} \sigma(\omega)$  is the frequency depending absorption coef. (in  $\text{cm}^{-1}$ ).

- Approximation of the Optically Thin Medium [ $\alpha(\omega) L_{abs} \ll 1$ ]:

$$\Delta I(\omega) = \frac{I_0 - I(\omega)}{I_0} = \alpha(\omega) L_{abs}$$

- The Number Density ( $\mathcal{N}$  in  $\text{cm}^{-3}$ ) is proportional to

# Background

- The Absorption Beer-Lambert Law:

$$I(\omega) = I_0 e^{-\alpha(\omega) L_{abs}}$$

$\alpha(\omega) = \mathcal{N} \sigma(\omega)$  is the frequency depending absorption coef. (in  $\text{cm}^{-1}$ ).

- Approximation of the Optically Thin Medium [ $\alpha(\omega) L_{abs} \ll 1$ ]:

$$\Delta I(\omega) = \frac{I_0 - I(\omega)}{I_0} = \alpha(\omega) L_{abs}$$

- The Number Density ( $\mathcal{N}$  in  $\text{cm}^{-3}$ ) is proportional to
  - the Gas Pressure

# Background

- The Absorption Beer-Lambert Law:

$$I(\omega) = I_0 e^{-\alpha(\omega) L_{abs}}$$

$\alpha(\omega) = \mathcal{N} \sigma(\omega)$  is the frequency depending absorption coef. (in  $\text{cm}^{-1}$ ).

- Approximation of the Optically Thin Medium [ $\alpha(\omega) L_{abs} \ll 1$ ]:

$$\Delta I(\omega) = \frac{I_0 - I(\omega)}{I_0} = \alpha(\omega) L_{abs}$$

- The Number Density ( $\mathcal{N}$  in  $\text{cm}^{-3}$ ) is proportional to
  - the Gas Pressure
  - the Concentration of each specific species

# Background

- The Absorption Beer-Lambert Law:

$$I(\omega) = I_0 e^{-\alpha(\omega) L_{abs}}$$

$\alpha(\omega) = \mathcal{N} \sigma(\omega)$  is the frequency depending absorption coef. (in  $\text{cm}^{-1}$ ).

- Approximation of the Optically Thin Medium [ $\alpha(\omega) L_{abs} \ll 1$ ]:

$$\Delta I(\omega) = \frac{I_0 - I(\omega)}{I_0} = \alpha(\omega) L_{abs}$$

- The Number Density ( $\mathcal{N}$  in  $\text{cm}^{-3}$ ) is proportional to
  - the Gas Pressure
  - the Concentration of each specific species
- The line absorption Crosssection ( $\sigma(\omega)$  in  $\text{cm}^2/\text{molecule}$ ), includes a Normalized Lineshape (like a Voigt profile whose width is pressure depending):

$$\int \sigma(\omega) d\omega = \mathcal{S}$$

where  $\mathcal{S}$  is the Line Intensity (in  $\text{cm}/\text{molecule}$  if  $\omega$  is in  $\text{cm}^{-1}$ ).

# Background

- The Absorption Beer-Lambert Law:

$$I(\omega) = I_0 e^{-\alpha(\omega) L_{abs}}$$

$\alpha(\omega) = \mathcal{N} \sigma(\omega)$  is the frequency depending absorption coef. (in  $\text{cm}^{-1}$ ).

- Approximation of the Optically Thin Medium [ $\alpha(\omega) L_{abs} \ll 1$ ]:

$$\Delta I(\omega) = \frac{I_0 - I(\omega)}{I_0} = \alpha(\omega) L_{abs}$$

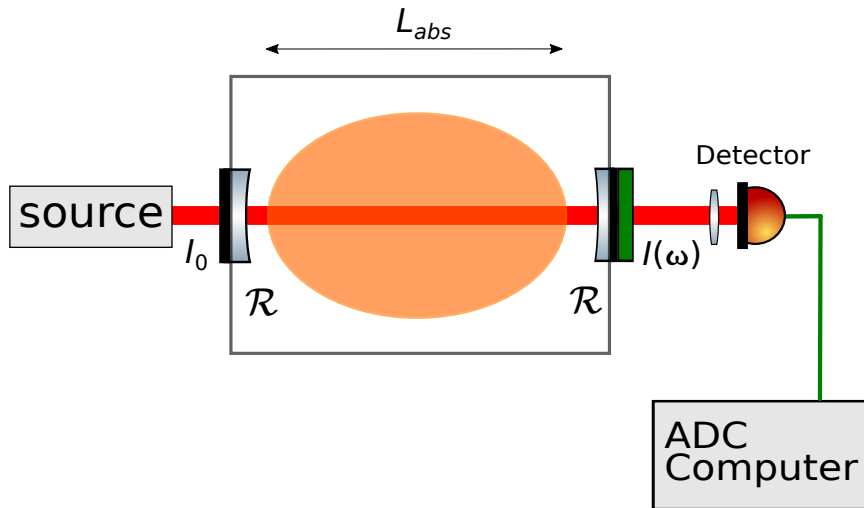
- The Number Density ( $\mathcal{N}$  in  $\text{cm}^{-3}$ ) is proportional to
  - the Gas Pressure
  - the Concentration of each specific species
- The line absorption Crosssection ( $\sigma(\omega)$  in  $\text{cm}^2/\text{molecule}$ ), includes a Normalized Lineshape (like a Voigt profile whose width is pressure depending):

$$\int \sigma(\omega) d\omega = \mathcal{S}$$

where  $\mathcal{S}$  is the Line Intensity (in  $\text{cm}/\text{molecule}$  if  $\omega$  is in  $\text{cm}^{-1}$ ).

- $\mathcal{S}$  is available in the database like HITRAN (<http://hitran.org/>)

# Absorption in a Resonant Cavity



# Main Features of a Symmetric Resonant Cavity

# Main Features of a Symmetric Resonant Cavity

- Cavity Finesse (Enhancement Factor)

$$\mathcal{F} = \frac{2\pi}{\mathcal{L}} = \frac{\pi\sqrt{\mathcal{R}}}{1-\mathcal{R}}$$



# Main Features of a Symmetric Resonant Cavity

- Cavity Finesse (Enhancement Factor)

$$\mathcal{F} = \frac{2\pi}{\mathcal{L}} = \frac{\pi\sqrt{\mathcal{R}}}{1-\mathcal{R}}$$

- Free Spectral Range (FSR)

$$FSR = \frac{c}{2L_{cav}}$$

# Main Features of a Symmetric Resonant Cavity

- Cavity Finesse (Enhancement Factor)

$$\mathcal{F} = \frac{2\pi}{\mathcal{L}} = \frac{\pi\sqrt{\mathcal{R}}}{1-\mathcal{R}}$$

- Free Spectral Range (FSR)

$$FSR = \frac{c}{2L_{cav}}$$

- Response Time (or Characteristics Time)

$$\tau_{RD} = \frac{\mathcal{F} L_{cav}}{\pi c} = \frac{\mathcal{F}}{2\pi} t_{rt} = \frac{1}{2\Delta_{cav}} = \frac{\mathcal{F}}{2\pi FSR}$$

# Main Features of a Symmetric Resonant Cavity

- Cavity Finesse (Enhancement Factor)

$$\mathcal{F} = \frac{2\pi}{\mathcal{L}} = \frac{\pi\sqrt{\mathcal{R}}}{1-\mathcal{R}}$$

- Free Spectral Range (FSR)

$$FSR = \frac{c}{2L_{cav}}$$

- Response Time (or Characteristics Time)

$$\tau_{RD} = \frac{\mathcal{F}L_{cav}}{\pi c} = \frac{\mathcal{F}}{2\pi} t_{rt} = \frac{1}{2\Delta_{cav}} = \frac{\mathcal{F}}{2\pi FSR}$$

- Equivalent Absorption Length

$$L_{eq} = \frac{2\mathcal{F}L_{cav}}{\pi} = 2c\tau_{RD}$$

# Main Features of a Symmetric Resonant Cavity

- Cavity Finesse (Enhancement Factor)

$$\mathcal{F} = \frac{2\pi}{\mathcal{L}} = \frac{\pi\sqrt{\mathcal{R}}}{1-\mathcal{R}}$$

- Free Spectral Range (FSR)

$$FSR = \frac{c}{2L_{cav}}$$

- Response Time (or Characteristics Time)

$$\tau_{RD} = \frac{\mathcal{F}L_{cav}}{\pi c} = \frac{\mathcal{F}}{2\pi} t_{rt} = \frac{1}{2\Delta_{cav}} = \frac{\mathcal{F}}{2\pi FSR}$$

- Equivalent Absorption Length

$$L_{eq} = \frac{2\mathcal{F}L_{cav}}{\pi} = 2c\tau_{RD}$$

- Trapped Power

$$\mathcal{I}_{cav} = \frac{\mathcal{F}}{\pi} \mathcal{I}_{in}$$

# Formalism: Transfer Function (“Filter”) of a Lossless Cavity

# Formalism: Transfer Function (“Filter”) of a Lossless Cavity

- In the Frequency Domain

$$\mathcal{I}_{out}(\omega) = |\mathcal{T}_{cav}(\omega) \cdot \mathcal{E}_{in}(\omega)|^2$$

with (obtained from multiple interferences)

$$\mathcal{T}_{cav}(\omega) = \frac{\mathcal{T} e^{-i\omega t_{rt}/2}}{1 + \mathcal{R} e^{i\omega t_{rt}}} = \frac{\mathcal{T}}{1 - \mathcal{R}} \sum_i \frac{1}{1 + i \left( \frac{\omega - i\omega_{FSR}}{\Delta_{cav}(\omega)} \right)}$$

and with

$$\mathcal{R}' = \mathcal{R} e^{-\alpha(\omega)L_{abs}}$$

# Formalism: Transfer Function (“Filter”) of a Lossless Cavity

- In the Frequency Domain

$$\mathcal{I}_{out}(\omega) = |\mathcal{T}_{cav}(\omega) \cdot \mathcal{E}_{in}(\omega)|^2$$

with (obtained from multiple interferences)

$$\mathcal{T}_{cav}(\omega) = \frac{\mathcal{T} e^{-i\omega t_{rt}/2}}{1 + \mathcal{R} e^{i\omega t_{rt}}} = \frac{\mathcal{T}}{1 - \mathcal{R}} \sum_i \frac{1}{1 + i \left( \frac{\omega - i\omega_{FSR}}{\Delta_{cav}(\omega)} \right)}$$

and with

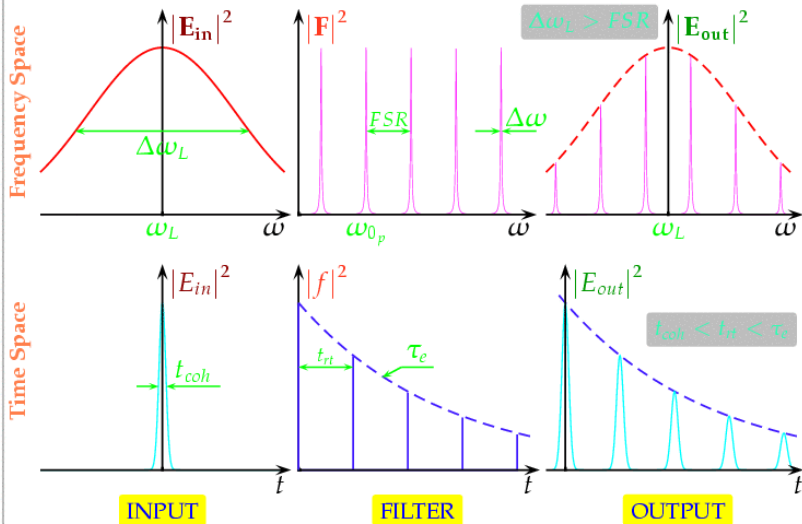
$$\mathcal{R}' = \mathcal{R} e^{-\alpha(\omega) L_{abs}}$$

- In the Time Domain

$$I_{out}(t) = \left| FT^{-1} \left[ \mathcal{T}_{cav}(\omega) \cdot \mathcal{E}_{in}(\omega) \right] \right|^2$$

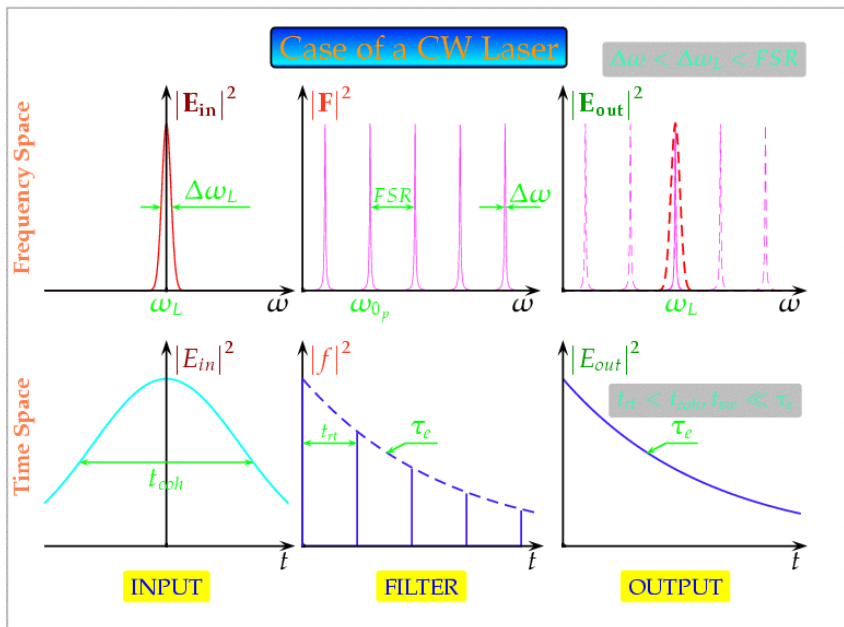
# Simulation: Pulsed Source

## Case of a Fourier Transform Limited Pulsed Laser





# Simulation: CW Source



# Absorption in a Resonant Cavity

# Absorption in a Resonant Cavity

- **Linear Absorption** at resonance (occupancy factor = 1)

# Absorption in a Resonant Cavity

- **Linear Absorption** at resonance (occupancy factor = 1)
  - Alteration of the Characteristics Time (CRDS)

$$\frac{1}{\tau_{RD}(\omega)} - \frac{1}{\tau_0} = \alpha(\omega) c$$

# Absorption in a Resonant Cavity

- **Linear Absorption** at resonance (occupancy factor = 1)
  - Alteration of the Characteristics Time (CRDS)

$$\frac{1}{\tau_{RD}(\omega)} - \frac{1}{\tau_0} = \alpha(\omega) c$$

- Alteration of the Detected Power

$$\frac{\Delta I(\omega)}{I_0} = \alpha(\omega) L_{eq}$$

# Absorption in a Resonant Cavity

- **Linear Absorption** at resonance (occupancy factor = 1)

- Alteration of the Characteristics Time (CRDS)

$$\frac{1}{\tau_{RD}(\omega)} - \frac{1}{\tau_0} = \alpha(\omega) c$$

- Alteration of the Detected Power

$$\frac{\Delta I(\omega)}{I_0} = \alpha(\omega) L_{eq}$$

- **NonLinear Absorption**

# Absorption in a Resonant Cavity

- **Linear Absorption** at resonance (occupancy factor = 1)

- Alteration of the Characteristics Time (CRDS)

$$\frac{1}{\tau_{RD}(\omega)} - \frac{1}{\tau_0} = \alpha(\omega) c$$

- Alteration of the Detected Power

$$\frac{\Delta I(\omega)}{I_0} = \alpha(\omega) L_{eq}$$

- **NonLinear Absorption**

- $\alpha(\omega, I)$

# Absorption in a Resonant Cavity

- **Linear Absorption** at resonance (occupancy factor = 1)

- Alteration of the Characteristics Time (CRDS)

$$\frac{1}{\tau_{RD}(\omega)} - \frac{1}{\tau_0} = \alpha(\omega) c$$

- Alteration of the Detected Power

$$\frac{\Delta I(\omega)}{I_0} = \alpha(\omega) L_{eq}$$

- **NonLinear Absorption**

- $\alpha(\omega, I)$
- Alteration of the Decay Shape (to nonexponential decay)



# Absorption in a Resonant Cavity

- **Linear Absorption** at resonance (occupancy factor = 1)

- Alteration of the Characteristics Time (CRDS)

$$\frac{1}{\tau_{RD}(\omega)} - \frac{1}{\tau_0} = \alpha(\omega) c$$

- Alteration of the Detected Power

$$\frac{\Delta I(\omega)}{I_0} = \alpha(\omega) L_{eq}$$

- **NonLinear Absorption**

- $\alpha(\omega, I)$
- Alteration of the Decay Shape (to nonexponential decay)
- **Lamb-dip, etc...**

# Absorption in a Resonant Cavity

- **Linear Absorption** at resonance (occupancy factor = 1)

- Alteration of the Characteristics Time (CRDS)

$$\frac{1}{\tau_{RD}(\omega)} - \frac{1}{\tau_0} = \alpha(\omega) c$$

- Alteration of the Detected Power

$$\frac{\Delta I(\omega)}{I_0} = \alpha(\omega) L_{eq}$$

- **NonLinear Absorption**

- $\alpha(\omega, I)$
- Alteration of the Decay Shape (to nonexponential decay)
- Lamb-dip, etc...



# Limit of Detection or Sensitivity

Analysis of the Signal to Noise Ratios (SNR) for the different techniques

# Limit of Detection or Sensitivity

Analysis of the Signal to Noise Ratios (SNR) for the different techniques

- Signal (Cavity Enhancement factor:  $\sim \mathcal{F}$ )

# Limit of Detection or Sensitivity

Analysis of the Signal to Noise Ratios (SNR) for the different techniques

- Signal (Cavity Enhancement factor:  $\sim \mathcal{F}$ )
- Source Intensity Fluctuations versus Photon-Shot-Noise (PSN)

$$\sqrt{\frac{2e\Delta\nu}{\eta \langle P_{eff} \rangle}} = (\alpha L_{eq})_{min}$$

# Limit of Detection or Sensitivity

Analysis of the Signal to Noise Ratios (SNR) for the different techniques

- Signal (Cavity Enhancement factor:  $\sim \mathcal{F}$ )
- Source Intensity Fluctuations versus Photon-Shot-Noise (PSN)

$$\sqrt{\frac{2e\Delta\nu}{\eta \langle P_{eff} \rangle}} = (\alpha L_{eq})_{min}$$

- CRDS is intrinsically Immune to Source Intensity Fluctuations (discontinuous acquisition)

# Limit of Detection or Sensitivity

Analysis of the Signal to Noise Ratios (SNR) for the different techniques

- Signal (Cavity Enhancement factor:  $\sim \mathcal{F}$ )
- Source Intensity Fluctuations versus Photon-Shot-Noise (PSN)

$$\sqrt{\frac{2e\Delta\nu}{\eta \langle P_{eff} \rangle}} = (\alpha L_{eq})_{min}$$

- CRDS is intrinsically Immune to Source Intensity Fluctuations (discontinuous acquisition)
- The “Direct” Absorption techniques require acquiring the Noise Immunity

# Limit of Detection or Sensitivity

Analysis of the Signal to Noise Ratios (SNR) for the different techniques

- Signal (Cavity Enhancement factor:  $\sim \mathcal{F}$ )
- Source Intensity Fluctuations versus Photon-Shot-Noise (PSN)

$$\sqrt{\frac{2e\Delta\nu}{\eta \langle P_{eff} \rangle}} = (\alpha L_{eq})_{min}$$

- CRDS is intrinsically Immune to Source Intensity Fluctuations (discontinuous acquisition)
- The “Direct” Absorption techniques require acquiring the Noise Immunity
  - Differential Absorption (DAS)



# Limit of Detection or Sensitivity

Analysis of the Signal to Noise Ratios (SNR) for the different techniques

- Signal (Cavity Enhancement factor:  $\sim \mathcal{F}$ )
- Source Intensity Fluctuations versus Photon-Shot-Noise (PSN)

$$\sqrt{\frac{2e\Delta\nu}{\eta \langle P_{eff} \rangle}} = (\alpha L_{eq})_{min}$$

- CRDS is intrinsically Immune to Source Intensity Fluctuations (discontinuous acquisition)
- The “Direct” Absorption techniques require acquiring the Noise Immunity
  - Differential Absorption (DAS)
  - **Amplitude Modulation**

# Limit of Detection or Sensitivity

Analysis of the Signal to Noise Ratios (SNR) for the different techniques

- Signal (Cavity Enhancement factor:  $\sim \mathcal{F}$ )
- Source Intensity Fluctuations versus Photon-Shot-Noise (PSN)

$$\sqrt{\frac{2e\Delta\nu}{\eta \langle P_{eff} \rangle}} = (\alpha L_{eq})_{min}$$

- CRDS is intrinsically Immune to Source Intensity Fluctuations (discontinuous acquisition)
- The “Direct” Absorption techniques require acquiring the Noise Immunity
  - Differential Absorption (DAS)
  - Amplitude Modulation
  - **Frequency/Phase Modulation (FMS)**

# Limit of Detection or Sensitivity

Analysis of the Signal to Noise Ratios (SNR) for the different techniques

- Signal (Cavity Enhancement factor:  $\sim \mathcal{F}$ )
- Source Intensity Fluctuations versus Photon-Shot-Noise (PSN)

$$\sqrt{\frac{2e\Delta\nu}{\eta \langle P_{eff} \rangle}} = (\alpha L_{eq})_{min}$$

- CRDS is intrinsically Immune to Source Intensity Fluctuations (discontinuous acquisition)
- The “Direct” Absorption techniques require acquiring the Noise Immunity
  - Differential Absorption (DAS)
  - Amplitude Modulation
  - Frequency/Phase Modulation (FMS)
  - **Beam Intensity Stabilization (AOM)**

# Limit of Detection or Sensitivity

Analysis of the Signal to Noise Ratios (SNR) for the different techniques

- Signal (Cavity Enhancement factor:  $\sim \mathcal{F}$ )
- Source Intensity Fluctuations versus Photon-Shot-Noise (PSN)

$$\sqrt{\frac{2e\Delta\nu}{\eta \langle P_{eff} \rangle}} = (\alpha L_{eq})_{min}$$

- CRDS is intrinsically Immune to Source Intensity Fluctuations (discontinuous acquisition)
- The “Direct” Absorption techniques require acquiring the Noise Immunity
  - Differential Absorption (DAS)
  - Amplitude Modulation
  - Frequency/Phase Modulation (FMS)
  - Beam Intensity Stabilization (AOM)
- NICE-OHMS benefits of both: CW acquisition, and full noise Immunity.

# NonLinear Absorption by CRDS

# NonLinear Absorption by CRDS

- **First** demonstrated in 1999 (Saturated Absorption in jet cooled  $\text{NO}_2$ ), Romanini, Dupré & Jost, in *Vib. Spectros.* 19, 93.

# NonLinear Absorption by CRDS

- **First** demonstrated in 1999 (Saturated Absorption in jet cooled  $\text{NO}_2$ ), Romanini, Dupré & Jost, in Vib. Spectros. 19, 93.
- **Then,**

# NonLinear Absorption by CRDS

- **First** demonstrated in 1999 (Saturated Absorption in jet cooled  $\text{NO}_2$ ), Romanini, Dupré & Jost, in *Vib. Spectros.* 19, 93.
- Then,
  - P. De Natale Group (Florence) in 2010 ( $\text{CO}_2$ ), *Phys. Rev. Let.* 104, 110801



# NonLinear Absorption by CRDS

- **First** demonstrated in 1999 (Saturated Absorption in jet cooled  $\text{NO}_2$ ), Romanini, Dupré & Jost, in Vib. Spectros. 19, 93.
- Then,
  - P. De Natale Group (Florence) in 2010 ( $\text{CO}_2$ ), Phys. Rev. Let. 104, 110801
  - S. Hu (Hefei) in 2017 (CO), Rev. Scient. Inst., 88, 043108

# NonLinear Absorption by CRDS

- **First** demonstrated in 1999 (Saturated Absorption in jet cooled  $\text{NO}_2$ ), Romanini, Dupré & Jost, in Vib. Spectros. 19, 93.
- Then,
  - P. De Natale Group (Florence) in 2010 ( $\text{CO}_2$ ), Phys. Rev. Let. 104, 110801
  - S. Hu (Hefei) in 2017 ( $\text{CO}$ ), Rev. Scient. Inst., 88, 043108
- **Applications:**

# NonLinear Absorption by CRDS

- **First** demonstrated in 1999 (Saturated Absorption in jet cooled  $\text{NO}_2$ ), Romanini, Dupré & Jost, in Vib. Spectros. 19, 93.
- Then,
  - P. De Natale Group (Florence) in 2010 ( $\text{CO}_2$ ), Phys. Rev. Let. 104, 110801
  - S. Hu (Hefei) in 2017 (CO), Rev. Scient. Inst., 88, 043108
- Applications:
  - High Resolution Spectroscopy

# NonLinear Absorption by CRDS

- **First** demonstrated in 1999 (Saturated Absorption in jet cooled  $\text{NO}_2$ ), Romanini, Dupré & Jost, in Vib. Spectros. 19, 93.
- Then,
  - P. De Natale Group (Florence) in 2010 ( $\text{CO}_2$ ), Phys. Rev. Let. 104, 110801
  - S. Hu (Hefei) in 2017 (CO), Rev. Scient. Inst., 88, 043108
- Applications:
  - High Resolution Spectroscopy
  - **Simultaneous determination of the number density and of the crosssection, from a single decay (CRDS)!**

# NonLinear Absorption by CRDS

- **First** demonstrated in 1999 (Saturated Absorption in jet cooled  $\text{NO}_2$ ), Romanini, Dupré & Jost, in *Vib. Spectros.* 19, 93.
- Then,
  - P. De Natale Group (Florence) in 2010 ( $\text{CO}_2$ ), *Phys. Rev. Let.* 104, 110801
  - S. Hu (Hefei) in 2017 ( $\text{CO}$ ), *Rev. Scient. Inst.*, 88, 043108
- Applications:
  - High Resolution Spectroscopy
  - Simultaneous determination of the number density and of the crosssection, from a single decay (CRDS)!
- **Attention**

# NonLinear Absorption by CRDS

- **First** demonstrated in 1999 (Saturated Absorption in jet cooled  $\text{NO}_2$ ), Romanini, Dupré & Jost, in Vib. Spectros. 19, 93.
- Then,
  - P. De Natale Group (Florence) in 2010 ( $\text{CO}_2$ ), Phys. Rev. Let. 104, 110801
  - S. Hu (Hefei) in 2017 (CO), Rev. Scient. Inst., 88, 043108
- Applications:
  - High Resolution Spectroscopy
  - Simultaneous determination of the number density and of the crosssection, from a single decay (CRDS)!
- Attention
  - **Requiring full control of the Intracavity Power**

# NonLinear Absorption by CRDS

- **First** demonstrated in 1999 (Saturated Absorption in jet cooled  $\text{NO}_2$ ), Romanini, Dupré & Jost, in Vib. Spectros. 19, 93.
- Then,
  - P. De Natale Group (Florence) in 2010 ( $\text{CO}_2$ ), Phys. Rev. Let. 104, 110801
  - S. Hu (Hefei) in 2017 (CO), Rev. Scient. Inst., 88, 043108
- Applications:
  - High Resolution Spectroscopy
  - Simultaneous determination of the number density and of the crosssection, from a single decay (CRDS)!
- Attention
  - Requiring full control of the Intracavity Power
  - **Modeling of the Nonlinear Interaction**

# NonLinear Absorption by CRDS

- **First** demonstrated in 1999 (Saturated Absorption in jet cooled  $\text{NO}_2$ ), Romanini, Dupré & Jost, in Vib. Spectros. 19, 93.
- Then,
  - P. De Natale Group (Florence) in 2010 ( $\text{CO}_2$ ), Phys. Rev. Let. 104, 110801
  - S. Hu (Hefei) in 2017 (CO), Rev. Scient. Inst., 88, 043108
- Applications:
  - High Resolution Spectroscopy
  - Simultaneous determination of the number density and of the crosssection, from a single decay (CRDS)!
- Attention
  - Requiring full control of the Intracavity Power
  - Modeling of the Nonlinear Interaction
  - **Data Weighting (according to the noise source, see CRDS)**

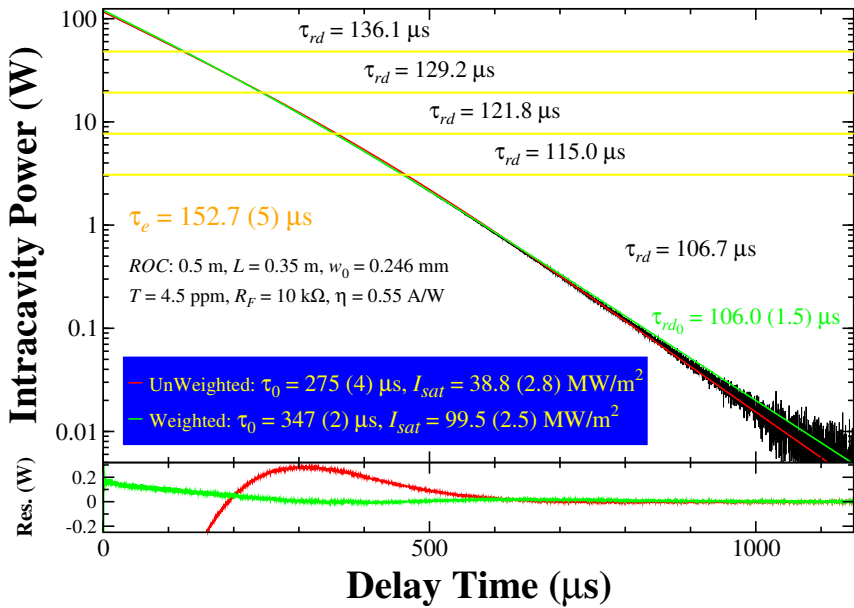


# NonLinear Absorption by CRDS

- **First** demonstrated in 1999 (Saturated Absorption in jet cooled  $\text{NO}_2$ ), Romanini, Dupré & Jost, in Vib. Spectros. 19, 93.
- Then,
  - P. De Natale Group (Florence) in 2010 ( $\text{CO}_2$ ), Phys. Rev. Let. 104, 110801
  - S. Hu (Hefei) in 2017 (CO), Rev. Scient. Inst., 88, 043108
- Applications:
  - High Resolution Spectroscopy
  - Simultaneous determination of the number density and of the crosssection, from a single decay (CRDS)!
- Attention
  - Requiring full control of the Intracavity Power
  - Modeling of the Nonlinear Interaction
  - Data Weighting (according to the noise source, see CRDS)
  - Crossover Resonances

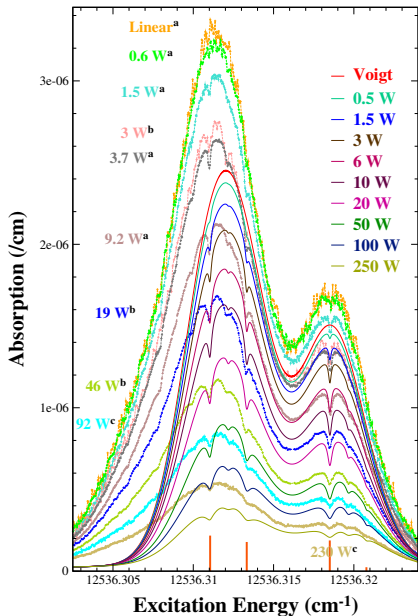
# NonExponential Decay

CRDS of Jet-Cooled  $\text{NO}_2$ : Decay of the line at  $12536.4464 \text{ cm}^{-1}$  ( ${}^qR_0(0), 3/2$ )



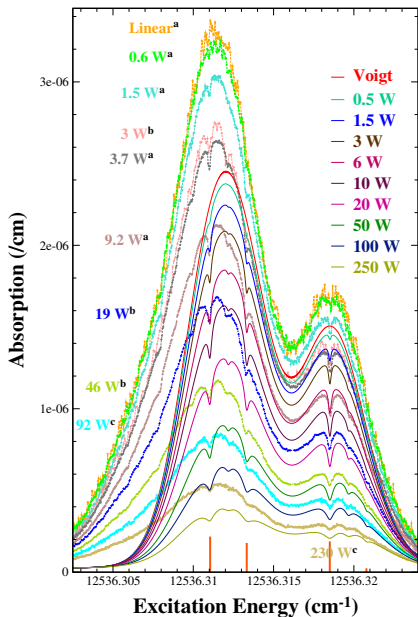
# Saturation in $\text{NO}_2$ (with Fine and Hyperfine Transitions)

Power dependence of the  ${}^0Q_{21}(0.5)$  Line Pattern

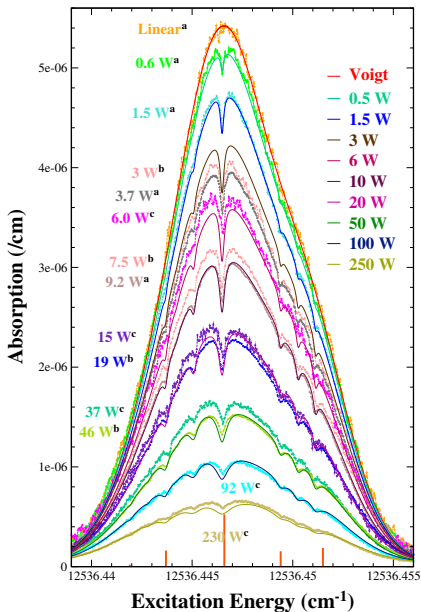


# Saturation in $\text{NO}_2$ (with Fine and Hyperfine Transitions)

Power dependence of the  ${}^0_0Q_{21}(0.5)$  Line Pattern

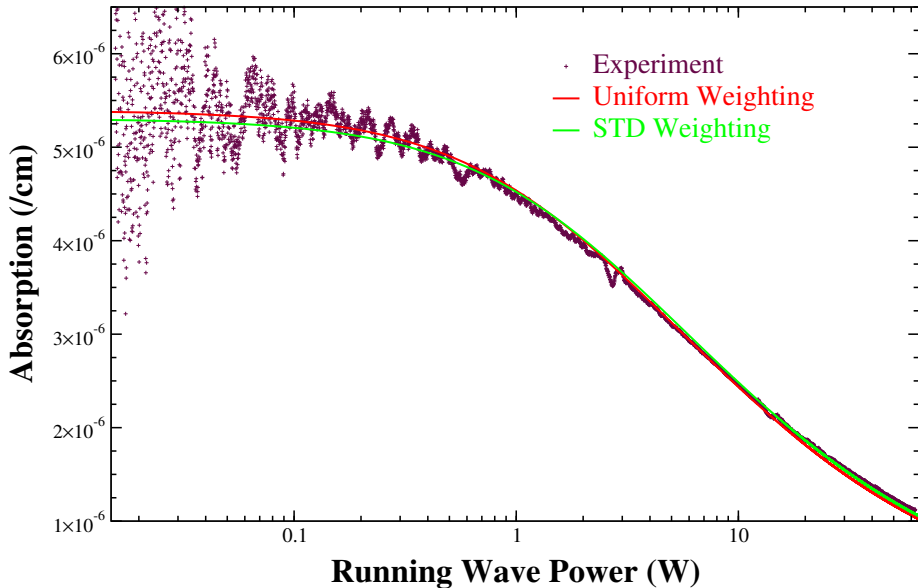


Power dependence of the  ${}^0_0R_{11}(0.5)$  Line Pattern



# Absorption versus Intracavity Power

*Absorption versus the Intracavity Power at the Center of the  ${}^9R_{11}(0.5)$  Line Pattern*



## **NICE-OHMS: History in a nutshell**

## NICE-OHMS: History in a nutshell

- **Pioneeringly developed at NIST** (J. Hall, J. Ye), first publication in 1996 on acetylene at  $1.064 \mu\text{m}$  (Nd:YAG/Ti:Sa), cavity finesse:  $\sim 100000$ ,  
**NEA  $\sim 1 \times 10^{-14} \text{ cm}^{-1} / \sqrt{\text{Hz}}$**

## NICE-OHMS: History in a nutshell

- **Pioneeringly developed at NIST** (J. Hall, J. Ye), first publication in 1996 on acetylene at  $1.064\ \mu\text{m}$  (Nd:YAG/Ti:Sa), cavity finesse:  $\sim 100000$ , **NEA  $\sim 1 \times 10^{-14}\ \text{cm}^{-1}/\sqrt{\text{Hz}}$**
- Since 2007: Ove Axner group (Umeå, SW), all fibered NIR (EDFL and DFB), MIR (OPO), more than 18 Publications ( $\mathcal{F} \sim 50000$ ), **NEA  $\sim 4 \times 10^{-13}\ \text{cm}^{-1}/\sqrt{\text{Hz}}$ , Technical Developments; 2017: Whispering-Gallery-Mode Laser**



## NICE-OHMS: History in a nutshell

- **Pioneeringly developed at NIST** (J. Hall, J. Ye), first publication in 1996 on acetylene at  $1.064\ \mu\text{m}$  (Nd:YAG/Ti:Sa), cavity finesse:  $\sim 100000$ ,  $\text{NEA} \sim 1 \times 10^{-14}\ \text{cm}^{-1}/\sqrt{\text{Hz}}$
- Since 2007: Ove Axner group (Umeå, SW), all fibered NIR (EDFL and DFB), MIR (OPO), more than 18 Publications ( $\mathcal{F} \sim 50000$ ),  $\text{NEA} \sim 4 \times 10^{-13}\ \text{cm}^{-1}/\sqrt{\text{Hz}}$ , **Technical Developments; 2017: Whispering-Gallery-Mode Laser**
- Since 2010: Ben McCall (UIUC, IL), **Molecular Ion** (Spectroscopy), Ti:Sa (Red), DFG and OPO (MIR), Jet Expansion, cavities of modest Finesse

## NICE-OHMS: History in a nutshell

- **Pioneeringly developed at NIST** (J. Hall, J. Ye), first publication in 1996 on acetylene at  $1.064\ \mu\text{m}$  (Nd:YAG/Ti:Sa), cavity finesse:  $\sim 100000$ ,  $\text{NEA} \sim 1 \times 10^{-14}\ \text{cm}^{-1}/\sqrt{\text{Hz}}$
- Since 2007: Ove Axner group (Umeå, SW), all fibered NIR (EDFL and DFB), MIR (OPO), more than 18 Publications ( $\mathcal{F} \sim 50000$ ),  $\text{NEA} \sim 4 \times 10^{-13}\ \text{cm}^{-1}/\sqrt{\text{Hz}}$ , **Technical Developments; 2017: Whispering-Gallery-Mode Laser**
- Since 2010: Ben McCall (UIUC, IL), **Molecular Ion** (Spectroscopy), Ti:Sa (Red), DFG and OPO (MIR), Jet Expansion, cavities of modest Finesse
- Since 2013, Frans Harren (Radboud Univ., Nijmegen, NL), NIR, **Trace Detection**

## NICE-OHMS: History in a nutshell

- **Pioneeringly developed at NIST** (J. Hall, J. Ye), first publication in 1996 on acetylene at  $1.064\ \mu\text{m}$  (Nd:YAG/Ti:Sa), cavity finesse:  $\sim 100000$ ,  $\text{NEA} \sim 1 \times 10^{-14}\ \text{cm}^{-1}/\sqrt{\text{Hz}}$
- Since 2007: Ove Axner group (Umeå, SW), all fibered NIR (EDFL and DFB), MIR (OPO), more than 18 Publications ( $\mathcal{F} \sim 50000$ ),  $\text{NEA} \sim 4 \times 10^{-13}\ \text{cm}^{-1}/\sqrt{\text{Hz}}$ , **Technical Developments; 2017: Whispering-Gallery-Mode Laser**
- Since 2010: Ben McCall (UIUC, IL), **Molecular Ion** (Spectroscopy), Ti:Sa (Red), DFG and OPO (MIR), Jet Expansion, cavities of modest Finesse
- Since 2013, Frans Harren (Radboud Univ., Nijmegen, NL), NIR, **Trace Detection**
- Since 2014, Livio Gianfrani (Naples Univ.): ECDL at  $1.39\ \mu\text{m}$  ( $\text{H}_2^{18}\text{O}$ ), **Metrology: Boltzmann Constant, Symmetrization postulate (detec. of forbidden transition), HD?**

# NICE-OHMS: History in a nutshell

- **Pioneeringly developed at NIST** (J. Hall, J. Ye), first publication in 1996 on acetylene at  $1.064\ \mu\text{m}$  (Nd:YAG/Ti:Sa), cavity finesse:  $\sim 100000$ ,  $\text{NEA} \sim 1 \times 10^{-14}\ \text{cm}^{-1}/\sqrt{\text{Hz}}$
- Since 2007: Ove Axner group (Umeå, SW), all fibered NIR (EDFL and DFB), MIR (OPO), more than 18 Publications ( $\mathcal{F} \sim 50000$ ),  $\text{NEA} \sim 4 \times 10^{-13}\ \text{cm}^{-1}/\sqrt{\text{Hz}}$ , **Technical Developments; 2017: Whispering-Gallery-Mode Laser**
- Since 2010: Ben McCall (UIUC, IL), **Molecular Ion** (Spectroscopy), Ti:Sa (Red), DFG and OPO (MIR), Jet Expansion, cavities of modest Finesse
- Since 2013, Frans Harren (Radboud Univ., Nijmegen, NL), NIR, **Trace Detection**
- Since 2014, Livio Gianfrani (Naples Univ.): ECDL at  $1.39\ \mu\text{m}$  ( $\text{H}_2^{18}\text{O}$ ), **Metrology**: Boltzmann Constant, Symmetrization postulate (detec. of forbidden transition), HD?
- Since 2015: Dual NICE-OHMS (CO, NIR), Shally Saraf, Robert Byer (Stanford University, CA), **Metrology** (Testing Lorentz Invariance, STAR Project)?

# NICE-OHMS: History in a nutshell

- **Pioneeringly developed at NIST** (J. Hall, J. Ye), first publication in 1996 on acetylene at  $1.064\ \mu\text{m}$  (Nd:YAG/Ti:Sa), cavity finesse:  $\sim 100000$ ,  $\text{NEA} \sim 1 \times 10^{-14}\ \text{cm}^{-1}/\sqrt{\text{Hz}}$
- Since 2007: Ove Axner group (Umeå, SW), all fibered NIR (EDFL and DFB), MIR (OPO), more than 18 Publications ( $\mathcal{F} \sim 50000$ ),  $\text{NEA} \sim 4 \times 10^{-13}\ \text{cm}^{-1}/\sqrt{\text{Hz}}$ , **Technical Developments; 2017: Whispering-Gallery-Mode Laser**
- Since 2010: Ben McCall (UIUC, IL), **Molecular Ion** (Spectroscopy), Ti:Sa (Red), DFG and OPO (MIR), Jet Expansion, cavities of modest Finesse
- Since 2013, Frans Harren (Radboud Univ., Nijmegen, NL), NIR, **Trace Detection**
- Since 2014, Livio Gianfrani (Naples Univ.): ECDL at  $1.39\ \mu\text{m}$  ( $\text{H}_2^{18}\text{O}$ ), **Metrology**: Boltzmann Constant, Symmetrization postulate (detec. of forbidden transition), HD?
- Since 2015: Dual NICE-OHMS (CO, NIR), Shally Saraf, Robert Byer (Stanford University, CA), **Metrology** (Testing Lorentz Invariance, STAR Project)?
- **Since 2015: National Tsing Hua University (Taiwan), Quantum-Dot ECDL at  $1.283\ \mu\text{m}$  ( $\text{N}_2\text{O}$  in Doppler), + CRDS, Atomic Parity NonConservation**

# NICE-OHMS: History in a nutshell

- **Pioneeringly developed at NIST** (J. Hall, J. Ye), first publication in 1996 on acetylene at  $1.064\ \mu\text{m}$  (Nd:YAG/Ti:Sa), cavity finesse:  $\sim 100000$ ,  $\text{NEA} \sim 1 \times 10^{-14}\ \text{cm}^{-1}/\sqrt{\text{Hz}}$
- Since 2007: Ove Axner group (Umeå, SW), all fibered NIR (EDFL and DFB), MIR (OPO), more than 18 Publications ( $\mathcal{F} \sim 50000$ ),  $\text{NEA} \sim 4 \times 10^{-13}\ \text{cm}^{-1}/\sqrt{\text{Hz}}$ , **Technical Developments; 2017: Whispering-Gallery-Mode Laser**
- Since 2010: Ben McCall (UIUC, IL), **Molecular Ion** (Spectroscopy), Ti:Sa (Red), DFG and OPO (MIR), Jet Expansion, cavities of modest Finesse
- Since 2013, Frans Harren (Radboud Univ., Nijmegen, NL), NIR, **Trace Detection**
- Since 2014, Livio Gianfrani (Naples Univ.): ECDL at  $1.39\ \mu\text{m}$  ( $\text{H}_2^{18}\text{O}$ ), **Metrology**: Boltzmann Constant, Symmetrization postulate (detec. of forbidden transition), HD?
- Since 2015: Dual NICE-OHMS (CO, NIR), Shally Saraf, Robert Byer (Stanford University, CA), **Metrology** (Testing Lorentz Invariance, STAR Project)?
- Since 2015: National Tsing Hua University (Taiwan), Quantum-Dot ECDL at  $1.283\ \mu\text{m}$  ( $\text{N}_2\text{O}$  in Doppler), + CRDS, **Atomic Parity NonConservation**
- Since 2016: Collaboration Dunkerque/Amsterdam (VU), **Metrology of Hydrogen**

# NICE-OHMS: History in a nutshell

- **Pioneeringly developed at NIST** (J. Hall, J. Ye), first publication in 1996 on acetylene at  $1.064\ \mu\text{m}$  (Nd:YAG/Ti:Sa), cavity finesse:  $\sim 100000$ ,  $\text{NEA} \sim 1 \times 10^{-14}\ \text{cm}^{-1}/\sqrt{\text{Hz}}$
- Since 2007: Ove Axner group (Umeå, SW), all fibered NIR (EDFL and DFB), MIR (OPO), more than 18 Publications ( $\mathcal{F} \sim 50000$ ),  $\text{NEA} \sim 4 \times 10^{-13}\ \text{cm}^{-1}/\sqrt{\text{Hz}}$ , **Technical Developments; 2017: Whispering-Gallery-Mode Laser**
- Since 2010: Ben McCall (UIUC, IL), **Molecular Ion** (Spectroscopy), Ti:Sa (Red), DFG and OPO (MIR), Jet Expansion, cavities of modest Finesse
- Since 2013, Frans Harren (Radboud Univ., Nijmegen, NL), NIR, **Trace Detection**
- Since 2014, Livio Gianfrani (Naples Univ.): ECDL at  $1.39\ \mu\text{m}$  ( $\text{H}_2^{18}\text{O}$ ), **Metrology**: Boltzmann Constant, Symmetrization postulate (detec. of forbidden transition), HD?
- Since 2015: Dual NICE-OHMS (CO, NIR), Shally Saraf, Robert Byer (Stanford University, CA), **Metrology** (Testing Lorentz Invariance, STAR Project)?
- Since 2015: National Tsing Hua University (Taiwan), Quantum-Dot ECDL at  $1.283\ \mu\text{m}$  ( $\text{N}_2\text{O}$  in Doppler), + CRDS, **Atomic Parity NonConservation**
- Since 2016: Collaboration Dunkerque/Amsterdam (VU), **Metrology** of Hydrogen
- Since 2017: Stefan Schäffer, Niels Bohr Institute (Copenhagen), **MOT** of  $^{88}\text{Sr}$  (locking against transition)

# Outline

- 1 Motivations
- 2 Introduction to CEAS
- 3 Absorption in Cavity
- 4 Saturated Absorption: Modeling and Simulations**
- 5 NICE-OHMS: Principles and Implementation
- 6  $C_2H_2$  in the NIR
- 7 HD in the NIR (Forbidden Transition)
- 8 Bibliography
- 9 Conclusions



## Saturation Modeling: an Insight

- Establishing the complex absorption in the Frequency domain (coupling of the EMF with the susceptibility) for a given Doppler Shift

## Saturation Modeling: an Insight

- Establishing the complex absorption in the Frequency domain (coupling of the EMF with the susceptibility) for a given Doppler Shift
- Solving the Liouville Equation for a (2)-level system to obtain the population and coherence terms of a specific sub-transition

## Saturation Modeling: an Insight

- Establishing the complex absorption in the Frequency domain (coupling of the EMF with the susceptibility) for a given Doppler Shift
- Solving the Liouville Equation for a (2)-level system to obtain the population and coherence terms of a specific sub-transition
- Using a perturbative approach to solve the coupled system of equations

## Saturation Modeling: an Insight

- Establishing the complex absorption in the Frequency domain (coupling of the EMF with the susceptibility) for a given Doppler Shift
- Solving the Liouville Equation for a (2)-level system to obtain the population and coherence terms of a specific sub-transition
- Using a perturbative approach to solve the coupled system of equations
- **Applying the SVEA**

## Saturation Modeling: an Insight

- Establishing the complex absorption in the Frequency domain (coupling of the EMF with the susceptibility) for a given Doppler Shift
- Solving the Liouville Equation for a (2)-level system to obtain the population and coherence terms of a specific sub-transition
- Using a perturbative approach to solve the coupled system of equations
- Applying the SVEA
- Establishing the Linear Absorption

## Saturation Modeling: an Insight

- Establishing the complex absorption in the Frequency domain (coupling of the EMF with the susceptibility) for a given Doppler Shift
- Solving the Liouville Equation for a (2)-level system to obtain the population and coherence terms of a specific sub-transition
- Using a perturbative approach to solve the coupled system of equations
- Applying the SVEA
- Establishing the Linear Absorption
- Establishing the Saturated Absorption (based on the Rabi Frequency)

## Saturation Modeling: an Insight

- Establishing the complex absorption in the Frequency domain (coupling of the EMF with the susceptibility) for a given Doppler Shift
- Solving the Liouville Equation for a (2)-level system to obtain the population and coherence terms of a specific sub-transition
- Using a perturbative approach to solve the coupled system of equations
- Applying the SVEA
- Establishing the Linear Absorption
- Establishing the Saturated Absorption (based on the Rabi Frequency)
- **Plugging specific EMFs (FMS) and considering the Stationary Response**

## Saturation Modeling: an Insight

- Establishing the complex absorption in the Frequency domain (coupling of the EMF with the susceptibility) for a given Doppler Shift
- Solving the Liouville Equation for a (2)-level system to obtain the population and coherence terms of a specific sub-transition
- Using a perturbative approach to solve the coupled system of equations
- Applying the SVEA
- Establishing the Linear Absorption
- Establishing the Saturated Absorption (based on the Rabi Frequency)
- Plugging specific EMFs (FMS) and considering the Stationary Response
  - Monochromatic (Radial Extension)



## Saturation Modeling: an Insight

- Establishing the complex absorption in the Frequency domain (coupling of the EMF with the susceptibility) for a given Doppler Shift
- Solving the Liouville Equation for a (2)-level system to obtain the population and coherence terms of a specific sub-transition
- Using a perturbative approach to solve the coupled system of equations
- Applying the SVEA
- Establishing the Linear Absorption
- Establishing the Saturated Absorption (based on the Rabi Frequency)
- Plugging specific EMFs (FMS) and considering the Stationary Response
  - Monochromatic (Radial Extension)
  - Gaussian (Impact Parameter)

# Saturation Modeling: an Insight

- Establishing the complex absorption in the Frequency domain (coupling of the EMF with the susceptibility) for a given Doppler Shift
- Solving the Liouville Equation for a (2)-level system to obtain the population and coherence terms of a specific sub-transition
- Using a perturbative approach to solve the coupled system of equations
- Applying the SVEA
- Establishing the Linear Absorption
- Establishing the Saturated Absorption (based on the Rabi Frequency)
- Plugging specific EMFs (FMS) and considering the Stationary Response
  - Monochromatic (Radial Extension)
  - Gaussian (Impact Parameter)
    - Transit-Time Broadening (vs. Power Broadening)

# Saturation Modeling: an Insight

- Establishing the complex absorption in the Frequency domain (coupling of the EMF with the susceptibility) for a given Doppler Shift
- Solving the Liouville Equation for a (2)-level system to obtain the population and coherence terms of a specific sub-transition
- Using a perturbative approach to solve the coupled system of equations
- Applying the SVEA
- Establishing the Linear Absorption
- Establishing the Saturated Absorption (based on the Rabi Frequency)
- Plugging specific EMFs (FMS) and considering the Stationary Response
  - Monochromatic (Radial Extension)
  - Gaussian (Impact Parameter)
    - Transit-Time Broadening (vs. Power Broadening)
    - Approximation based on the spectral extension of products involved ( $\otimes$ ,  $\times$ )

## Saturation Modeling: an Insight

- Establishing the complex absorption in the Frequency domain (coupling of the EMF with the susceptibility) for a given Doppler Shift
- Solving the Liouville Equation for a (2)-level system to obtain the population and coherence terms of a specific sub-transition
- Using a perturbative approach to solve the coupled system of equations
- Applying the SVEA
- Establishing the Linear Absorption
- Establishing the Saturated Absorption (based on the Rabi Frequency)
- Plugging specific EMFs (FMS) and considering the Stationary Response
  - Monochromatic (Radial Extension)
  - Gaussian (Impact Parameter)
    - Transit-Time Broadening (vs. Power Broadening)
    - Approximation based on the spectral extension of products involved ( $\otimes$ ,  $\times$ )
- Numerical Integration over the Doppler Shift

# Saturation Modeling: an Insight

- Establishing the complex absorption in the Frequency domain (coupling of the EMF with the susceptibility) for a given Doppler Shift
- Solving the Liouville Equation for a (2)-level system to obtain the population and coherence terms of a specific sub-transition
- Using a perturbative approach to solve the coupled system of equations
- Applying the SVEA
- Establishing the Linear Absorption
- Establishing the Saturated Absorption (based on the Rabi Frequency)
- Plugging specific EMFs (FMS) and considering the Stationary Response
  - Monochromatic (Radial Extension)
  - Gaussian (Impact Parameter)
    - Transit-Time Broadening (vs. Power Broadening)
    - Approximation based on the spectral extension of products involved ( $\otimes$ ,  $\times$ )
- Numerical Integration over the Doppler Shift
- **Integration over the Impact Parameter**

## Saturation Modeling: an Insight

- Establishing the complex absorption in the Frequency domain (coupling of the EMF with the susceptibility) for a given Doppler Shift
- Solving the Liouville Equation for a (2)-level system to obtain the population and coherence terms of a specific sub-transition
- Using a perturbative approach to solve the coupled system of equations
- Applying the SVEA
- Establishing the Linear Absorption
- Establishing the Saturated Absorption (based on the Rabi Frequency)
- Plugging specific EMFs (FMS) and considering the Stationary Response
  - Monochromatic (Radial Extension)
  - Gaussian (Impact Parameter)
    - Transit-Time Broadening (vs. Power Broadening)
    - Approximation based on the spectral extension of products involved ( $\otimes$ ,  $\times$ )
- Numerical Integration over the Doppler Shift
- Integration over the Impact Parameter
- Integration over the Transit-Time Rate (Maxwell Boltzmann)

# Saturation Modeling: an Insight

- Establishing the complex absorption in the Frequency domain (coupling of the EMF with the susceptibility) for a given Doppler Shift
- Solving the Liouville Equation for a (2)-level system to obtain the population and coherence terms of a specific sub-transition
- Using a perturbative approach to solve the coupled system of equations
- Applying the SVEA
- Establishing the Linear Absorption
- Establishing the Saturated Absorption (based on the Rabi Frequency)
- Plugging specific EMFs (FMS) and considering the Stationary Response
  - Monochromatic (Radial Extension)
  - Gaussian (Impact Parameter)
    - Transit-Time Broadening (vs. Power Broadening)
    - Approximation based on the spectral extension of products involved ( $\otimes$ ,  $\times$ )
- Numerical Integration over the Doppler Shift
- Integration over the Impact Parameter
- Integration over the Transit-Time Rate (Maxwell Boltzmann)
- **Summation of the Degenerated Zeeman Sub-Transitions (Polarization)**

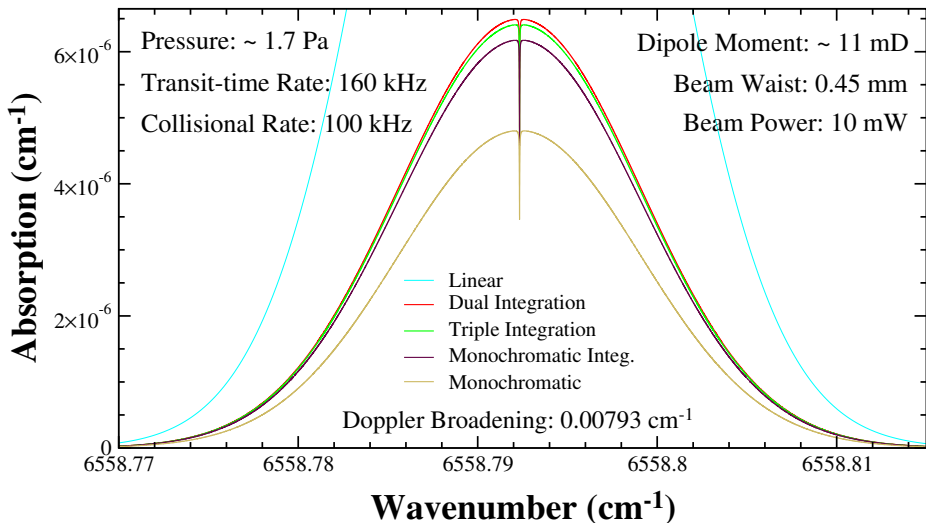
# Saturation Modeling: an Insight

- Establishing the complex absorption in the Frequency domain (coupling of the EMF with the susceptibility) for a given Doppler Shift
- Solving the Liouville Equation for a (2)-level system to obtain the population and coherence terms of a specific sub-transition
- Using a perturbative approach to solve the coupled system of equations
- Applying the SVEA
- Establishing the Linear Absorption
- Establishing the Saturated Absorption (based on the Rabi Frequency)
- Plugging specific EMFs (FMS) and considering the Stationary Response
  - Monochromatic (Radial Extension)
  - Gaussian (Impact Parameter)
    - Transit-Time Broadening (vs. Power Broadening)
    - Approximation based on the spectral extension of products involved ( $\otimes$ ,  $\times$ )
- Numerical Integration over the Doppler Shift
- Integration over the Impact Parameter
- Integration over the Transit-Time Rate (Maxwell Boltzmann)
- Summation of the Degenerated Zeeman Sub-Transitions (Polarization)
- **No Saturation Coefficient is used**



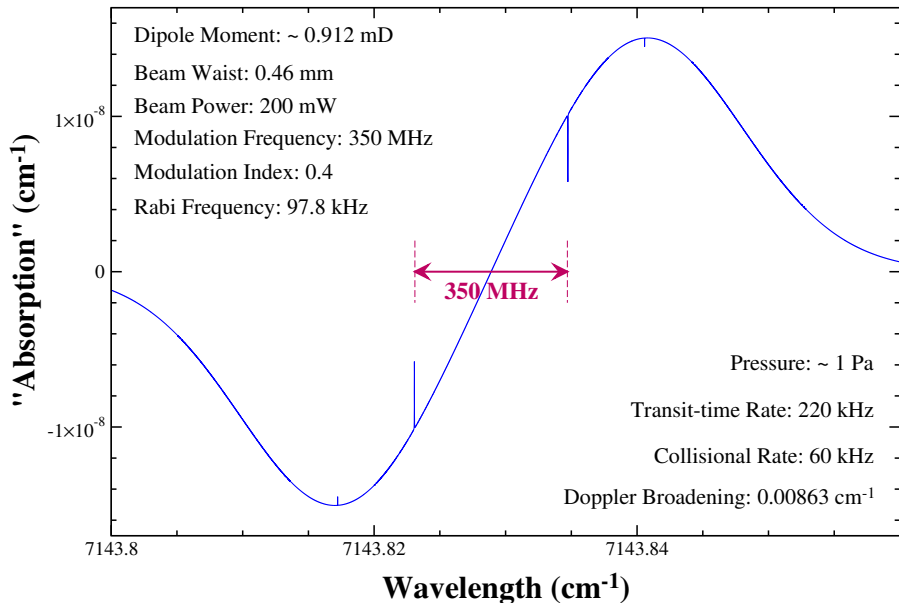
# Simulation and Line Profile Analysis

## Saturation Analysis: $C_2H_2$ Transition R(0) at $6558.79233\text{ cm}^{-1}$ (polyad 10)



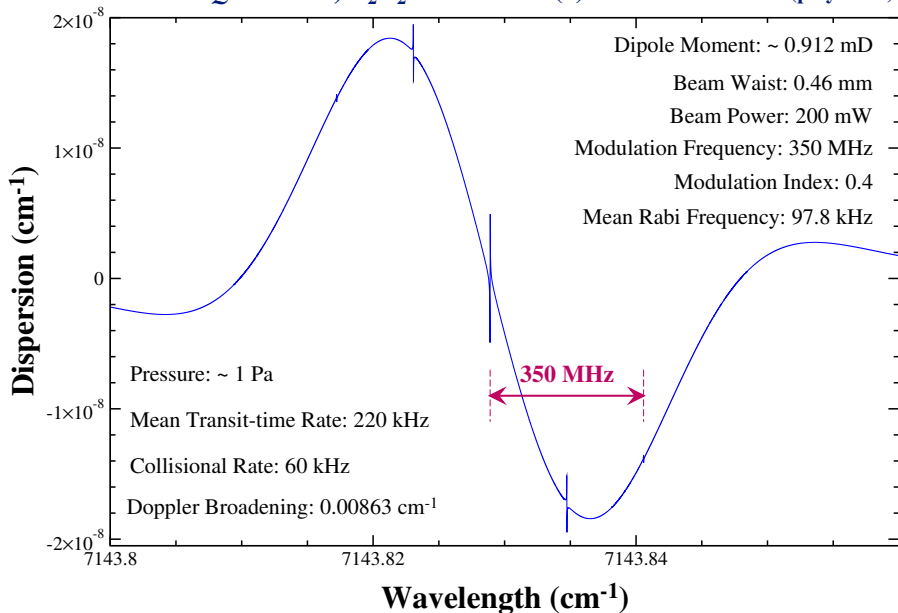
# $C_2H_2$ NICE-OHMS Simulation (Absorption)

*NICE-OHMS in Phase,  $C_2H_2$  Transition R(0) at  $7143.8289\text{ cm}^{-1}$  (polyad 11)*



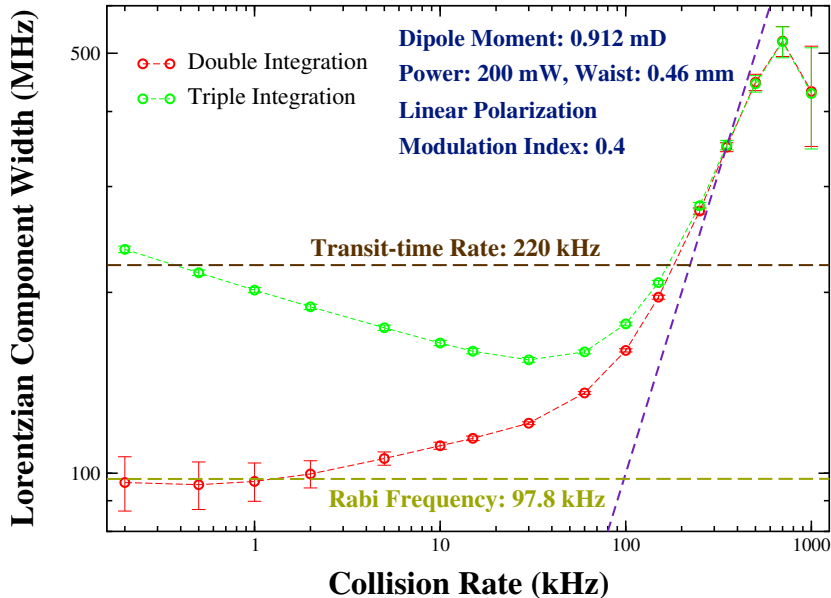
# $C_2H_2$ NICE-OHMS Simulation (Dispersion)

*NICE-OHMS in Quadrature,  $C_2H_2$  Transition R(0) at  $7143.8289\text{ cm}^{-1}$  (polyad 11)*



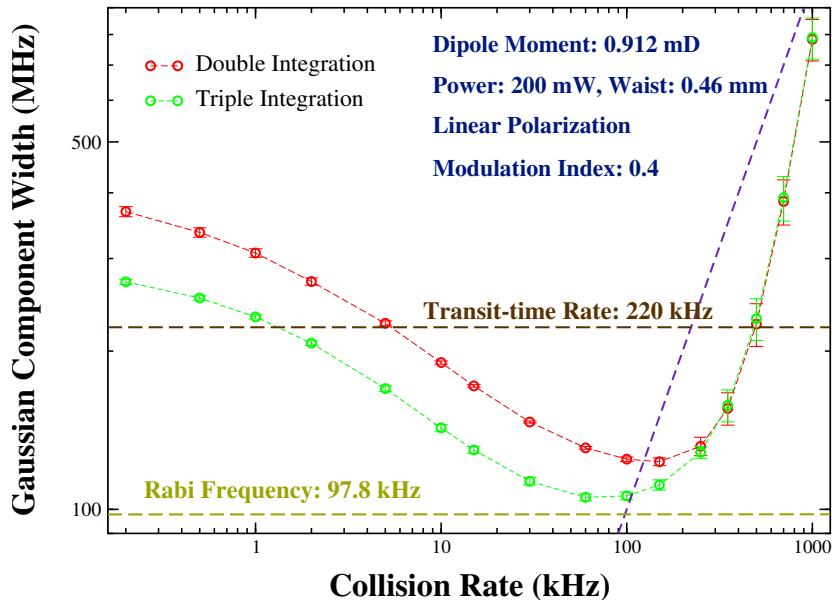
# C<sub>2</sub>H<sub>2</sub> Simulation (Dispersion): Lorentzian Component

Saturation Analysis: C<sub>2</sub>H<sub>2</sub> Transition R(0) at 7143.8289 cm<sup>-1</sup> (polyad 11)



# C<sub>2</sub>H<sub>2</sub> Simulation (Dispersion): Gaussian Component

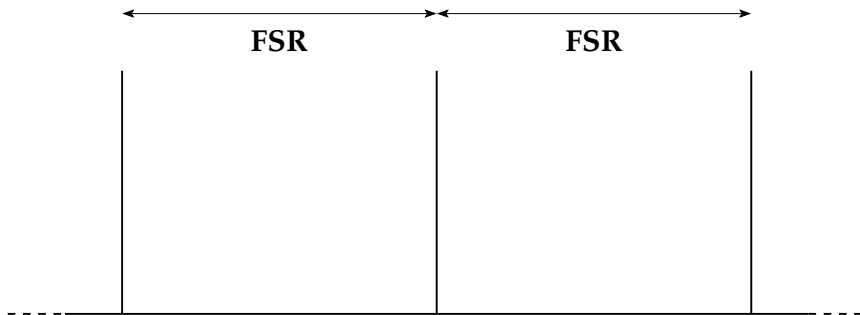
Saturation Analysis: C<sub>2</sub>H<sub>2</sub> Transition R(0) at 7143.8289 cm<sup>-1</sup> (polyad 11)



# Outline

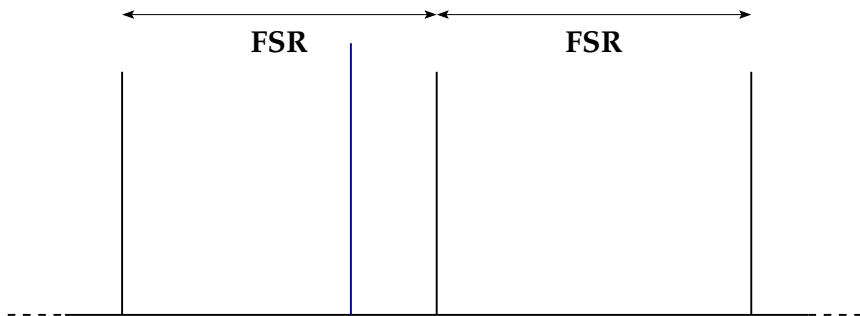
- 1 Motivations
- 2 Introduction to CEAS
- 3 Absorption in Cavity
- 4 Saturated Absorption: Modeling and Simulations
- 5 NICE-OHMS: Principles and Implementation**
- 6  $C_2H_2$  in the NIR
- 7 HD in the NIR (Forbidden Transition)
- 8 Bibliography
- 9 Conclusions

# Frequency Modulation in Cavity



## Phase Modulations in NICE-OHMS

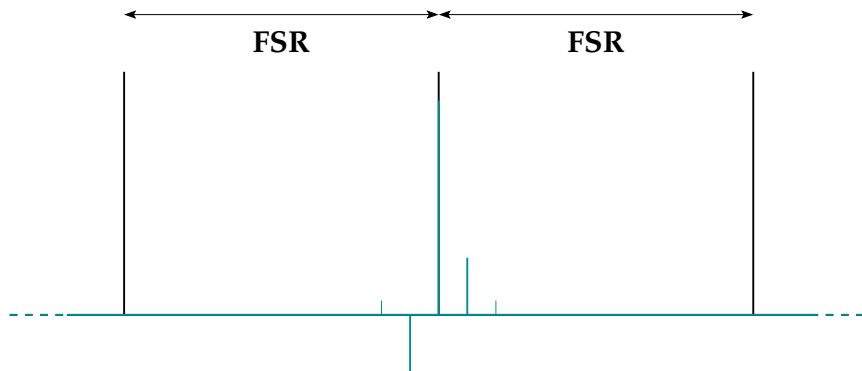
# Frequency Modulation in Cavity



## Phase Modulations in NICE-OHMS

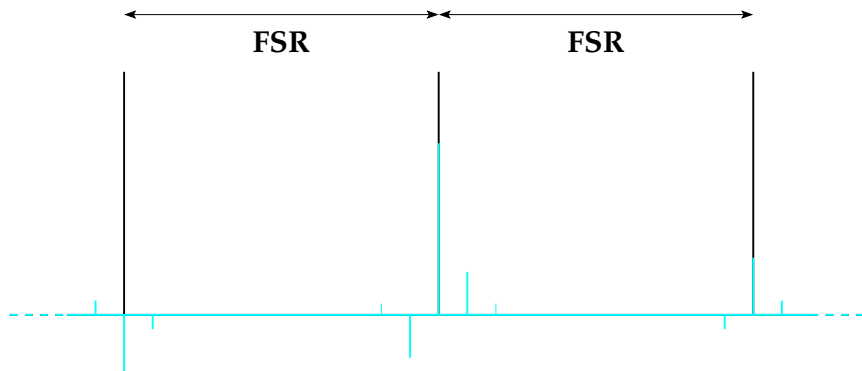


# Frequency Modulation in Cavity



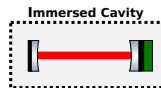
## Phase Modulations in NICE-OHMS

# Frequency Modulation in Cavity

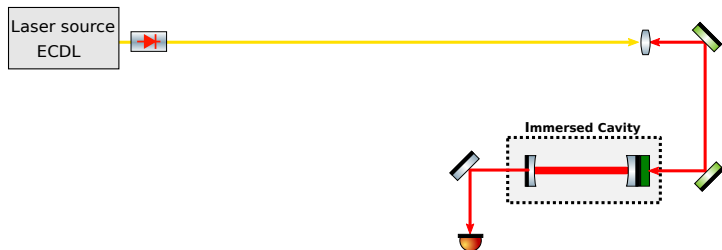


## Phase Modulations in NICE-OHMS

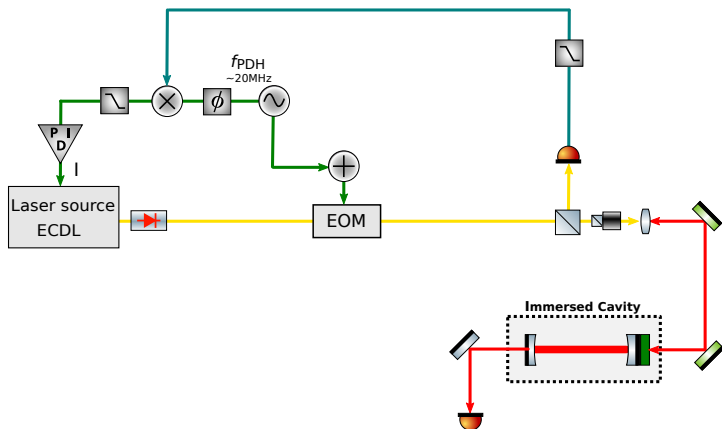
# NICE-OHMS Implementation



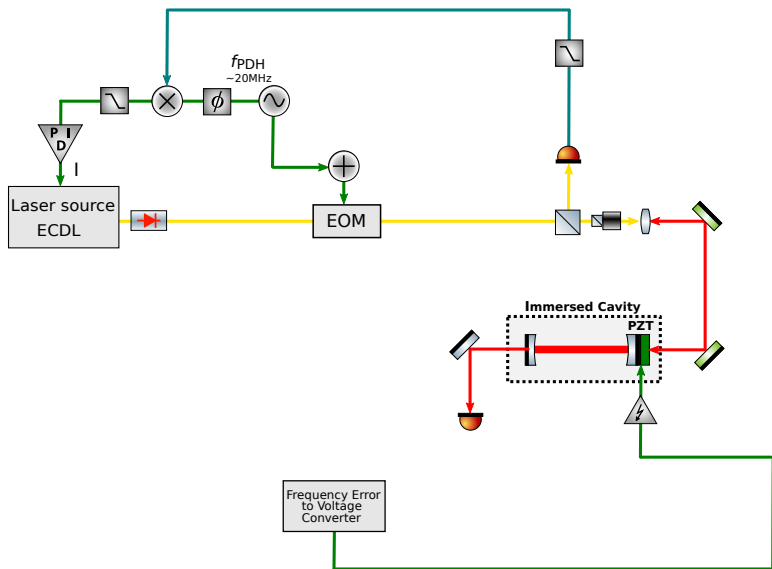
# NICE-OHMS Implementation



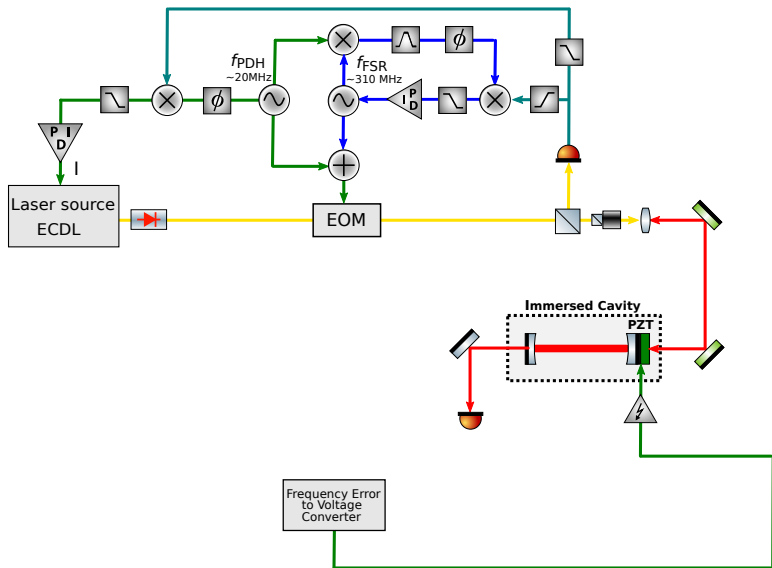
# NICE-OHMS Implementation



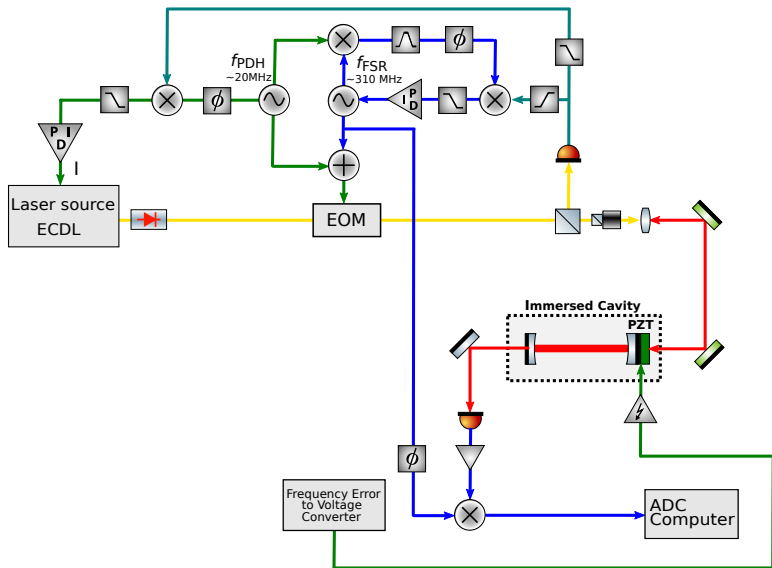
# NICE-OHMS Implementation



# NICE-OHMS Implementation

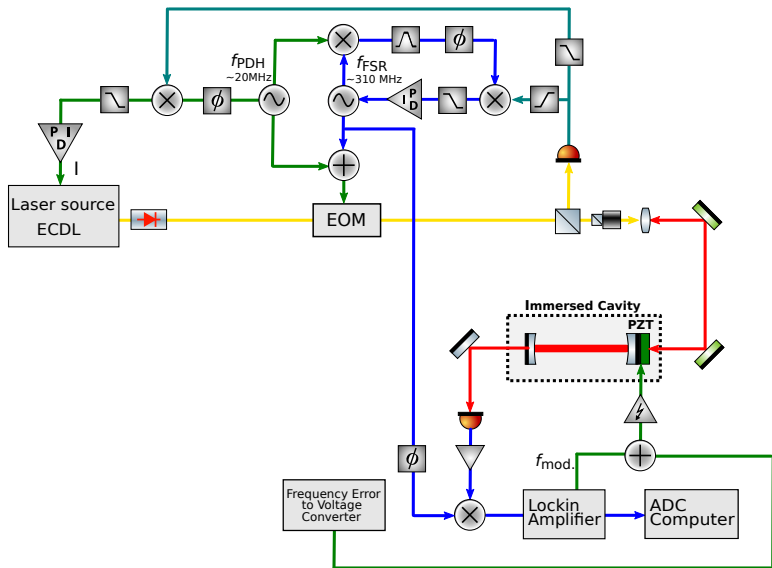


# NICE-OHMS Implementation

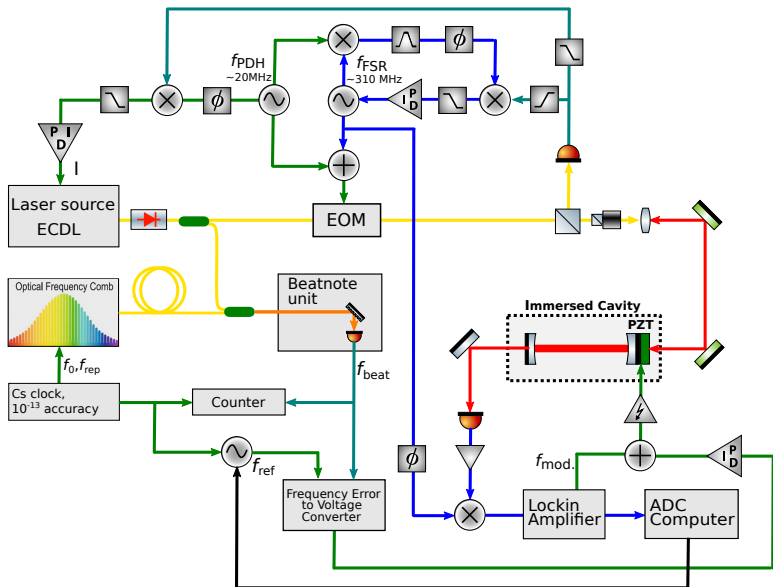




# NICE-OHMS Implementation



# NICE-OHMS Implementation

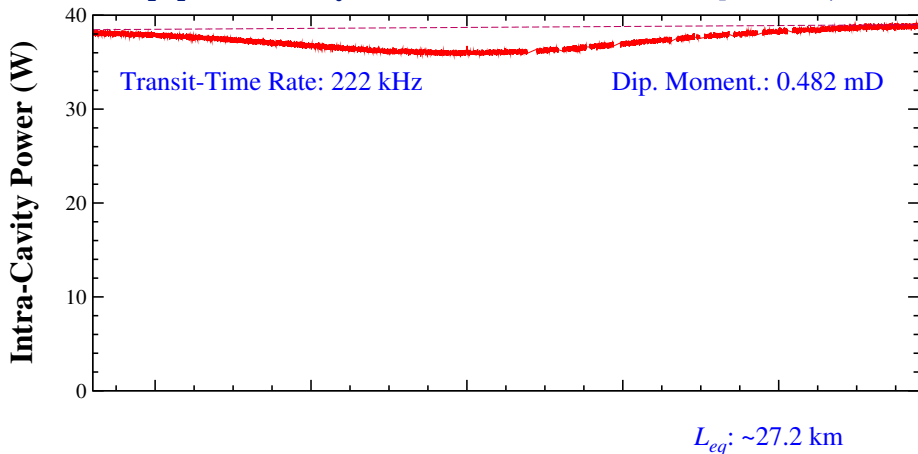


# Outline

- 1 Motivations
- 2 Introduction to CEAS
- 3 Absorption in Cavity
- 4 Saturated Absorption: Modeling and Simulations
- 5 NICE-OHMS: Principles and Implementation
- 6  $C_2H_2$  in the NIR**
- 7 HD in the NIR (Forbidden Transition)
- 8 Bibliography
- 9 Conclusions

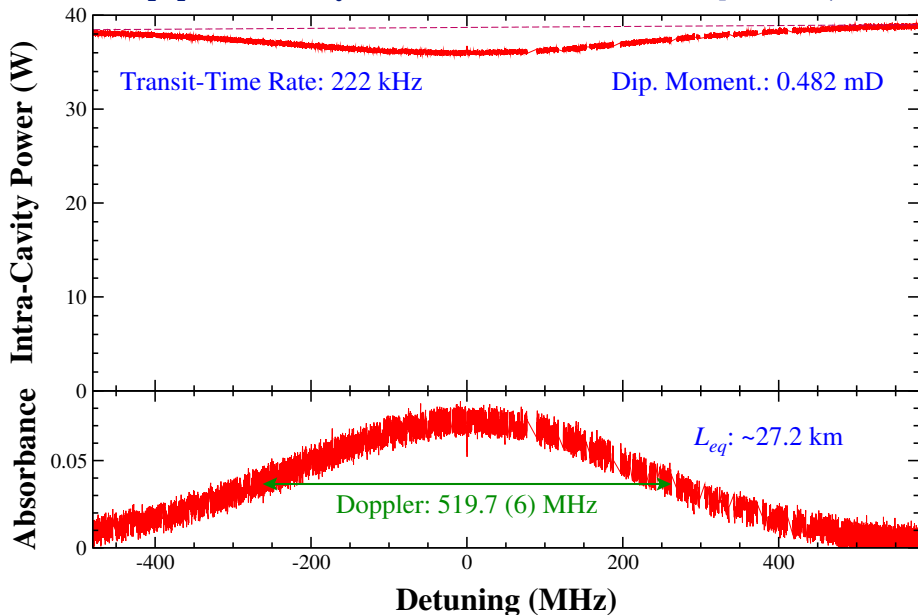
# Direct Absorption of $C_2H_2$ (P 11, $\nu_1 + \nu_2 + (2\nu_4 + \nu_5)^1 \leftarrow 0$ )

$C_2H_2$ , Transition  $R_e(4)$  ( $7239.79077\text{ cm}^{-1}$ ), Direct Absorption at  $7\text{ }\mu\text{bar}$



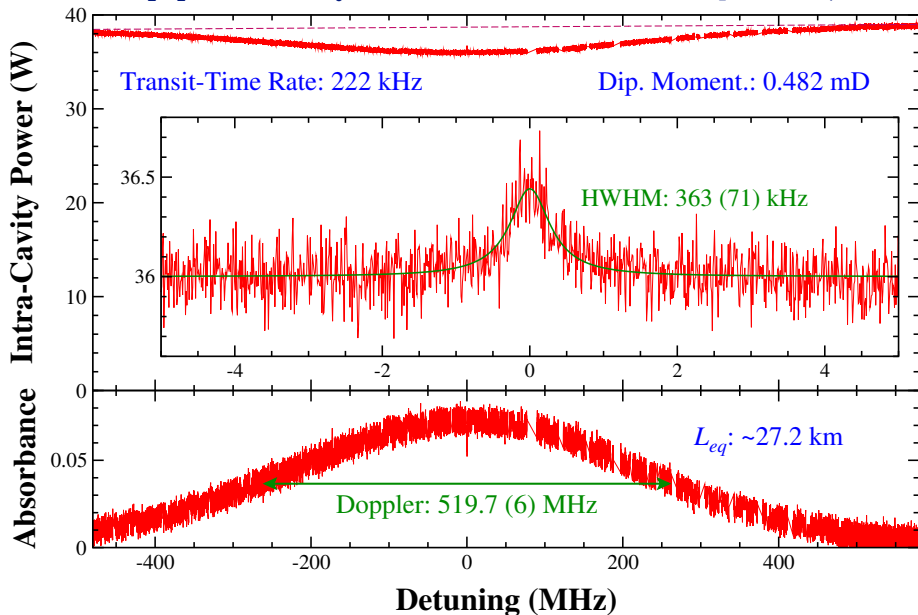
# Direct Absorption of $C_2H_2$ (P 11, $\nu_1 + \nu_2 + (2\nu_4 + \nu_5)^1 \leftarrow 0$ )

$C_2H_2$ , Transition  $R_e(4)$  ( $7239.79077\text{ cm}^{-1}$ ), Direct Absorption at  $7\text{ }\mu\text{bar}$



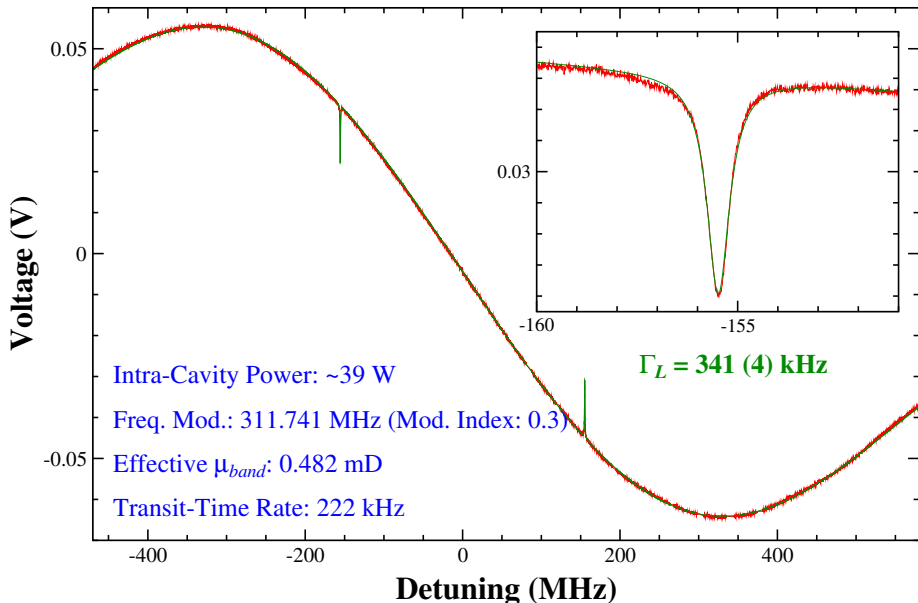
# Direct Absorption of $C_2H_2$ (P 11, $\nu_1 + \nu_2 + (2\nu_4 + \nu_5)^1 \leftarrow 0$ )

$C_2H_2$ , Transition  $R_e(4)$  ( $7239.79077 \text{ cm}^{-1}$ ), Direct Absorption at  $7 \mu\text{bar}$



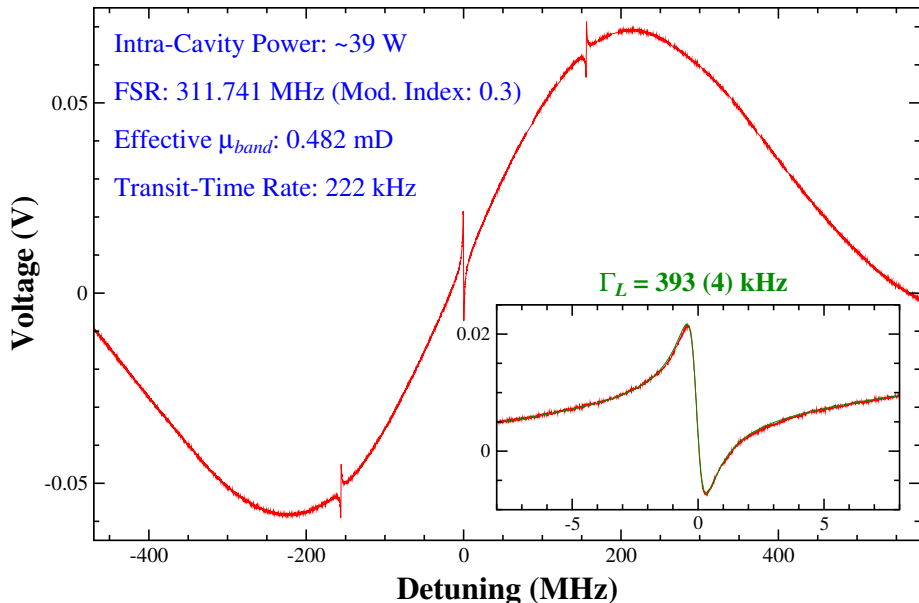
# NICE-OHMS “Absorption” of $C_2H_2$ (Polyad 11)

$C_2H_2$ , Transition  $R_e(4)$  ( $7239.79077\text{ cm}^{-1}$ ), NICE-OHMS in Phase at  $7\text{ }\mu\text{bar}$



# NICE-OHMS “Dispersion” of $C_2H_2$ (Polyad 11)

$C_2H_2$ , Transition  $R_e(4)$  ( $7239.79077\text{ cm}^{-1}$ ), NICE-OHMS in Quadrature at  $7\text{ }\mu\text{bar}$





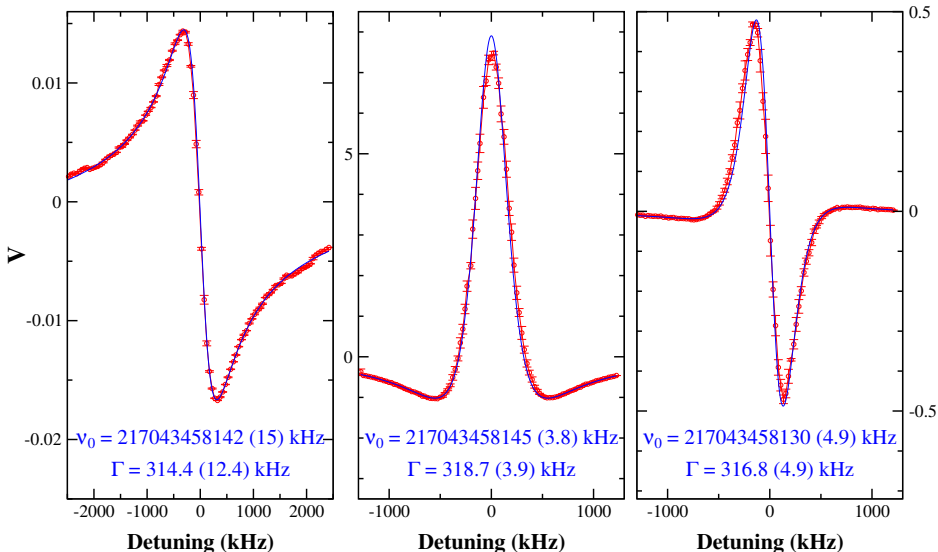
# NICE-OHMS Dispersion of $C_2H_2$ (Polyad 11) with OFC

$C_2H_2$ , Transition  $R_e(4)$  ( $7239.79077\text{ cm}^{-1}$ ), NICE-OHMS in Quadrature at  $2\text{ }\mu\text{bar}$

No Dithering

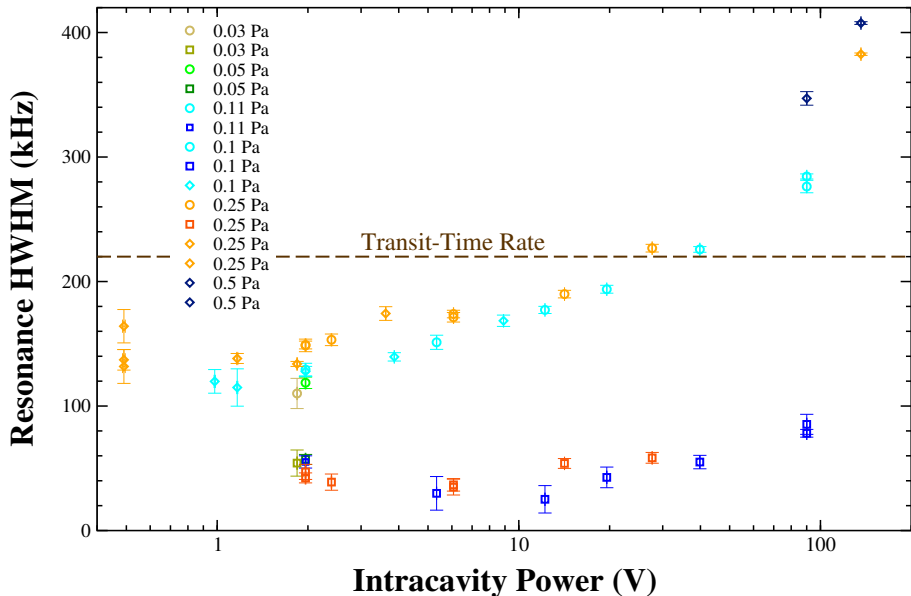
Dithering 1f (596.5 Hz?)

Dithering 2f



# Resonance Width Power Dependence

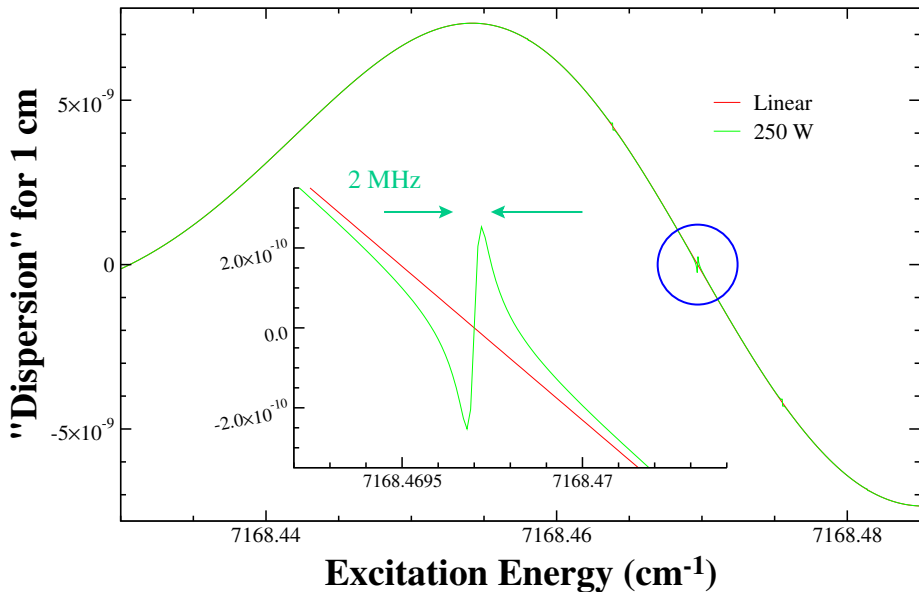
*Power Broadening, NICE-OHMS of C<sub>2</sub>H<sub>2</sub>, Transition R<sub>2</sub>,  $\nu_1 + \nu_2 + (2\nu_4 + \nu_5)^1$*



# Outline

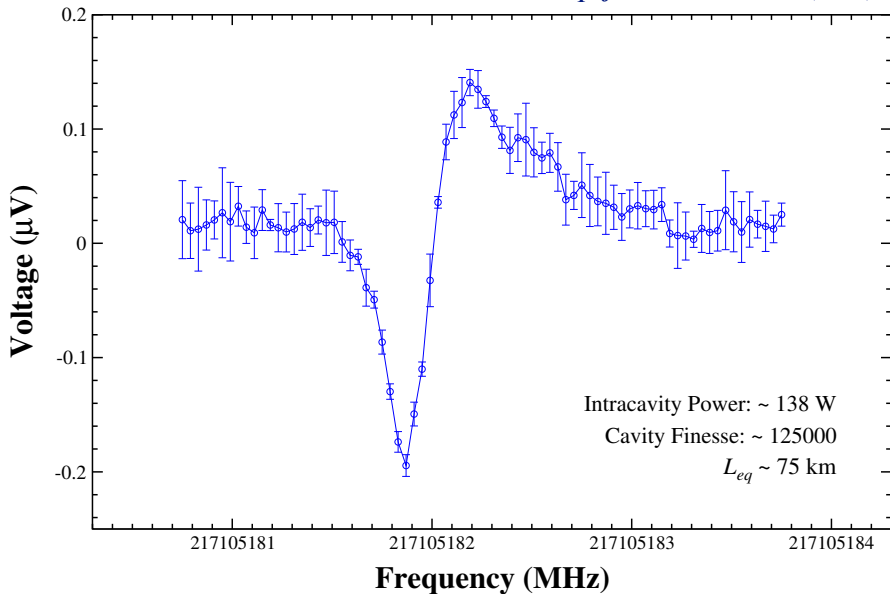
- 1 Motivations
- 2 Introduction to CEAS
- 3 Absorption in Cavity
- 4 Saturated Absorption: Modeling and Simulations
- 5 NICE-OHMS: Principles and Implementation
- 6  $C_2H_2$  in the NIR
- 7 HD in the NIR (Forbidden Transition)**
- 8 Bibliography
- 9 Conclusions

# Simulation of HD: First Overtone, Transition $R(0)$



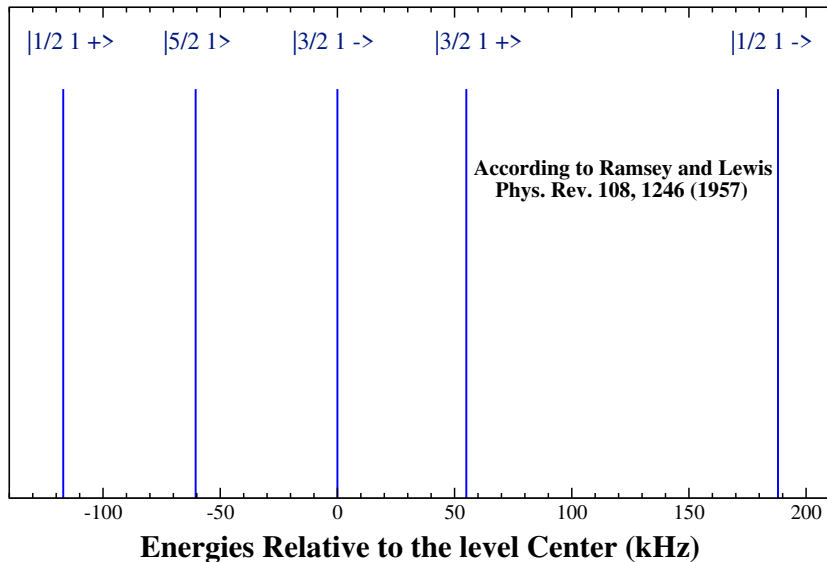
# HD: 1st Overtone, Transition $R(1)$

1-f WM-NICE-OHMS: HD Transition  $R_1$  of the 1<sup>st</sup> Overtone (1 Pa)



# HD Hyperfine Structure according to N. Ramsey

## HD Ground State: Hyperfine Energy Levels for $N=1$



## Discussion on HD (Work in Progress)

- New Transition Frequency: 217.105 181 891 (15) THz,  $< 1 \times 10^{-10}$ ,  
(published value: 217.105 192 (30) THz [Kassi/Campargue JMS, 2011])

## Discussion on HD (Work in Progress)

- New Transition Frequency: 217.105 181 891 (15) THz,  $< 1 \times 10^{-10}$ ,  
(published value: 217.105 192 (30) THz [Kassi/Campargue JMS, 2011])
- Improvement by 3 Orders of Magnitude



## Discussion on HD (Work in Progress)

- New Transition Frequency: 217.105 181 891 (15) THz,  $< 1 \times 10^{-10}$ ,  
(published value: 217.105 192 (30) THz [Kassi/Campargue JMS, 2011])
- Improvement by 3 Orders of Magnitude
- *R(1), R(2), R(3) observed*

## Discussion on HD (Work in Progress)

- New Transition Frequency: 217.105 181 891 (15) THz,  $< 1 \times 10^{-10}$ ,  
(published value: 217.105 192 (30) THz [Kassi/Campargue JMS, 2011])
- Improvement by 3 Orders of Magnitude
- $R(1)$ ,  $R(2)$ ,  $R(3)$  observed
- Sensitivity  $\sim 10^{-12} \text{ cm}^{-1}$

## Discussion on HD (Work in Progress)

- New Transition Frequency: 217.105 181 891 (15) THz,  $< 1 \times 10^{-10}$ ,  
(published value: 217.105 192 (30) THz [Kassi/Campargue JMS, 2011])
- Improvement by 3 Orders of Magnitude
- $R(1)$ ,  $R(2)$ ,  $R(3)$  observed
- Sensitivity  $\sim 10^{-12} \text{ cm}^{-1}$
- Resonance Line Shape (Asymmetry)?

## Discussion on HD (Work in Progress)

- New Transition Frequency: 217.105 181 891 (15) THz,  $< 1 \times 10^{-10}$ ,  
(published value: 217.105 192 (30) THz [Kassi/Campargue JMS, 2011])
- Improvement by 3 Orders of Magnitude
- $R(1)$ ,  $R(2)$ ,  $R(3)$  observed
- Sensitivity  $\sim 10^{-12} \text{ cm}^{-1}$
- Resonance Line Shape (Asymmetry)?
  - Mean Transit-Time Rate:  $\sim 660 \text{ kHz}$

## Discussion on HD (Work in Progress)

- New Transition Frequency: 217.105 181 891 (15) THz,  $< 1 \times 10^{-10}$ ,  
(published value: 217.105 192 (30) THz [Kassi/Campargue JMS, 2011])
- Improvement by 3 Orders of Magnitude
- $R(1)$ ,  $R(2)$ ,  $R(3)$  observed
- Sensitivity  $\sim 10^{-12} \text{ cm}^{-1}$
- Resonance Line Shape (Asymmetry)?
  - Mean Transit-Time Rate:  $\sim 660 \text{ kHz}$
  - Rabi Frequency ( $\sim 21 \text{ kHz}$ )?

## Discussion on HD (Work in Progress)

- New Transition Frequency: 217.105 181 891 (15) THz,  $< 1 \times 10^{-10}$ ,  
(published value: 217.105 192 (30) THz [Kassi/Campargue JMS, 2011])
- Improvement by 3 Orders of Magnitude
- $R(1)$ ,  $R(2)$ ,  $R(3)$  observed
- Sensitivity  $\sim 10^{-12} \text{ cm}^{-1}$
- Resonance Line Shape (Asymmetry)?
  - Mean Transit-Time Rate:  $\sim 660 \text{ kHz}$
  - Rabi Frequency ( $\sim 21 \text{ kHz}$ )?
  - Recoil ( $\sim 35 \text{ kHz}$ )?

## Discussion on HD (Work in Progress)

- New Transition Frequency: 217.105 181 891 (15) THz,  $< 1 \times 10^{-10}$ , (published value: 217.105 192 (30) THz [Kassi/Campargue JMS, 2011])
- Improvement by 3 Orders of Magnitude
- $R(1)$ ,  $R(2)$ ,  $R(3)$  observed
- Sensitivity  $\sim 10^{-12} \text{ cm}^{-1}$
- Resonance Line Shape (Asymmetry)?
  - Mean Transit-Time Rate:  $\sim 660 \text{ kHz}$
  - Rabi Frequency ( $\sim 21 \text{ kHz}$ )?
  - Recoil ( $\sim 35 \text{ kHz}$ )?
  - Evidence of the Hyperfine Structure (spreads over 600 kHz)?

## Discussion on HD (Work in Progress)

- New Transition Frequency: 217.105 181 891 (15) THz,  $< 1 \times 10^{-10}$ , (published value: 217.105 192 (30) THz [Kassi/Campargue JMS, 2011])
- Improvement by 3 Orders of Magnitude
- $R(1)$ ,  $R(2)$ ,  $R(3)$  observed
- Sensitivity  $\sim 10^{-12} \text{ cm}^{-1}$
- Resonance Line Shape (Asymmetry)?
  - Mean Transit-Time Rate:  $\sim 660 \text{ kHz}$
  - Rabi Frequency ( $\sim 21 \text{ kHz}$ )?
  - Recoil ( $\sim 35 \text{ kHz}$ )?
  - Evidence of the Hyperfine Structure (spreads over 600 kHz)?
- Pressure Broadening Coefficient?



## Discussion on HD (Work in Progress)

- New Transition Frequency: 217.105 181 891 (15) THz,  $< 1 \times 10^{-10}$ , (published value: 217.105 192 (30) THz [Kassi/Campargue JMS, 2011])
- Improvement by 3 Orders of Magnitude
- $R(1)$ ,  $R(2)$ ,  $R(3)$  observed
- Sensitivity  $\sim 10^{-12} \text{ cm}^{-1}$
- Resonance Line Shape (Asymmetry)?
  - Mean Transit-Time Rate:  $\sim 660 \text{ kHz}$
  - Rabi Frequency ( $\sim 21 \text{ kHz}$ )?
  - Recoil ( $\sim 35 \text{ kHz}$ )?
  - Evidence of the Hyperfine Structure (spreads over 600 kHz)?
- Pressure Broadening Coefficient?
- Pressure Shift?

## Discussion on HD (Work in Progress)

- New Transition Frequency: 217.105 181 891 (15) THz,  $< 1 \times 10^{-10}$ , (published value: 217.105 192 (30) THz [Kassi/Campargue JMS, 2011])
- Improvement by 3 Orders of Magnitude
- $R(1)$ ,  $R(2)$ ,  $R(3)$  observed
- Sensitivity  $\sim 10^{-12} \text{ cm}^{-1}$
- Resonance Line Shape (Asymmetry)?
  - Mean Transit-Time Rate:  $\sim 660 \text{ kHz}$
  - Rabi Frequency ( $\sim 21 \text{ kHz}$ )?
  - Recoil ( $\sim 35 \text{ kHz}$ )?
  - Evidence of the Hyperfine Structure (spreads over 600 kHz)?
- Pressure Broadening Coefficient?
- Pressure Shift?
- Comparison with CRDS (S. Hu group, in PRL)

## Discussion on HD (Work in Progress)

- New Transition Frequency: 217.105 181 891 (15) THz,  $< 1 \times 10^{-10}$ , (published value: 217.105 192 (30) THz [Kassi/Campargue JMS, 2011])
- Improvement by 3 Orders of Magnitude
- $R(1)$ ,  $R(2)$ ,  $R(3)$  observed
- Sensitivity  $\sim 10^{-12} \text{ cm}^{-1}$
- Resonance Line Shape (Asymmetry)?
  - Mean Transit-Time Rate:  $\sim 660 \text{ kHz}$
  - Rabi Frequency ( $\sim 21 \text{ kHz}$ )?
  - Recoil ( $\sim 35 \text{ kHz}$ )?
  - Evidence of the Hyperfine Structure (spreads over 600 kHz)?
- Pressure Broadening Coefficient?
- Pressure Shift?
- Comparison with CRDS (S. Hu group, in PRL)
-

# Outline

- 1 Motivations
- 2 Introduction to CEAS
- 3 Absorption in Cavity
- 4 Saturated Absorption: Modeling and Simulations
- 5 NICE-OHMS: Principles and Implementation
- 6  $C_2H_2$  in the NIR
- 7 HD in the NIR (Forbidden Transition)
- 8 Bibliography**
- 9 Conclusions

# Bibliography

- Broad-Band Cavity Ring-Down Spectroscopy, Chem. Rev., 103, 5239 (2003)
- Chemical Sensing Using Fiber Cavity Ring-Down Spectroscopy, Sensors, 10, 1716 (2010)
- Probing Molecular Species by Cavity Ringdown Laser Absorption Spectroscopy, Application to the Spectroscopy and Dynamics of Jet-Cooled NO<sub>2</sub>, CR. Acad. Sci. Paris Série IV, 2, 929 (2001)
- Cavity-Enhanced Spectroscopy and Sensing, Ed. by G. Gagliardi and H.-P. Loock, Springer (2014)
- Cavity Ringdown Spectroscopy: An Ultratrace-Absorption Measurement Technique, Ed. by K.W. Busch and M.A. Busch, ACS Symposium Series, 1999
- Sub-Doppler frequency metrology in HD for test of fundamental physics, Cozijn et al., Phys. Rev. Letters, 120, 153002 (2018)
- Toward a Determination of the Proton-Electron Mass Ratio from the Lamb-Dip Measurement of HD, Tao et al. Phys. Rev. Letters, 120, 153001 (2018)

# Outline

- 1 Motivations
- 2 Introduction to CEAS
- 3 Absorption in Cavity
- 4 Saturated Absorption: Modeling and Simulations
- 5 NICE-OHMS: Principles and Implementation
- 6  $C_2H_2$  in the NIR
- 7 HD in the NIR (Forbidden Transition)
- 8 Bibliography
- 9 Conclusions**

# Conclusions (NICE-OHMS in Amsterdam)

- **Comparing NICE-OHMS vs CRDS**

# Conclusions (NICE-OHMS in Amsterdam)

- **Comparing NICE-OHMS vs CRDS**
- **Work in Progress**



# Conclusions (NICE-OHMS in Amsterdam)

- **Comparing NICE-OHMS vs CRDS**
- **Work in Progress**
- **An Innovative Technique, even to Saturate weak Transitions**

# Conclusions (NICE-OHMS in Amsterdam)

- **Comparing NICE-OHMS vs CRDS**
- **Work in Progress**
- **An Innovative Technique, even to Saturate weak Transitions**
- **HD:  $R(0)$ ,  $P(1)$ , etc..**

## Conclusions (NICE-OHMS in Amsterdam)

- Comparing NICE-OHMS vs CRDS
- Work in Progress
- An Innovative Technique, even to Saturate weak Transitions
- HD:  $R(0)$ ,  $P(1)$ , etc..
- Higher Finesse: from  $\sim 125\,000$  to  $500\,000$ ?

# Conclusions (NICE-OHMS in Amsterdam)

- **Comparing NICE-OHMS vs CRDS**
- **Work in Progress**
- **An Innovative Technique, even to Saturate weak Transitions**
- **HD:  $R(0)$ ,  $P(1)$ , etc..**
- **Higher Finesse: from  $\sim 125\,000$  to  $500\,000$ ?**
- **Full Validation of the NICE-OHMS Technique on  $C_2H_2$**

## Conclusions (NICE-OHMS in Amsterdam)

- **Comparing NICE-OHMS vs CRDS**
- **Work in Progress**
- **An Innovative Technique, even to Saturate weak Transitions**
- **HD:  $R(0)$ ,  $P(1)$ , etc..**
- **Higher Finesse: from  $\sim 125\,000$  to  $500\,000$ ?**
- **Full Validation of the NICE-OHMS Technique on  $C_2H_2$**
- **Saturation Model Validation?**

# Conclusions (NICE-OHMS in Amsterdam)

- **Comparing NICE-OHMS vs CRDS**
- **Work in Progress**
- **An Innovative Technique, even to Saturate weak Transitions**
- **HD:  $R(0)$ ,  $P(1)$ , etc..**
- **Higher Finesse: from  $\sim 125\,000$  to  $500\,000$ ?**
- **Full Validation of the NICE-OHMS Technique on  $C_2H_2$**
- **Saturation Model Validation?**
- **Line Shape asymmetries: Role of the Hyperfine Structure?**

# Conclusions (NICE-OHMS in Amsterdam)

- **Comparing NICE-OHMS vs CRDS**
- **Work in Progress**
- **An Innovative Technique, even to Saturate weak Transitions**
- **HD:  $R(0)$ ,  $P(1)$ , etc..**
- **Higher Finesse: from  $\sim 125\,000$  to  $500\,000$ ?**
- **Full Validation of the NICE-OHMS Technique on  $C_2H_2$**
- **Saturation Model Validation?**
- **Line Shape asymmetries: Role of the Hyperfine Structure?**
- **Resonance Narrowing?**

# Conclusions (NICE-OHMS in Amsterdam)

- **Comparing NICE-OHMS vs CRDS**
- **Work in Progress**
- **An Innovative Technique, even to Saturate weak Transitions**
- **HD:  $R(0)$ ,  $P(1)$ , etc..**
- **Higher Finesse: from  $\sim 125\,000$  to  $500\,000$ ?**
- **Full Validation of the NICE-OHMS Technique on  $C_2H_2$**
- **Saturation Model Validation?**
- **Line Shape asymmetries: Role of the Hyperfine Structure?**
- **Resonance Narrowing?**
- **Sub-kHz Precision Range?**



# Conclusions (NICE-OHMS in Amsterdam)

- **Comparing NICE-OHMS vs CRDS**
- **Work in Progress**
- **An Innovative Technique, even to Saturate weak Transitions**
- **HD:  $R(0)$ ,  $P(1)$ , etc..**
- **Higher Finesse: from  $\sim 125\,000$  to  $500\,000$ ?**
- **Full Validation of the NICE-OHMS Technique on  $C_2H_2$**
- **Saturation Model Validation?**
- **Line Shape asymmetries: Role of the Hyperfine Structure?**
- **Resonance Narrowing?**
- **Sub-kHz Precision Range?**
- **Etc..**

# Acknowledgments

Thank for your Attention



**Frank Cozijn, Edcel Salumbides & Wim Ubachs**