External Cavity Quantum Cascade Lasers: Recent Advances and Applications

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OUTLINE

- Motivation
- Quantum Cascade Lasers (QCLs)
- External Cavity QCLs
  - Performance characteristics
  - Advantages and disadvantages in spectroscopic sensing
- Example Applications of EC-QCLs
- Preliminary results and tests of a new EC-QCL design
- Summary and Future Directions

IQCLSW '08

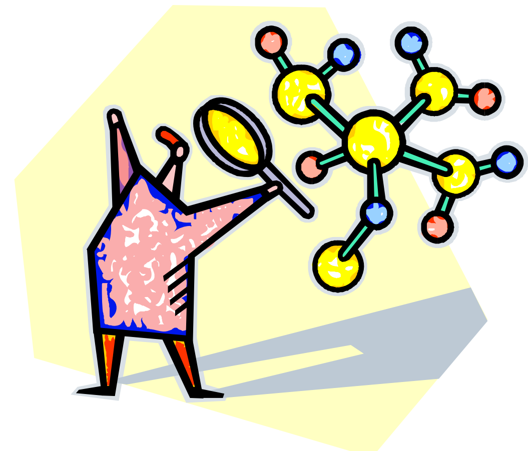
September 14-19, 2008
Monte Verita, Switzerland

Financial Support: DoE-STTR and NSF - MIRTHE

Trace Gas Sensing Applications

Environmental Monitoring

Fundamental Science



Urban and Industrial Emission Measurements

Industrial Process Control

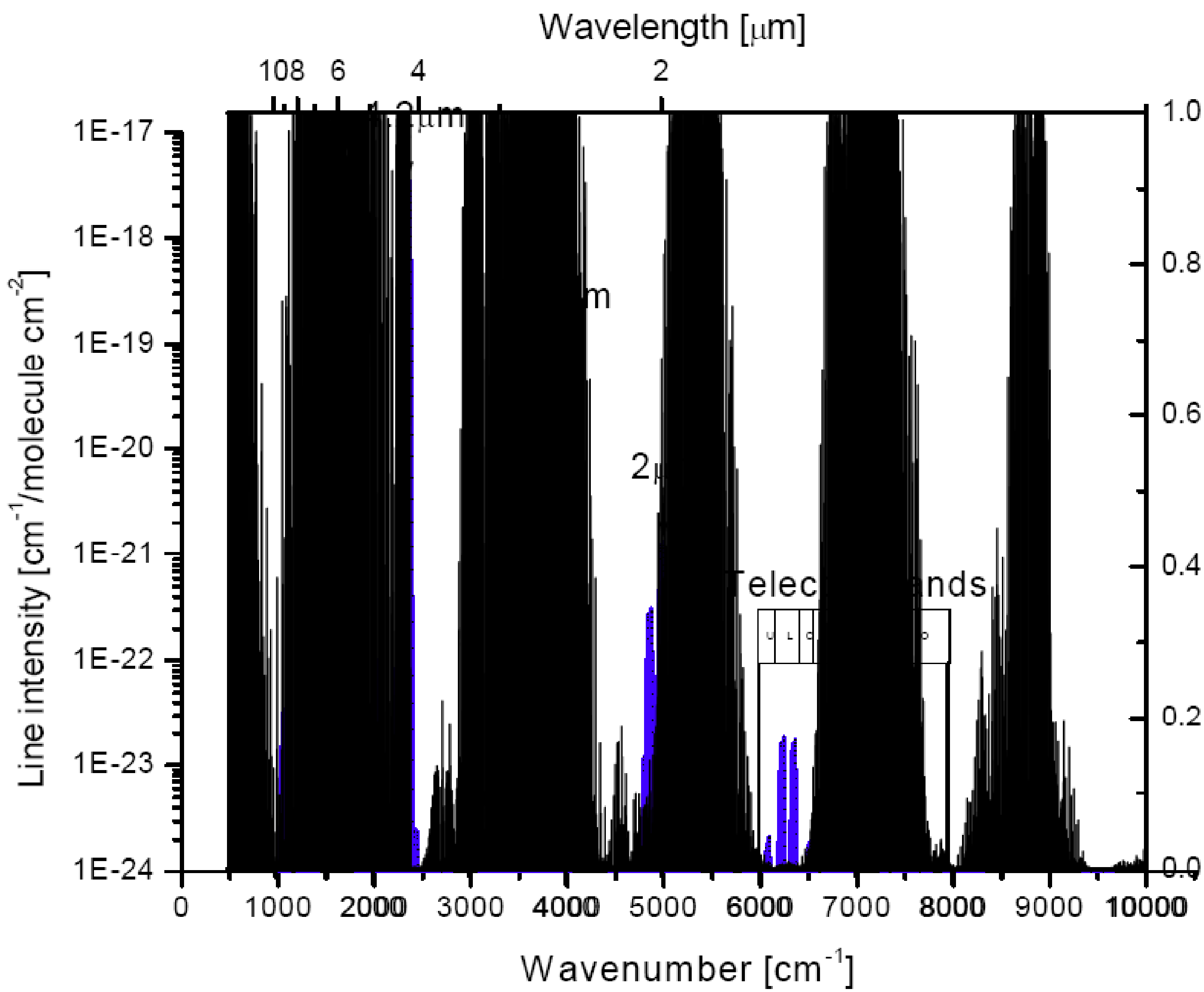
Applications in Medicine and Life Sciences

Law Enforcement and National Security

Laser Absorption Spectroscopy

- High sensitivity
- High selectivity
- Non-destructive
- Fast
- No sample preparation
- Remote sensing
- Field deployable

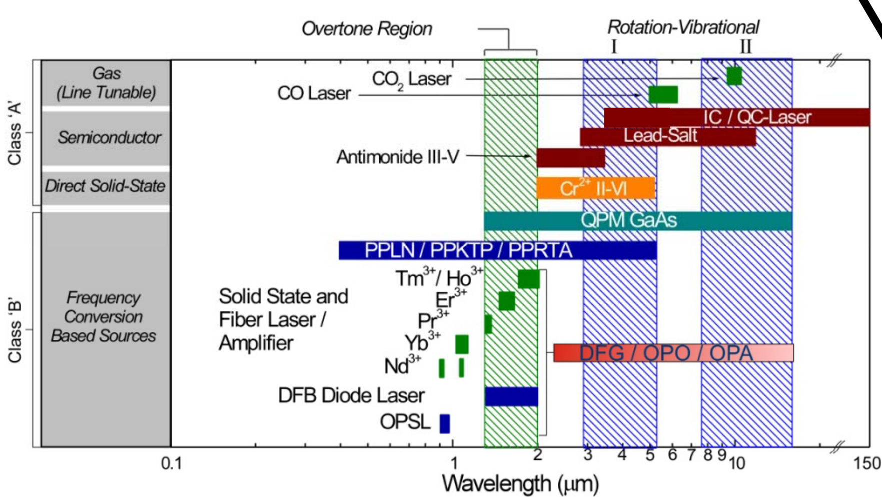
CO_2 absorption spectrum



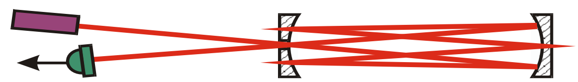
Source: HITRAN 2000 database

Spectroscopic techniques for trace-gas detection

LASER SOURCE

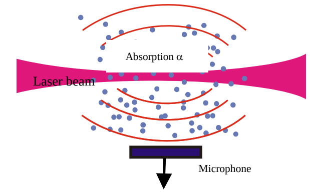


Multipass cell spectroscopy

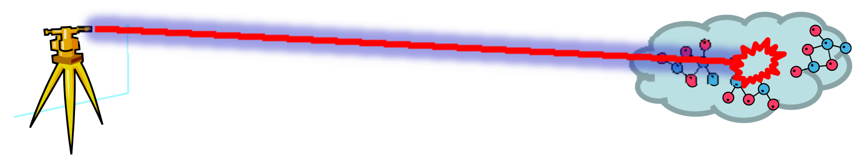


Cavity Ring Down Spectroscopy (CRDS)
Cavity Enhanced Spectroscopy

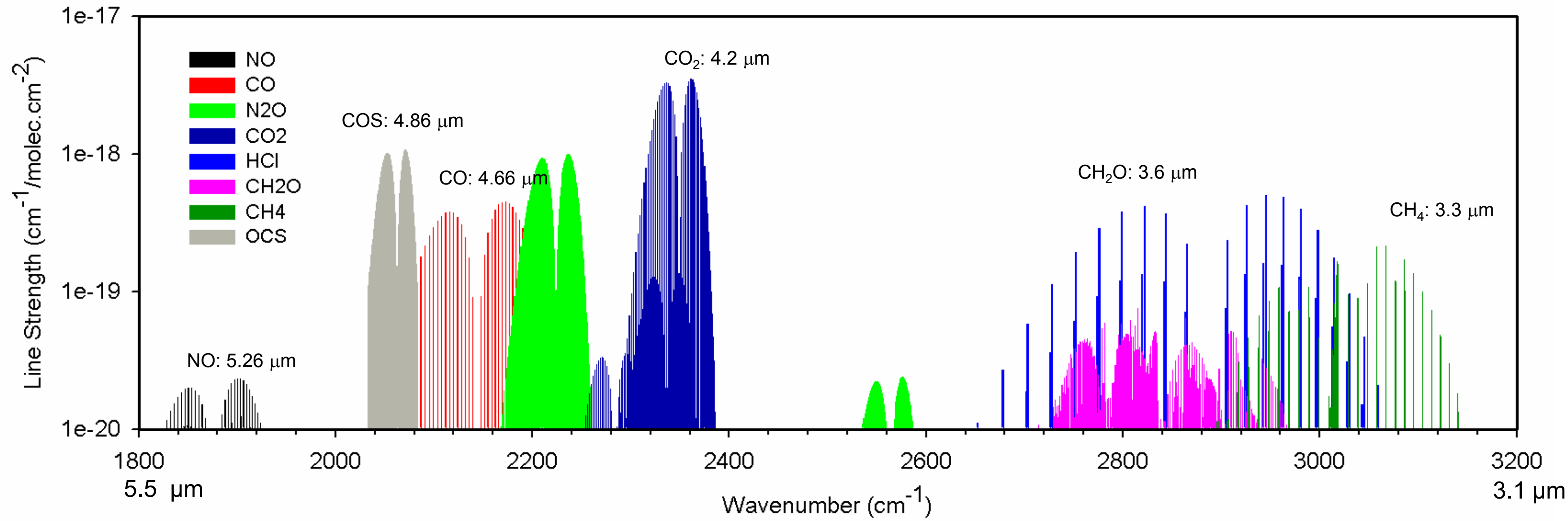
Photoaccoustic Spectroscopy

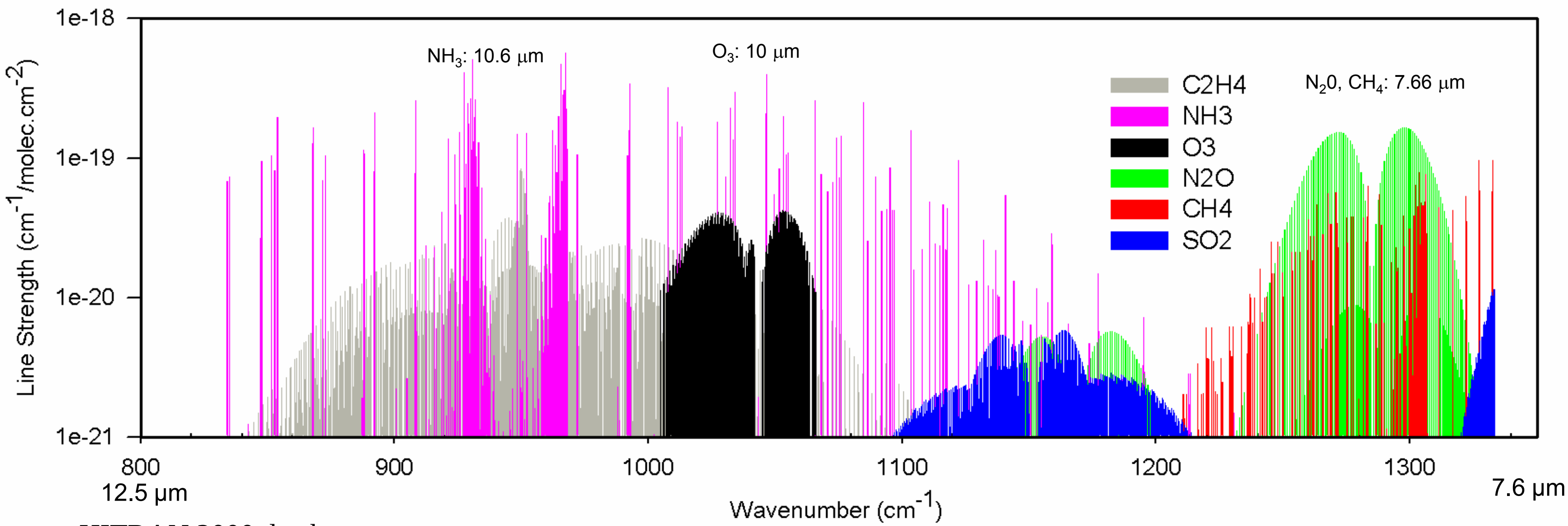


Remote sensing



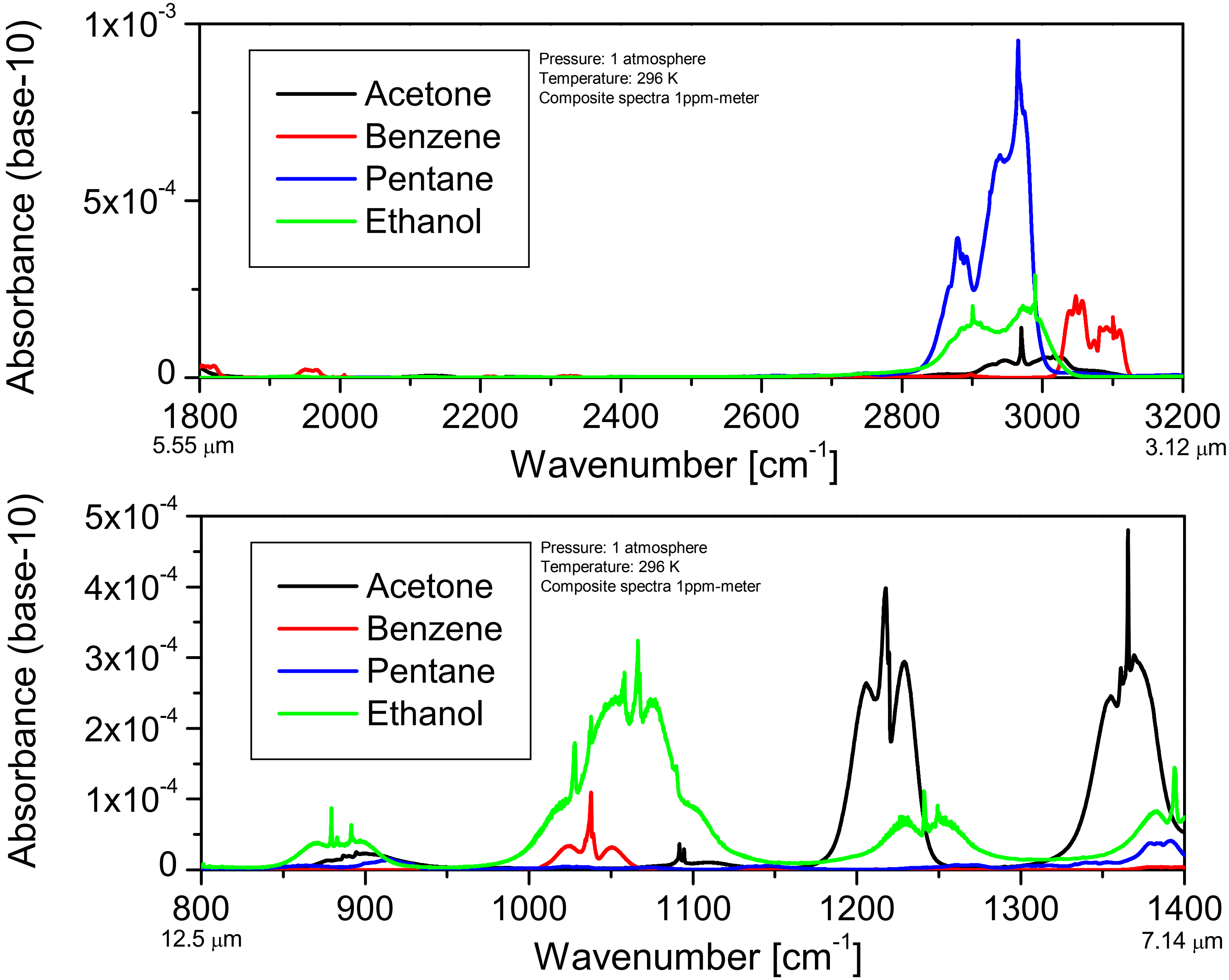
Example Molecular Absorption Spectra within Mid-IR "Atmospheric Windows"





Source: HITRAN 2000 database

Example Absorption Spectra of Broadband Absorbing Molecules



Source: PNNL spectroscopic database

Breath Biomarkers in Humans

As many as 400 different molecules in breath, many with well defined biochemical pathways

BROADBAND ABSORBERS

Compound	Concentration	Physiological basis / Pathology Indication
Acetaldehyde	ppb	Ethanol metabolism
Acetone	ppm	Decarboxylation of acetoacetate, diabetes
Ammonia	ppb	protein metabolism, liver and renal disease
Carbon dioxide	%	Product of respiration, Heliobacter pylori
Carbon disulfide	ppb	Gut bacteria, schizophrenia
Carbon monoxide	ppm	Production catalyzed by *heme oxygenase*
Carbonyl sulfide	ppb	Gut bacteria, liver disease
Ethane	ppb	Lipid peroxidation and oxidative stress
Ethanol	ppb	Gut bacteria
Ethylene	ppb	Lipid peroxidation, oxidative stress, cancer
Hydrocarbons	ppb	Lipid peroxidation/metabolism
Hydrogen	ppm	Gut bacteria
Isoprene	ppb	Cholesterol biosynthesis
Methane	ppm	Gut bacteria
Methanethiol	ppb	Methionine metabolism
Methanol	ppb	Metabolism of fruit
Methylamine	ppb	Protein metabolism
Nitric oxide	ppb	Production catalyzed by *nitric oxide synthase*
Oxygen	%	Required for normal respiration
Pentane	ppb	Lipid peroxidation, oxidative stress
Water	%	Product of respiration

Terence Risby, Johns Hopkins University

Mid-IR Source Requirements for Laser Spectroscopy

SPECTROSCOPIC GOALS	REQUIRED LASER PERFORMANCE
Sensitivity (% to ppt)	Wavelength, Power
Selectivity (Spectral Resolution)	Single Mode Operation and Narrow Linewidth
Multiple Molecular Targets, Broadband Absorbers	Tunable Wavelength
Remote sensing (directionality) or High-Finesse Cavity (mode-matching)	High Beam Quality
Rapid Data Acquisition	Fast Wavelength Tuning/ Modulation capabilities
Autonomous, no consumables	No cryogenic cooling
Field deployment	Compact & Robust

Quantum Cascade Laser: Basic Facts

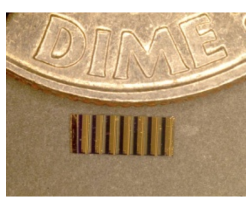
- Laser wavelengths cover the Mid-IR range (~3 – 24μm, band structure engineering)
- High laser power

  (>500mW cw, >5W peak for pulsed)
- Tunable single frequency operation

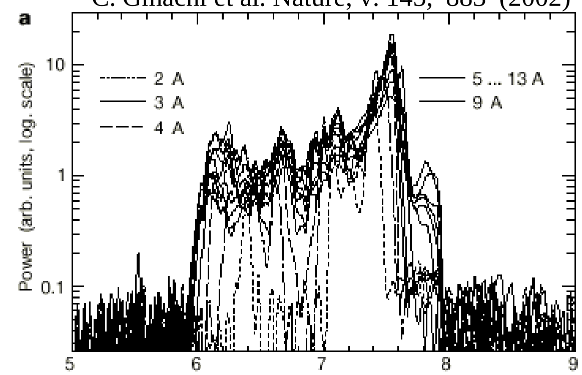
  tuning: DFB (up to ~10 cm^{-1}), EC (>200 cm^{-1})
- High quantum efficiency

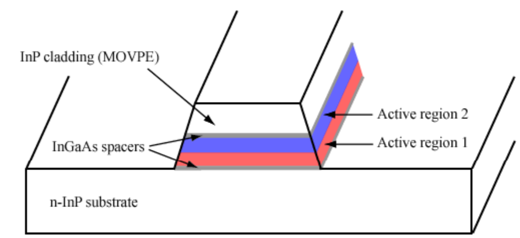
  (Cascading: 1 electron = N photons)
- High reliability, long lifetime
- Room temperature operation

  (CW: above RT)
- Compact

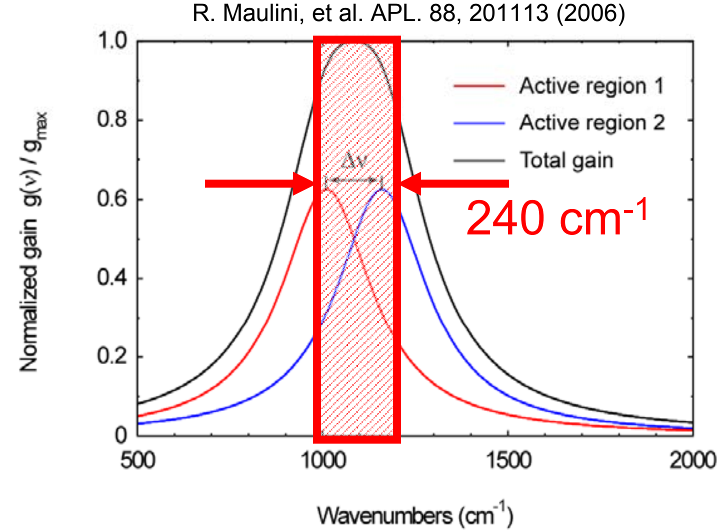


C. Gmachl et al. Nature, v. 145, 883 (2002)

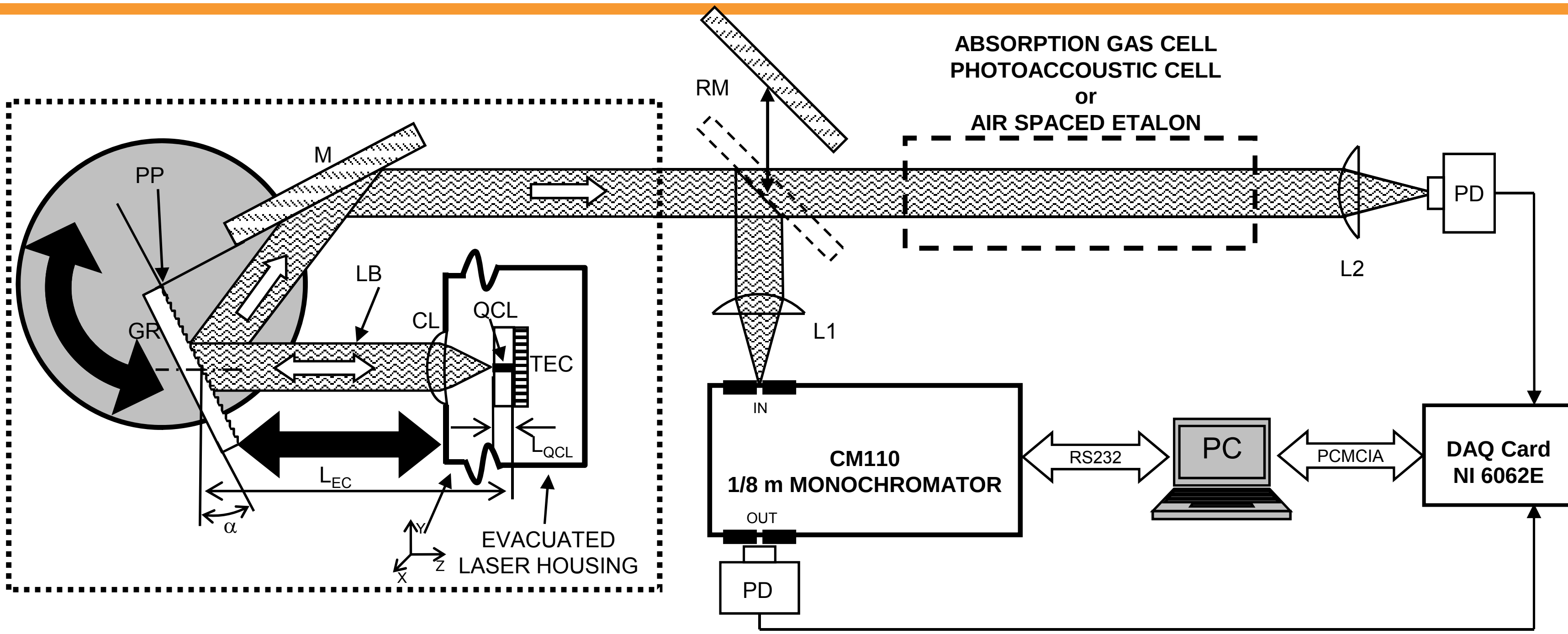


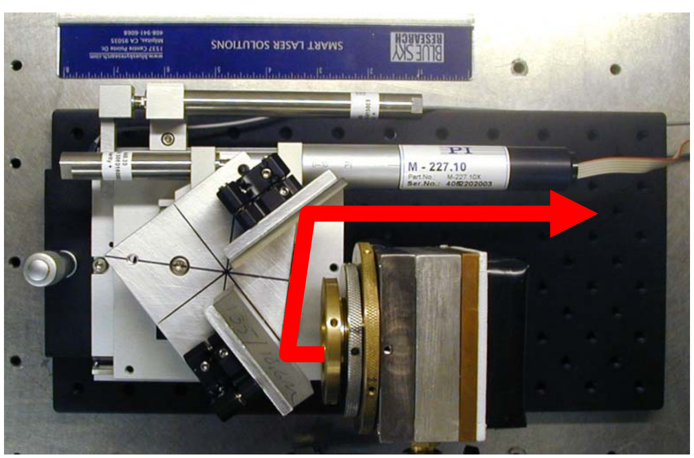


R. Maulini, et al. APL. 88, 201113 (2006)



Tunable external cavity QCL based spectrometer

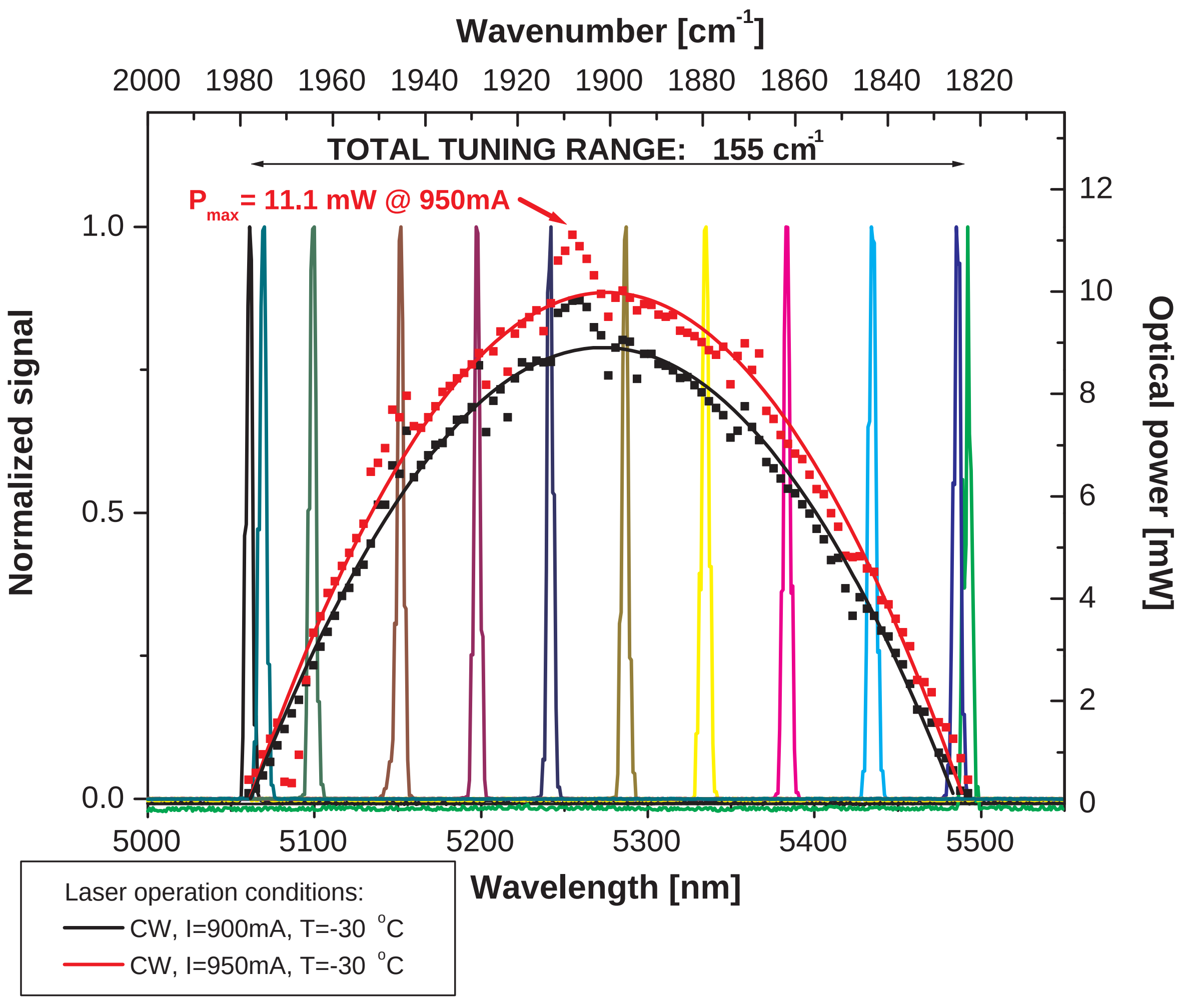




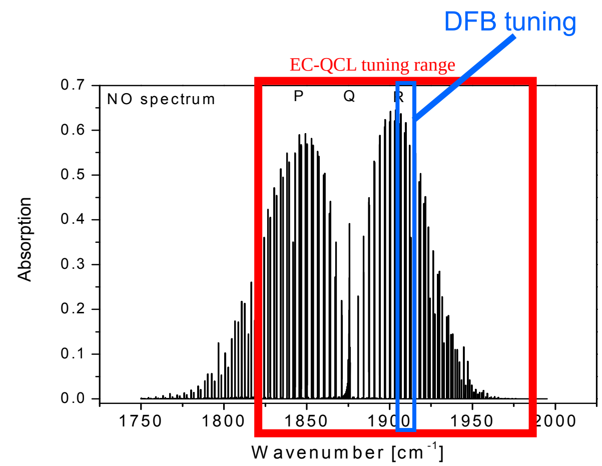
- High resolution mode-hop free wavelength tuning
  - PZT controlled EC-length
  - PZT controlled grating angle
  - QCL current control

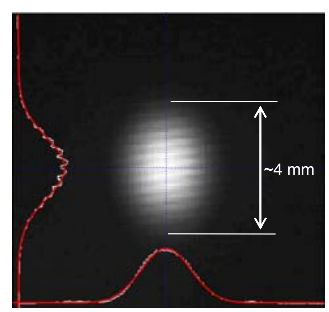
  } ~50Hz
- Motorized coarse grating angle tuning 2-3 sec./scan
- Vacuum tight QCL enclosure with build-in 3D lens positioner + TEC laser cooling + chilled water cooling

Wide Wavelength Tuning of a 5.3μm EC-QCL



- Coarse wavelength tuning of **155 cm^{-1}** is performed by varying diffraction grating angle
- Max. CW power ~**11mW**





In collaboration with:

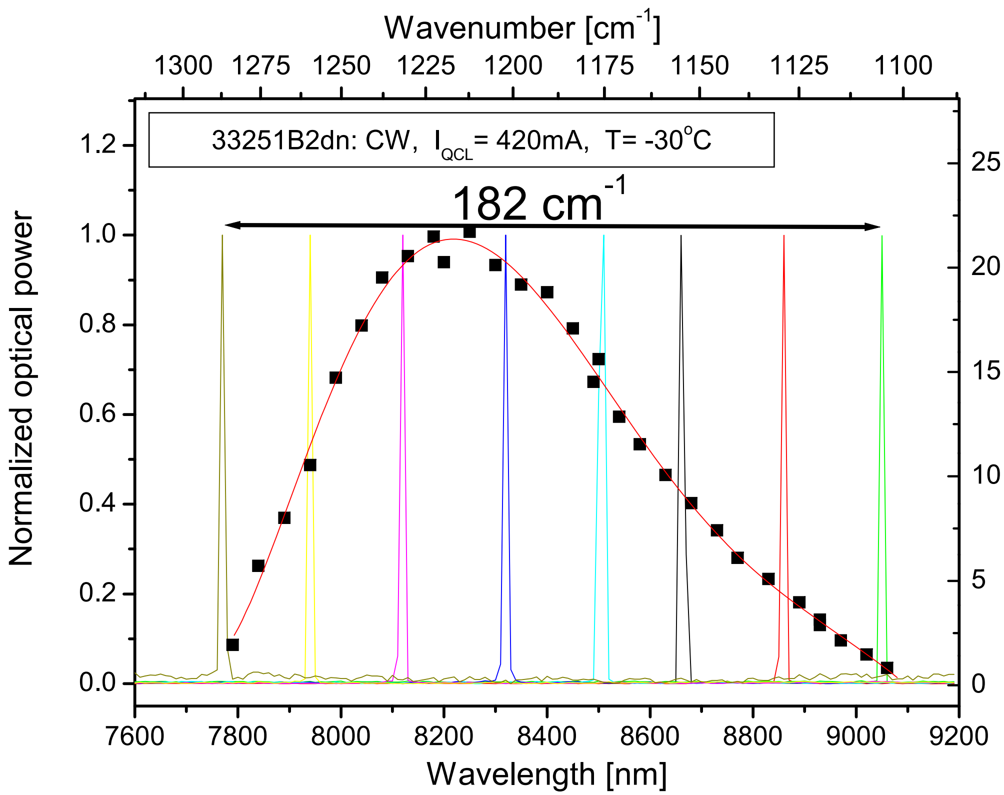
G. Wysocki et al. APB92 p.305 (2008)

EC-QCL emitting at λ = 8.4 μm

$P_{EC\text{-}opt}$ up to **50mW (cw)**

AR coating:

$R_{AR} \approx$ **5 × 10^{-4}**



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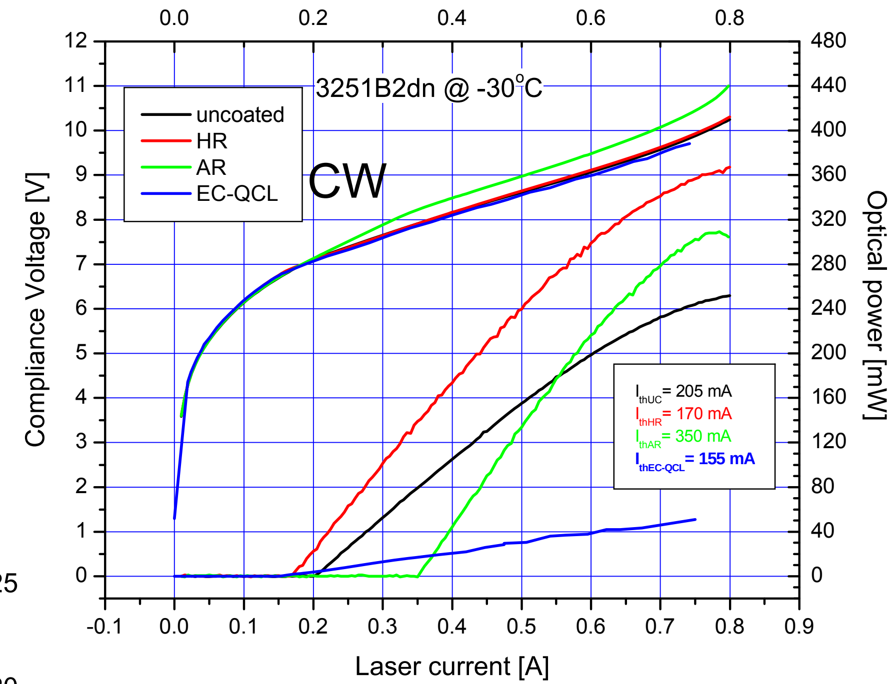


Tunability **182 cm^{-1}**
@8.4 μm (7.77μm - 9.05μm)
15.3 % of the center wavelength

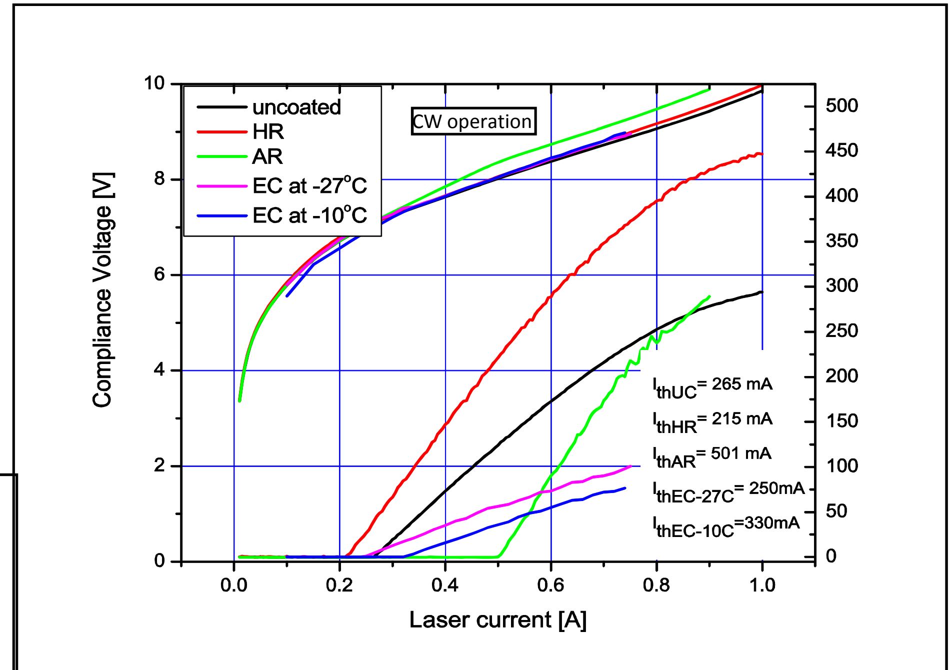
In collaboration with:

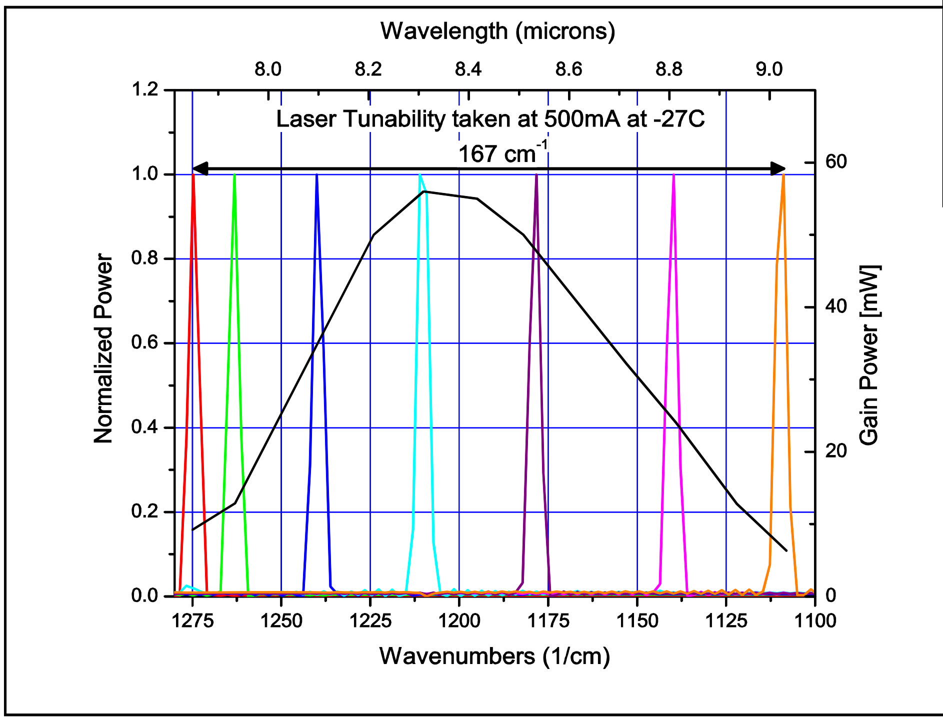
EC-QCL emitting at λ = 8.4 μm

$P_{EC\text{-}opt}$ **>100mW (cw)**

AR coating:

$R_{AR} \approx$ **~10^{-4}**





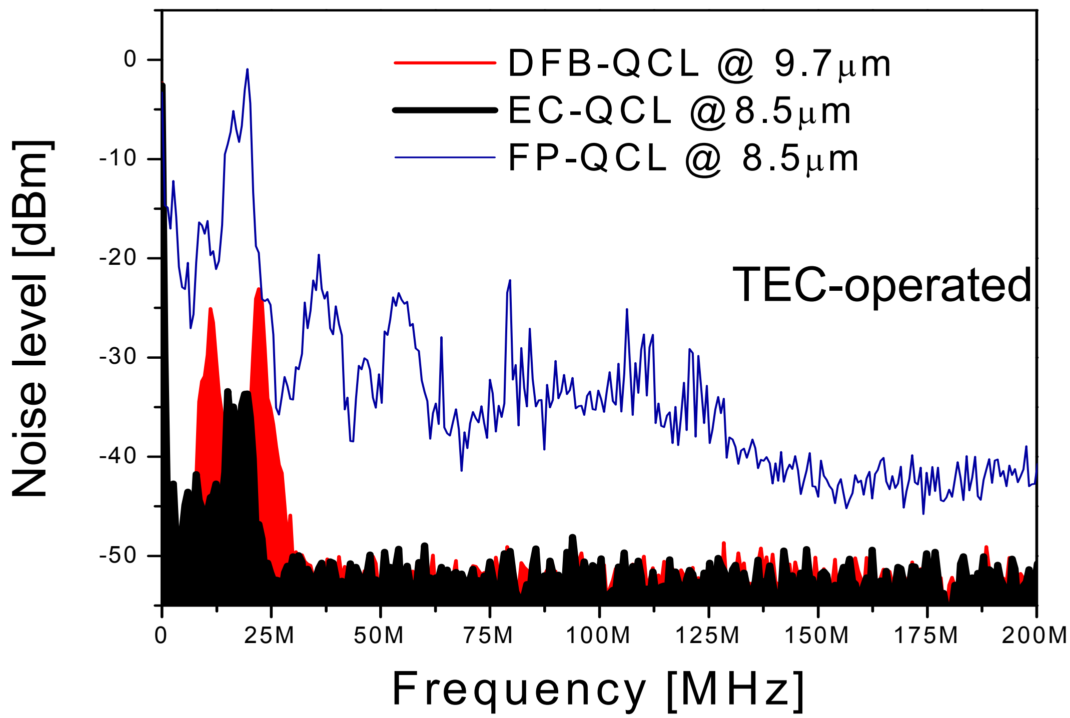
Tunability **167 cm^{-1}**

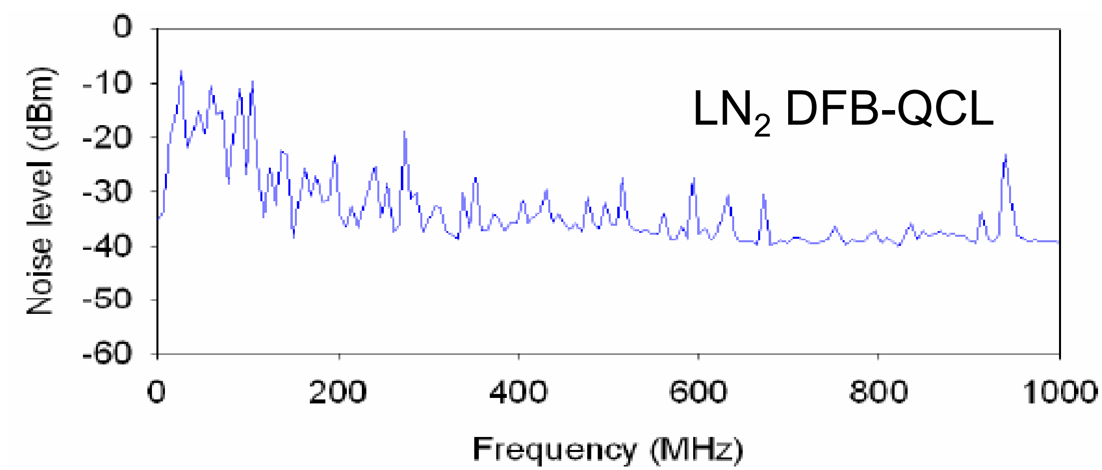
@8.4 μm

14 % of the center wavelength

In collaboration with:

EC-QCL Noise





- EC-QCL outperforms DFB-QCLs in terms of RF noise figure
- EC feedback reduces laser excess noise
  - ~L_{QCL}/L_{EC} reduction of injection current fluctuations impact
  - Strong feedback gives >30dB SMSR
- The excess noise has direct Impact on QCL-based systems performance
- FP "close to threshold" is not the best way to obtain single-mode lasing

In collaboration with Dr. Damien Weidmann
Rutherford Appleton Labs, UK

EC-QCL Pros & Cons

PROS:

- Broad tuning (operation wavelength flexibility)
- High power (as for most spectroscopic applications)
- Good beam quality
- Narrow linewidth
- Lower excess noise

CONS:

- Slow tuning
- Power fluctuations (especially in BB coarse tuning)
- Non-monolitic (requires external optics)

Available Applications of EC-QCLs

Excellent performance in:

- **Applications with critical wavelength restrictions**
- **Applications with pure intensity modulation**
  - Low resolution
    - QCL current (laser linewidth ≈ grating bandwidth)
  - High resolution (available with external AM modulation only)
    - Chopper
    - AOM, EOM (difficult and expensive in mid-IR)
- **Applications with external wavelength modulation**
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- **Applications with external modulation/detection means**
  - "Sample" modulation (Zeeman, Faraday, Stark modulation)
  - Heterodyne spectroscopy
  - CRDS

Poor performance in:

- **Applications with direct wavelength modulation:**
  - Most popular: LAS, WM-LAS, FM-LAS,
  - Photoacoustic detection (WM-PAS, WM-QEPAS)
- **High-res applications with direct AM modulation**

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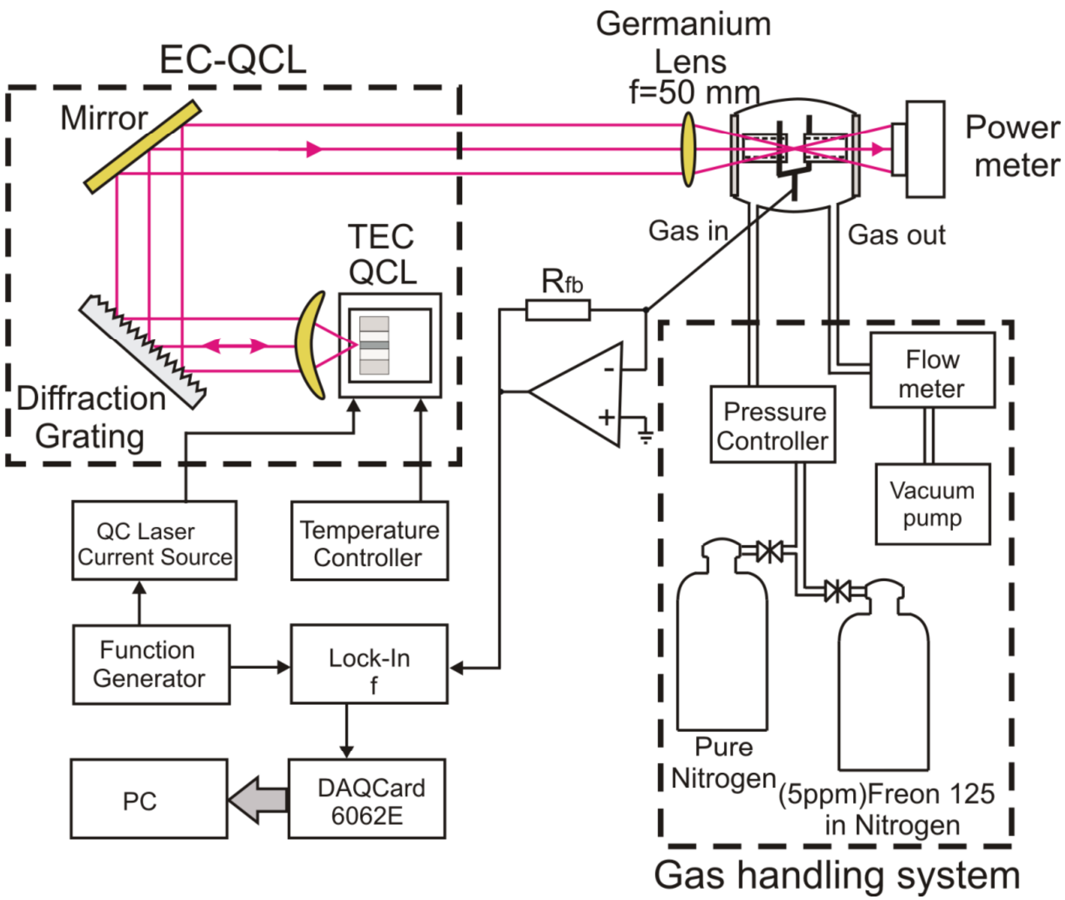
QCL based Quartz-Enhanced Photoacoustic Gas Sensor

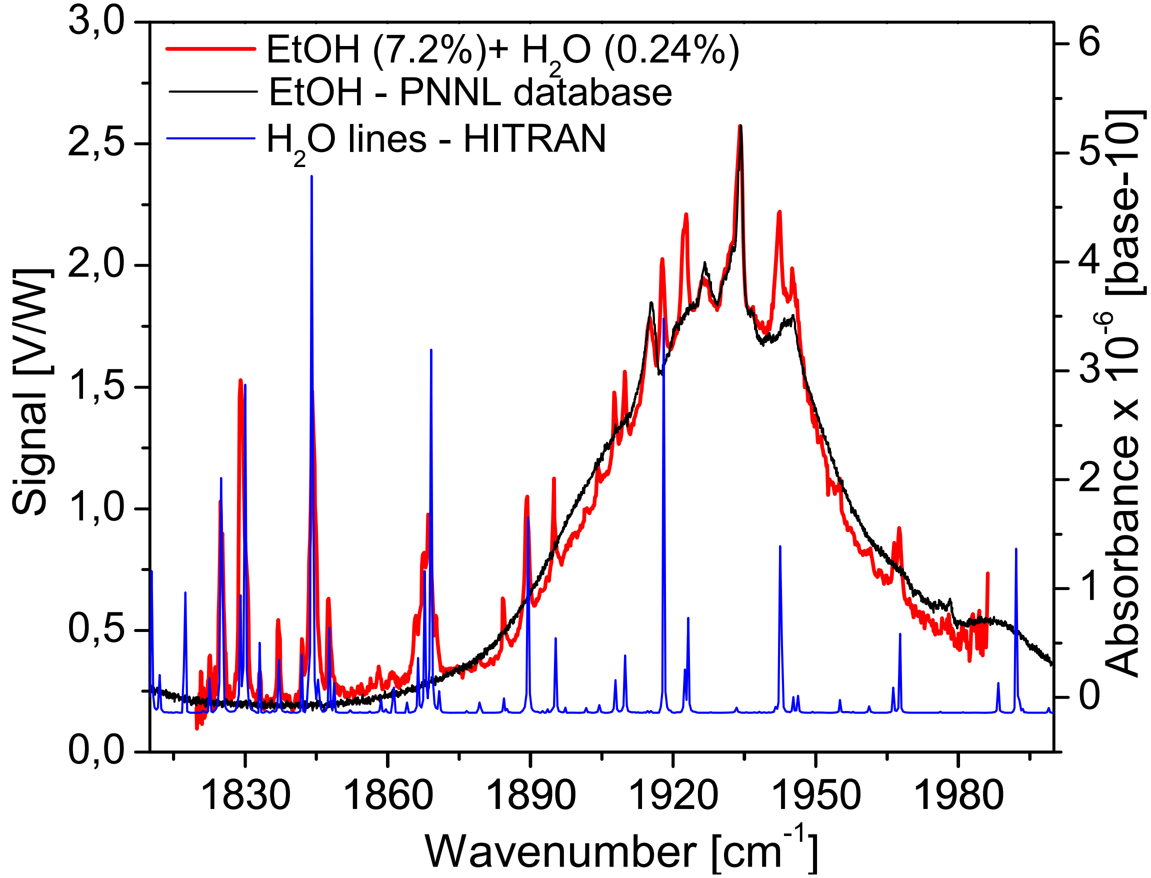


QEPAS characteristics:

- High sensitivity (ppm to ppb)
- Excellent dynamic range
- Immune to environmental noise
- Ultra-small sample volume (< 1 mm^3)
- Sensitivity is limited by the fundamental thermal quartz tuning fork (QTF) noise
- Compact, rugged and low cost
- Potential for trace gas sensor networks

R. Lewicki, G. Wysocki, A.A. Kosterev, and F.K. Tittel, Opt. Express **15**, 7357 (2007)

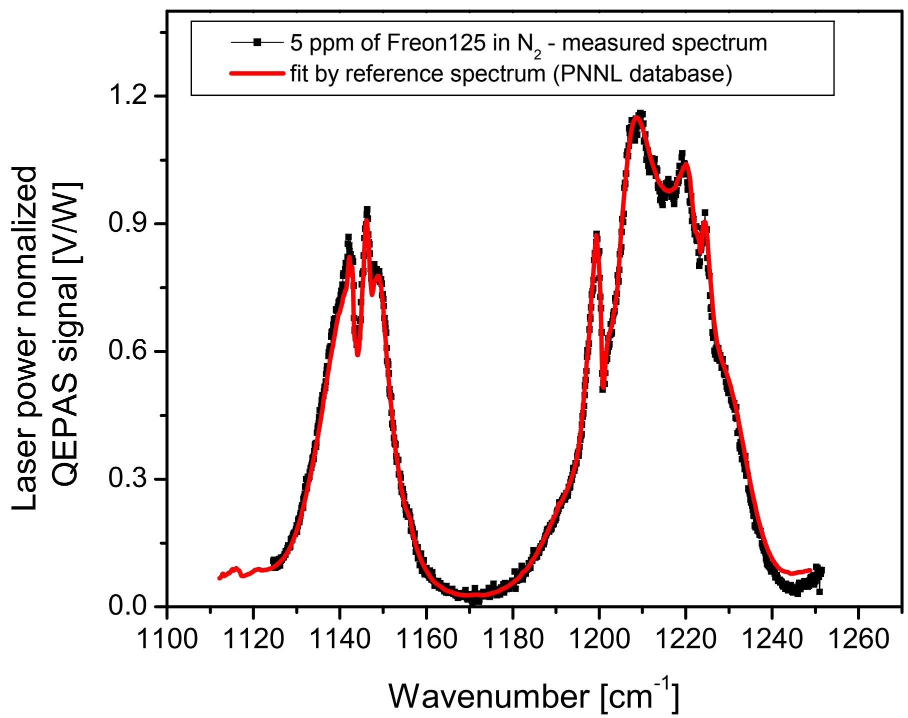
QEPAS ethanol spectrum between 1825 & 1980 cm^{-1}



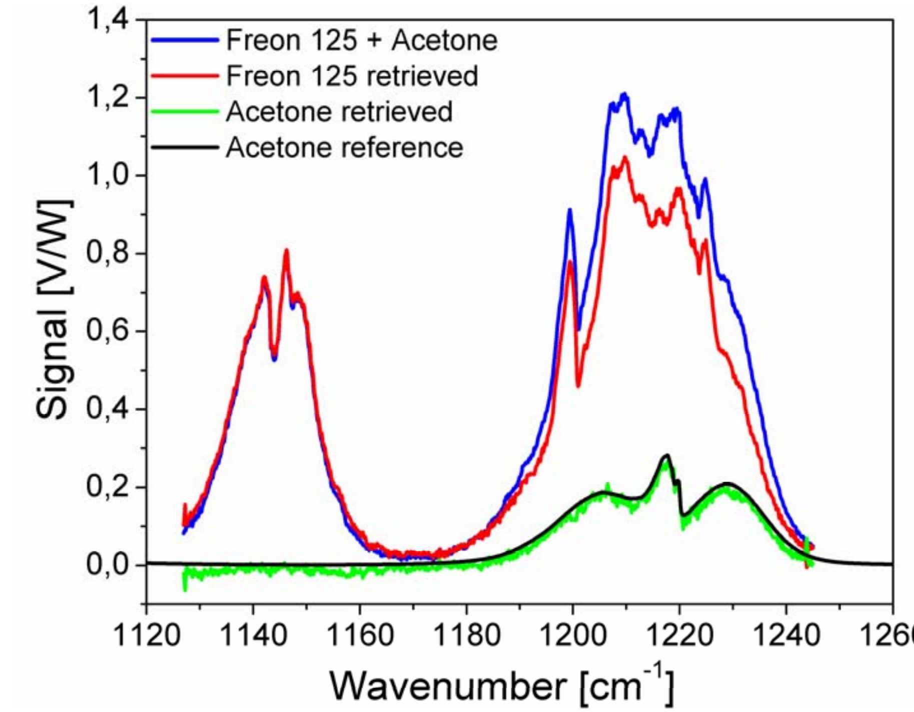
- Reference spectrum from the PNNL spectral database (black line).
- Sharp features on the ethanol spectrum correspond to the water absorption lines.
- Blue line depicts water absorption spectrum simulated using HITRAN database.
- Estimated resolution of a coarse wavelength scan **~1.2cm^{-1}**

Spectroscopy of Broadband Absorbers with Widely Tunable EC-QCL at λ = 8.4 μm

QEPAS concentration measurement of Freon 125 (5ppm mixture in N_2)



QEPAS concentration measurement of a Freon 125 and acetone mixture

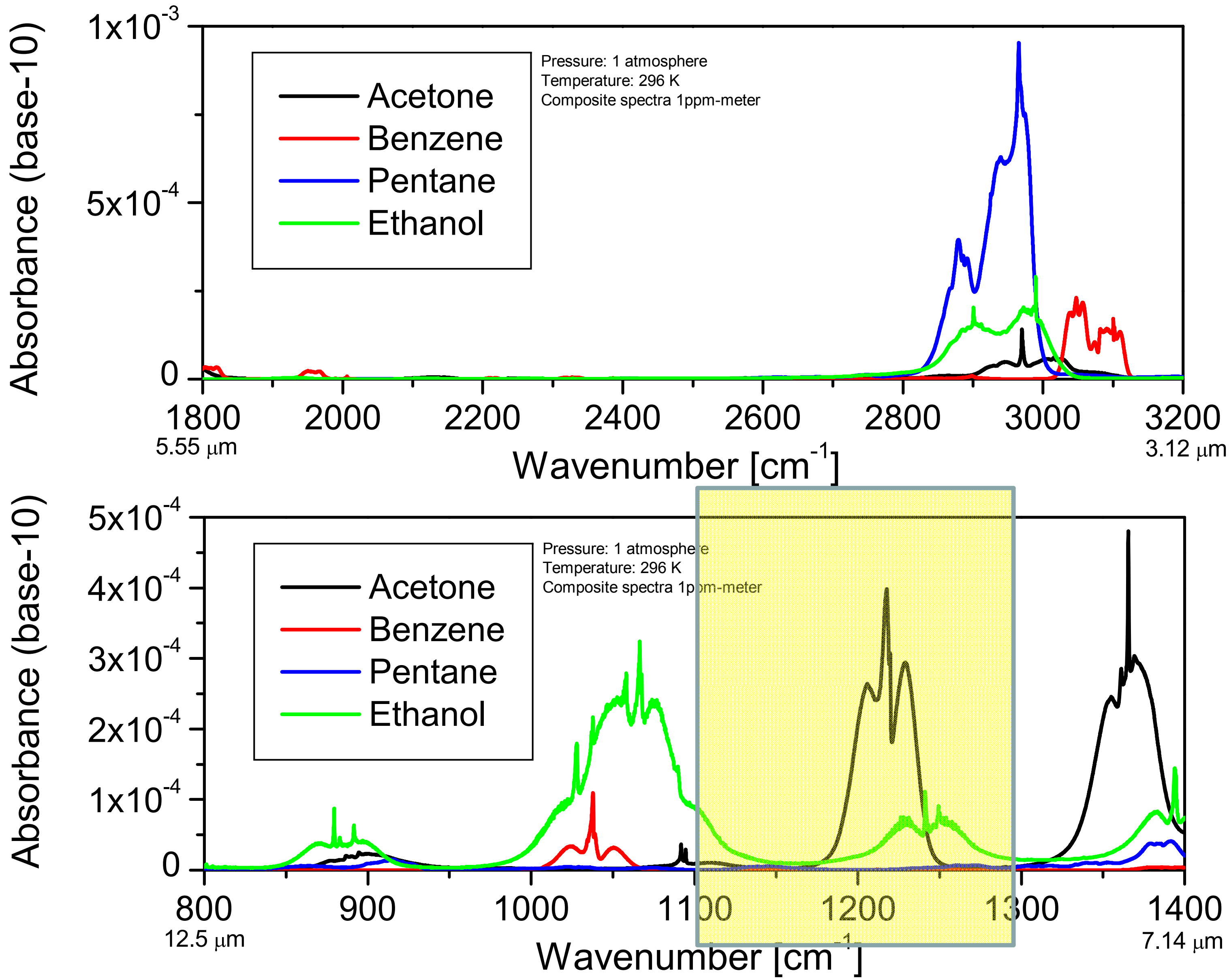


- Minimum detection limit (1σ) of **~3 ppb** was obtained for Freon 125 with an average laser power of 6.6 mW

- Wide tunability enables excellent molecular selectivity for broad band absorbers

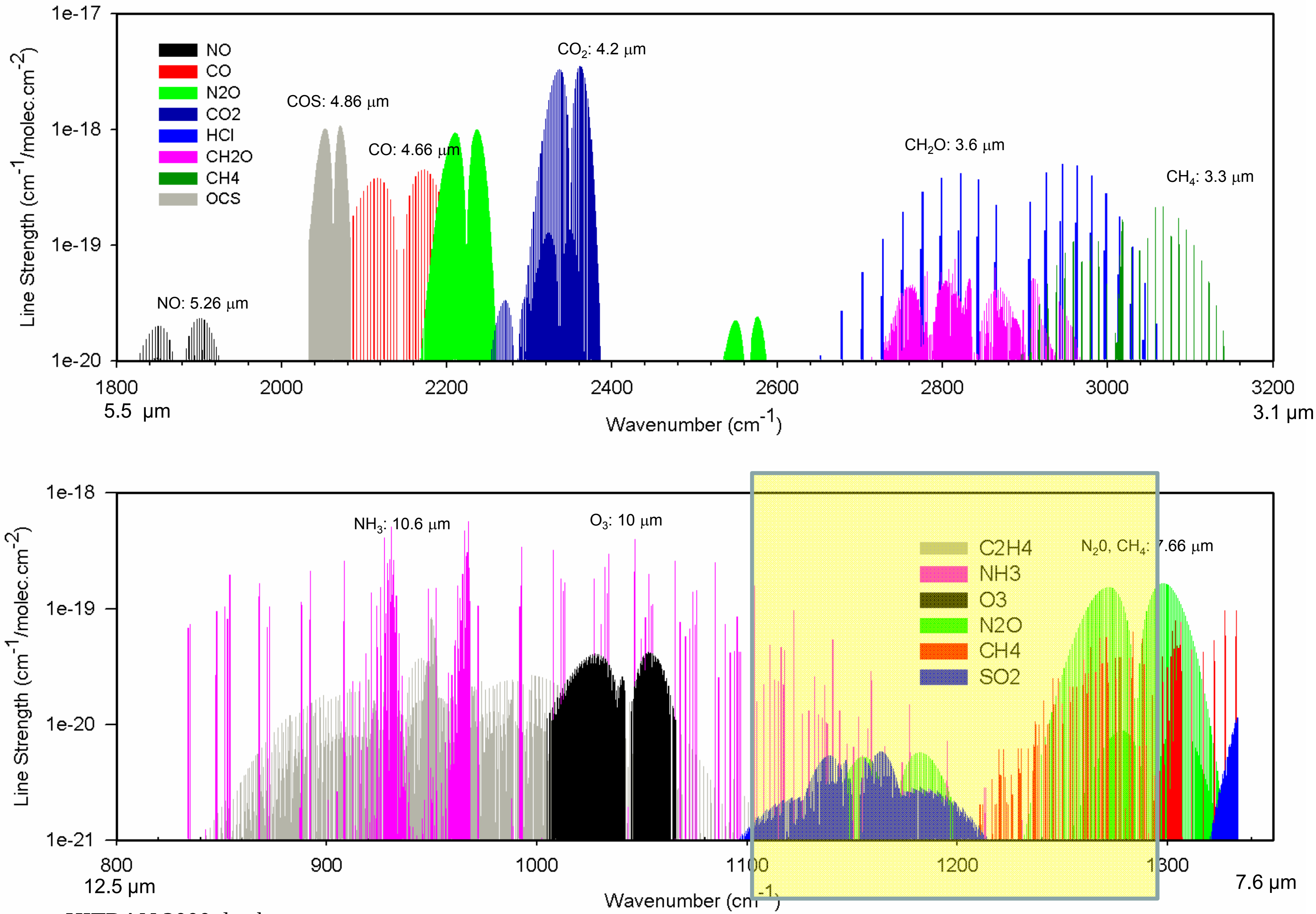
R. Lewicki, G. Wysocki, A.A. Kosterev, and F.K. Tittel, Opt. Express **15**, 7357 (2007)

Example Absorption Spectra of Broadband Absorbing Molecules



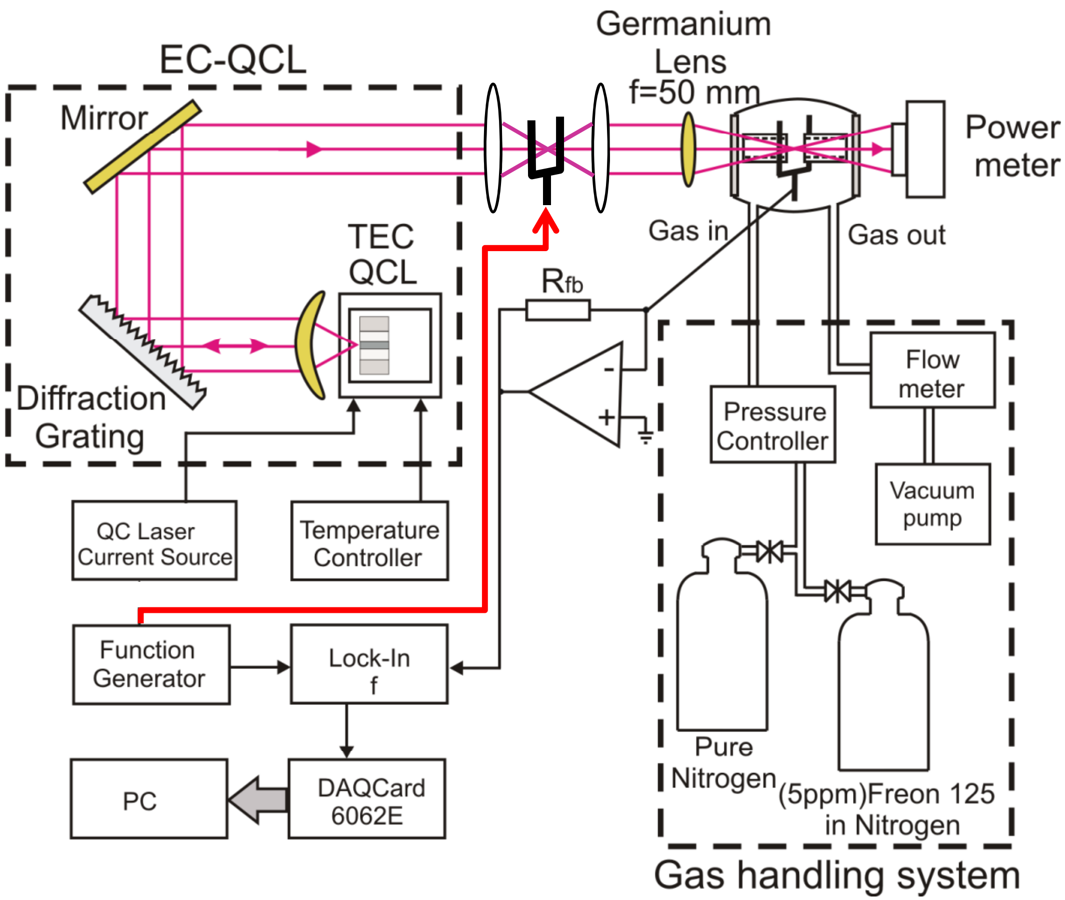
Source: PNNL spectroscopic database

Example Molecular Absorption Spectra within Mid-IR "Atmospheric Windows"



Source: HITRAN 2000 database

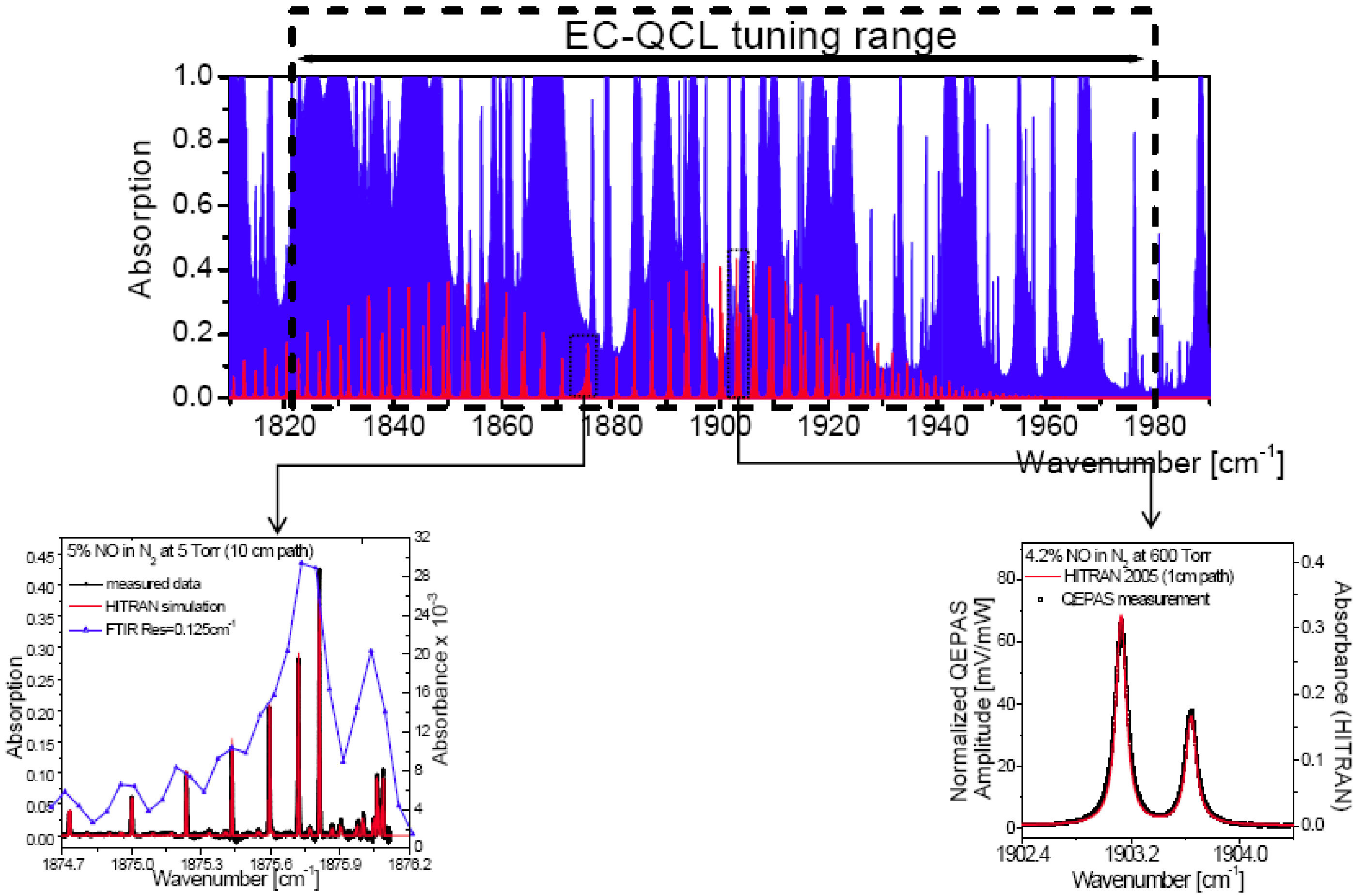
High resolution EC-QCL based QEPAS



External AM:

- QTF used as a mechanical chopper at *f=~32kHz*
- No chirp associated with the laser current modulation
- High resolution mode-hop-free tuning is possible

High resolution spectroscopy with a 5.3μm EC-QCL



- Mode hop free scan of up to ~**2.5 cm^{-1}** with a resolution **<0.001cm^{-1}** (30MHz) can be performed anywhere within the tuning range

G. Wysocki et al. APB92 p.305 (2008)

In collaboration with:

unine

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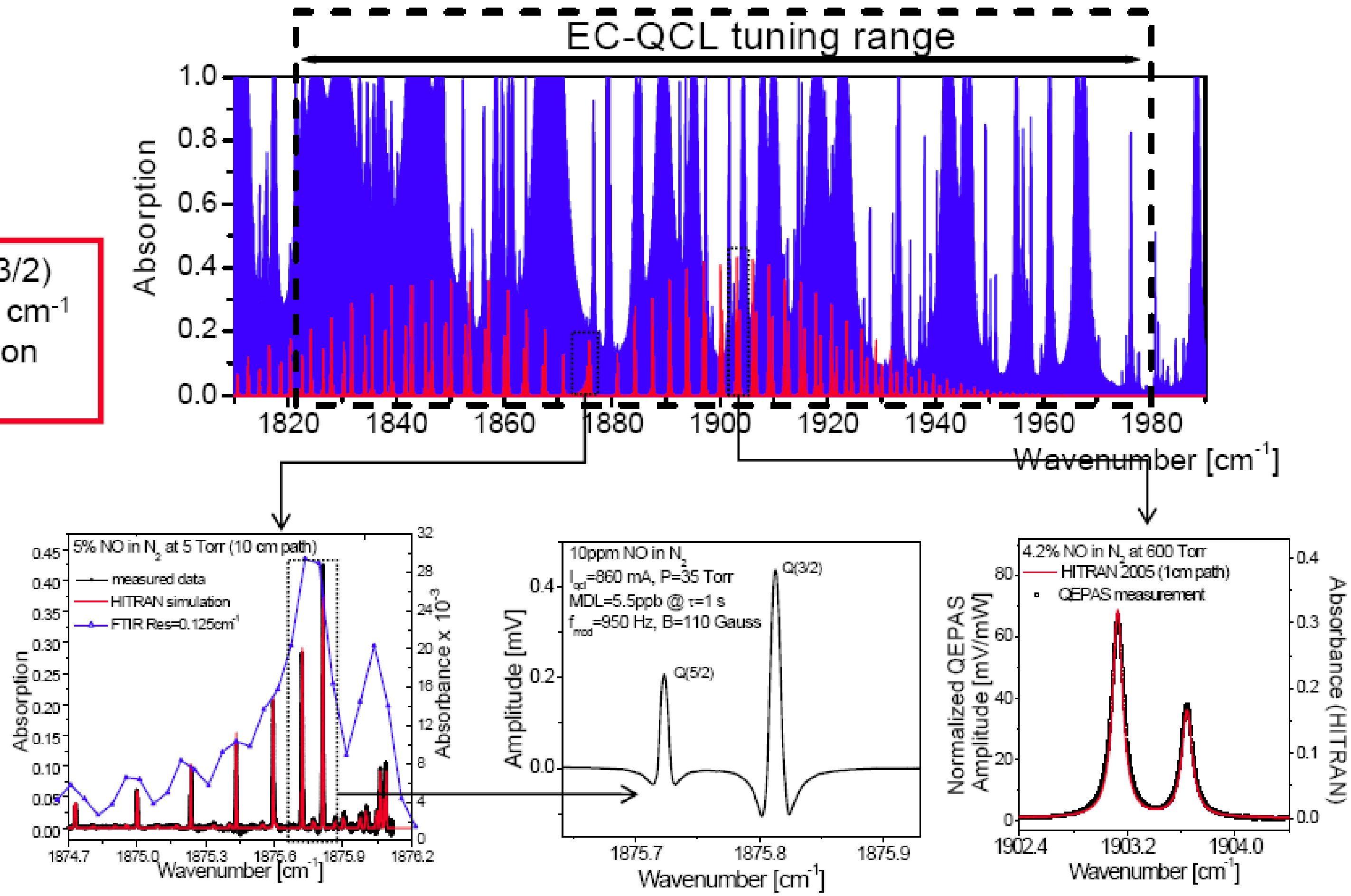
Access to NO Q(3/2) transition at 1875.8 cm^{-1} for Faraday rotation spectroscopy



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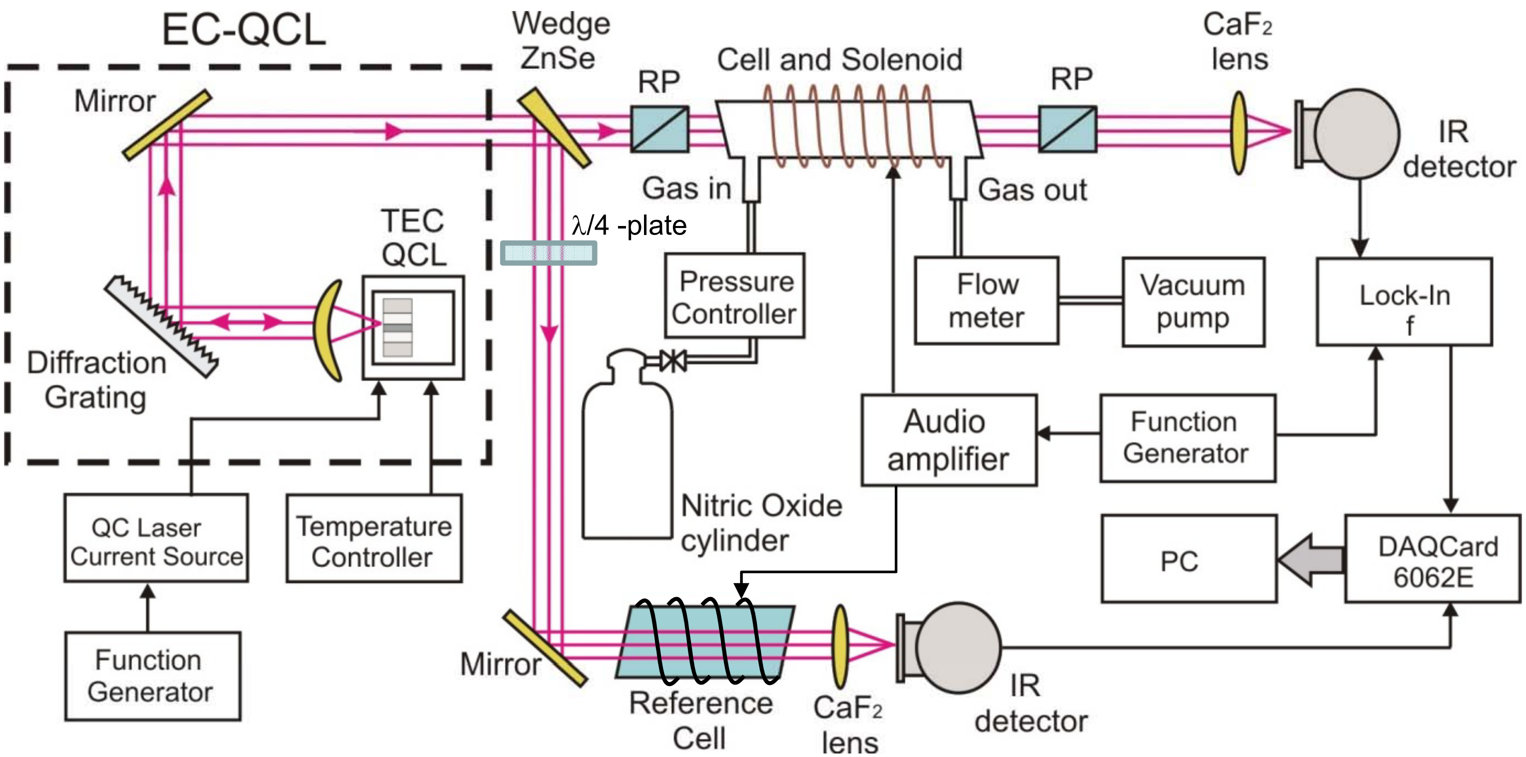
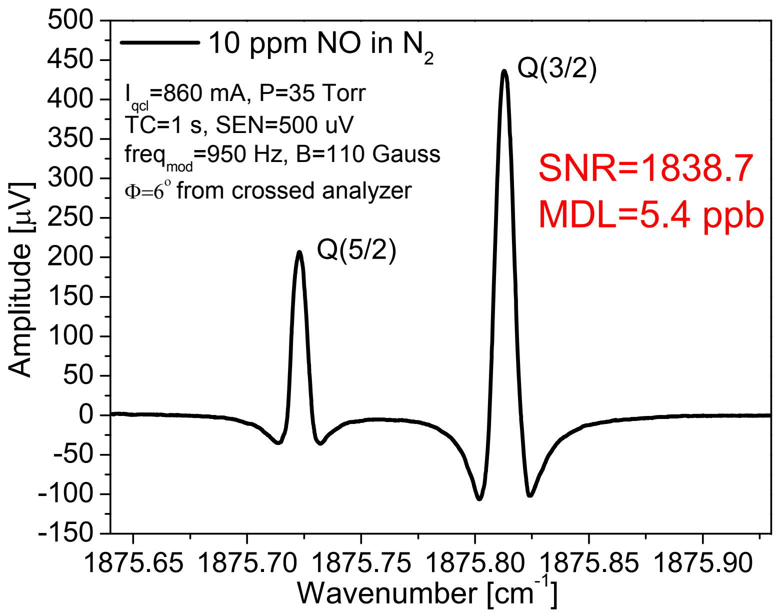
EC-QCL Based Faraday Rotation Spectrometer



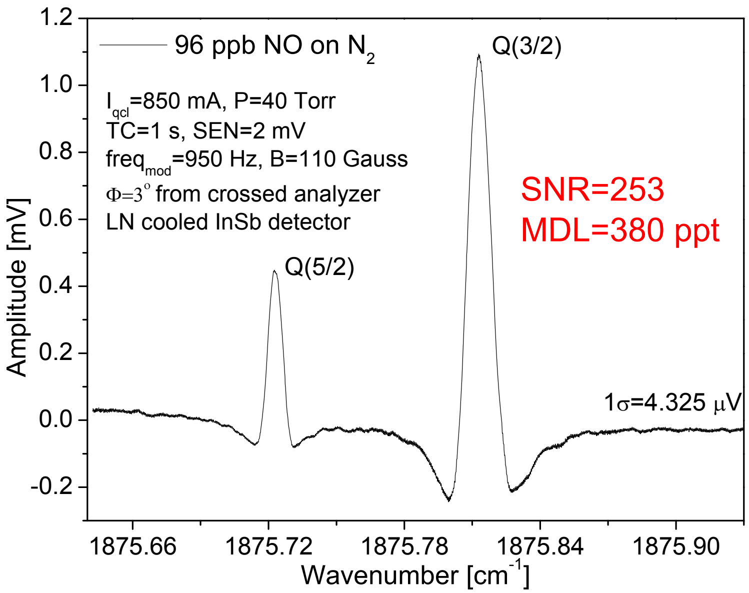
- EC-QCL Operating at 5.3μm – NO Fundamental Band
- 44cm effective optical pathlength
- Rochon Polarizer Extinction Ratio $<10^{-5}$
- Equivalent minimum detectable absorption coefficient → ~**1.4×10^{-9}** cm^{-1}
- Not sensitive to water interference
- Sensitivity Not Limited by Interference Fringes
- Gas Cell Volume (~ 250ml)
- Easy and Robust Optical Alignment
- Continuous NO Monitoring (Absorption Line Locking enabled with mode-hop free tuning using Zeeman Modulation at 3rd harmonic)

High Resolution Faraday Spectroscopy of NO at λ = 5.33μm

TEC-cooled MCT Detector → D*≈10^{10}



LN$_2$-cooled InSb Detector → D*≈10^{11}



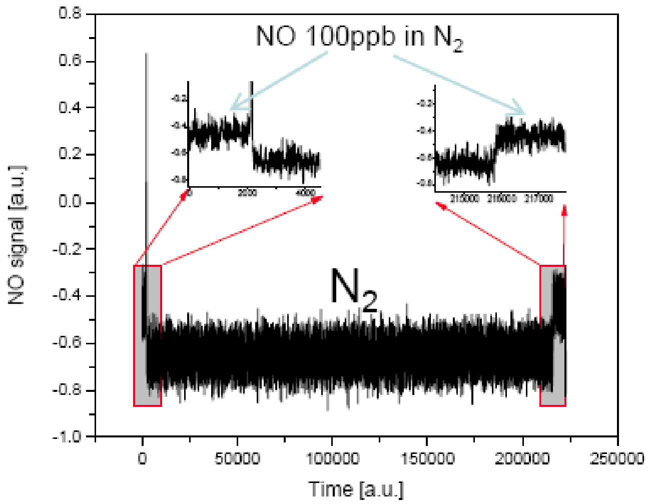
- Minimum detection limit (1σ) of **~5.4 ppb**
- MDL primarily limited by the detector noise

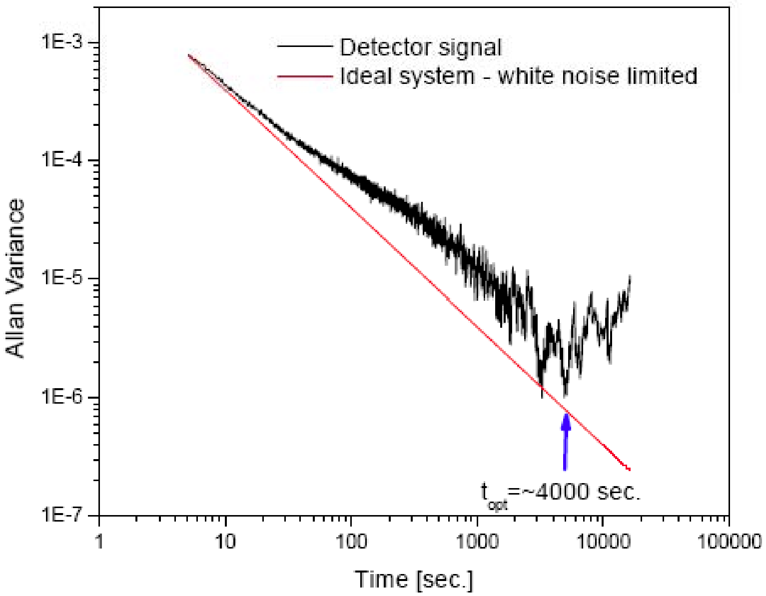
- Minimum detection limit (1σ) of **~ 0.38 ppb**
- MDL primarily limited by the quality of polarizers

- **<u>Excellent dynamic range</u>**

Continuous Monitoring of NO

- TEC-cooled detector allows **continuous unattended operation** of the sensor at the expense of lower MDL
- Preliminary results show excellent system stability (>1000 sec.)
- Suppression of an electromagnetic noise will further improve MDL





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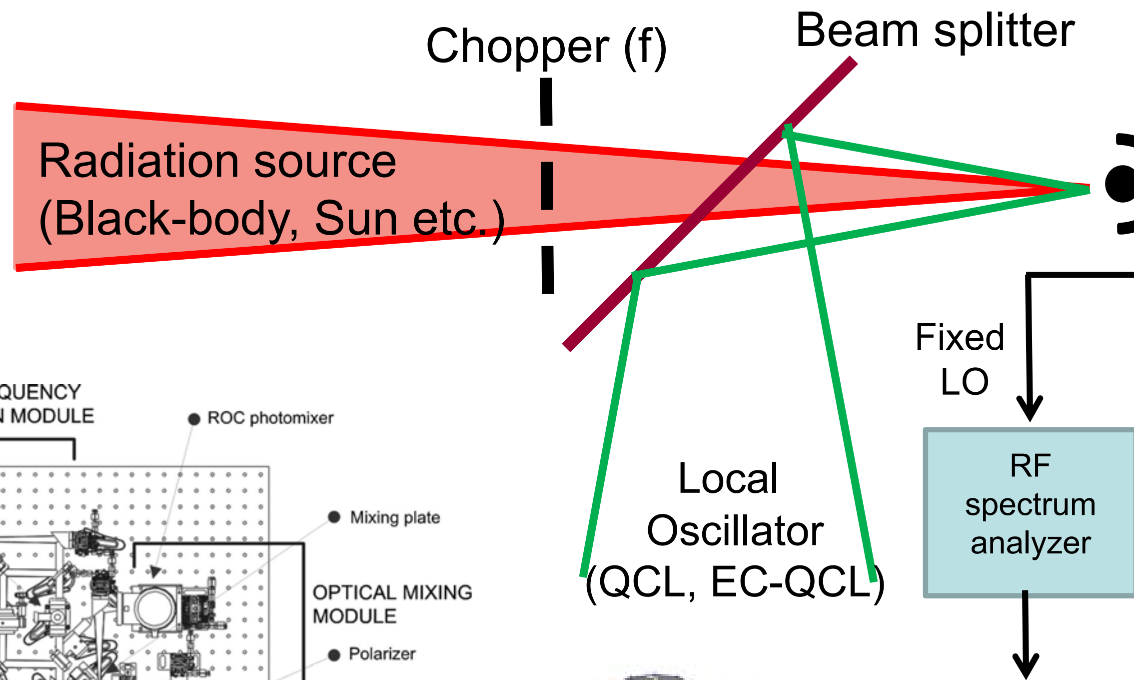
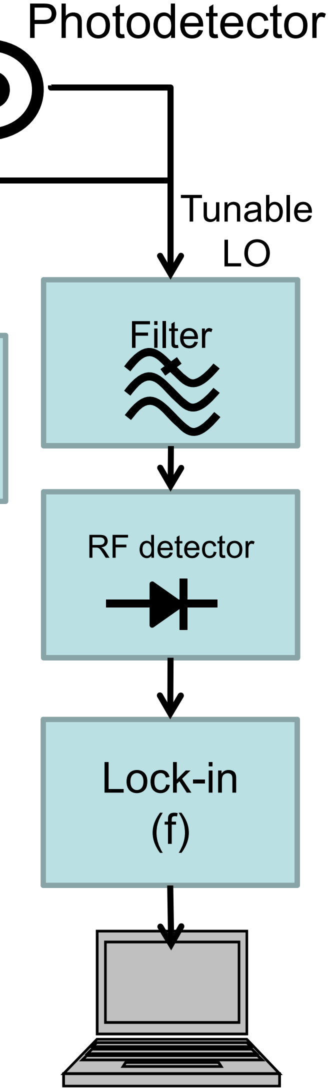
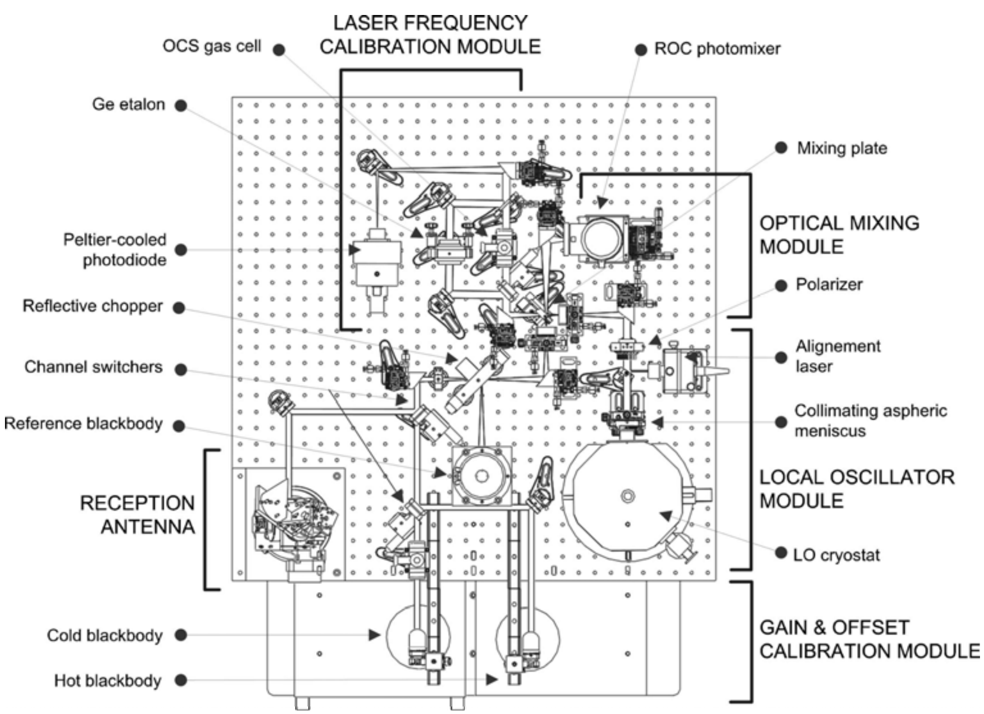
EC-QCL Based Heterodyne Radiometry







<u>In collaboration with Dr. Damien Weidmann @ Rutherford Appleton Labs in UK</u>

EC-QCL Based Heterodyne Radiometry

- Sensitivity → shot noise limited

$$NEP = \frac{h\nu}{\eta}\sqrt{\frac{B}{\tau}}$$

$\lambda = 10\ \mu m$
$B = 100\ MHz \Rightarrow$ **$NEP = 4.10^{-16} W$**
$\tau = 1 s$

- Tunable LO / Fixed LO + snapshot of RF spectrum

- Spectral resolution/Instrumental linewidth → electronic filters

$$\frac{\Delta\lambda}{\lambda} < 10^{-6}$$

- Passive remote sensing → Absorption/Emission

- Spatial resolution → Coherent FoV

$$FoV = \frac{4\lambda}{\pi D}$$

$\lambda = 10\ \mu m$
$D = 10\ cm \Rightarrow$ **$FoV = 0.13\ mrad$**

- Polarization sensitive detection

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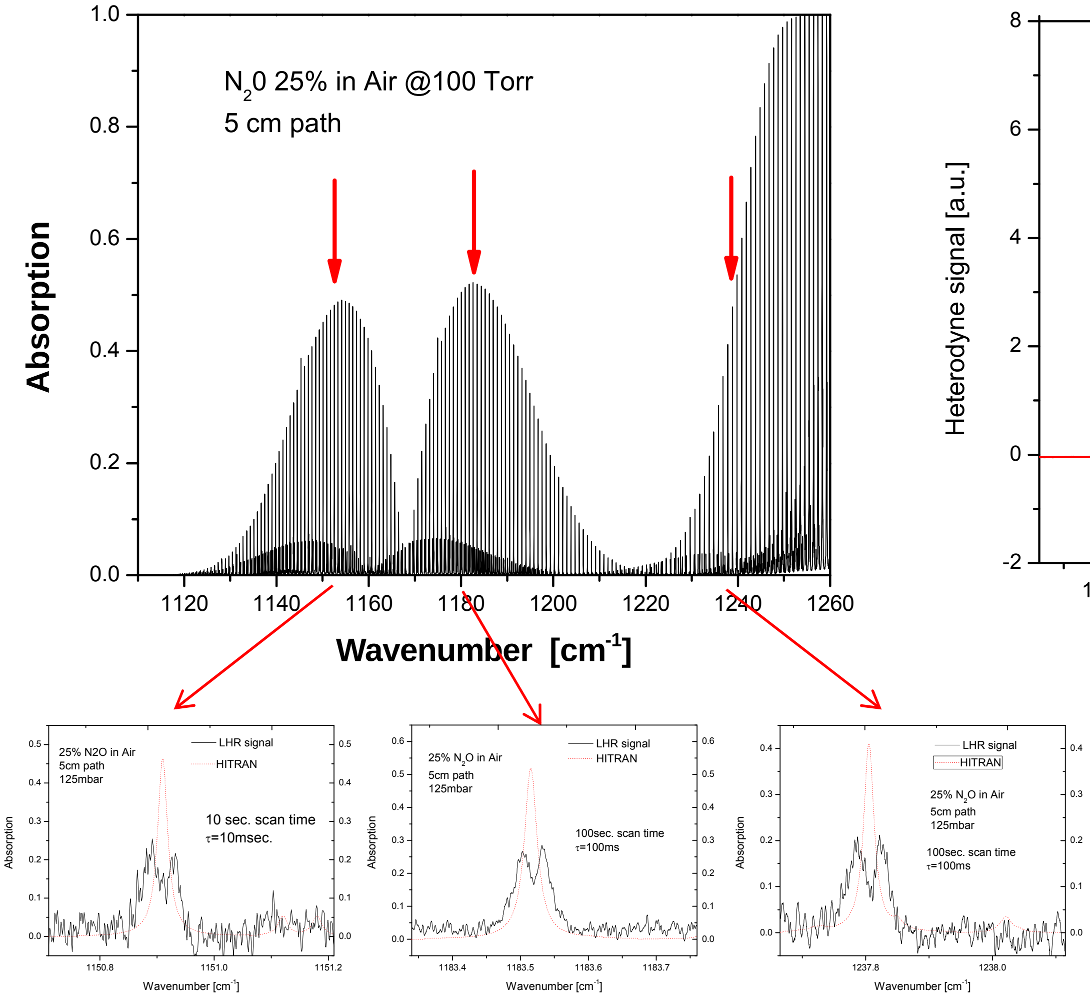
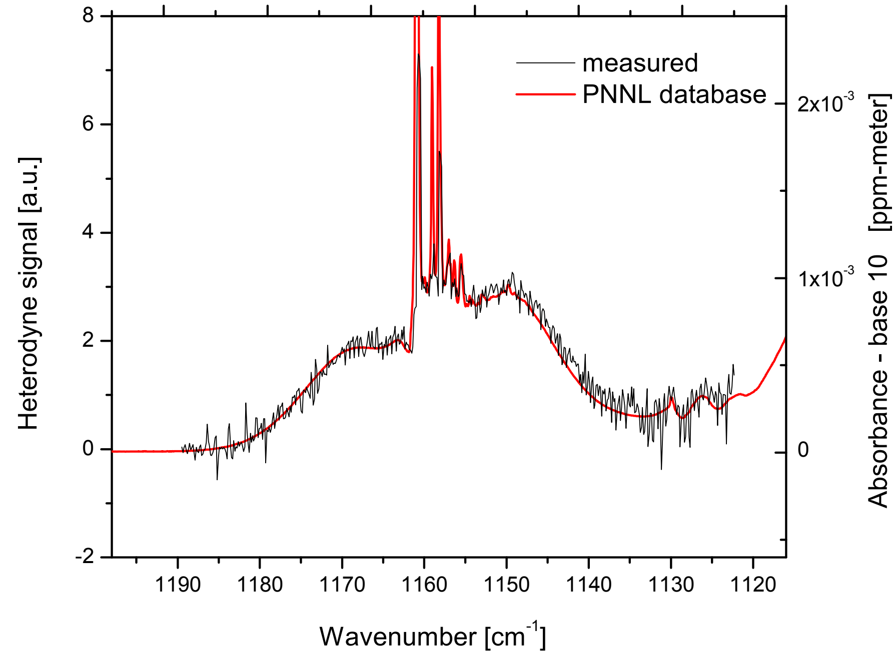
EC-QCL Based Heterodyne Radiometry

High resolution LHR of N_2O



Broad-band LHR of Freon 12



<u>Enabled applications:</u>

- Atmospheric trace-gas profiling.
- Remote multispecies analysis

<u>In collaboration with Dr. Damien Weidmann @ Rutherford Appleton Labs in UK</u>

How to improve the EC-QCL tuning speed?

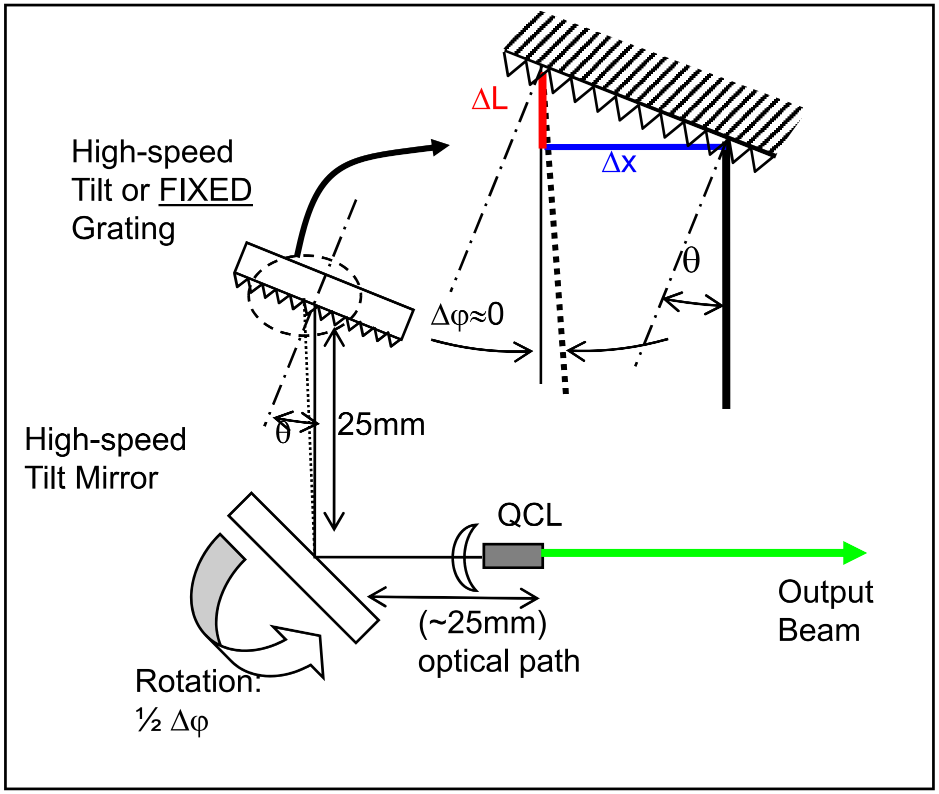
- Avoid low frequency **mechanical vibration resonances**
- Use **small displacements and low inertia** scanning elements (or EOM AOM beam steering → next step)
- Beam steering at **KHz rates** is possible with a **compact piezo-actuated tilt platform** (mirror steering)

New approach to achieve KHz scanning rates:

- Replace the slow translation stages with high speed tilt-platforms
- Folded laser cavity arrangement
- **Mode-hop-free tuning**

Folded ECDL tuning concept (configuration #1 and #2)

Change in the optical path;
$\Delta L = \Delta x \tan(\theta) = L \tan(\Delta\varphi) \tan(\theta)$



Configuration #1 "*Dual actuator single-folded Littrow cavity arrangement* ":
- The grating is mounted on similar tilt platform for precise (independent) grating angle tuning

Advantages: fully electronically-controlled wavelength tuning
Limitations: requires precise placement of the diffraction grating on the tilt actuator

Configuration #2 "*Single actuator single-folded Littrow cavity arrangement* ":
- The grating is fixed

Advantages: requires only one actuator, offers robust system with simplified system adjustment
Limitations: requires careful selection of the diffraction grating (blaze angles >45deg.)

The output laser beam has to be extracted from a second laser facet

ECDL tuning concept (configuration #2)

Change in the optical path;
$\Delta L = \Delta x \tan(\theta) = L_1 \tan(\Delta\varphi) \tan(\theta)$

$L_{tot} = L_1 + L_2$

$$\theta = a\tan\left(\sqrt{\frac{L_{tot}}{L_1}}\right)$$

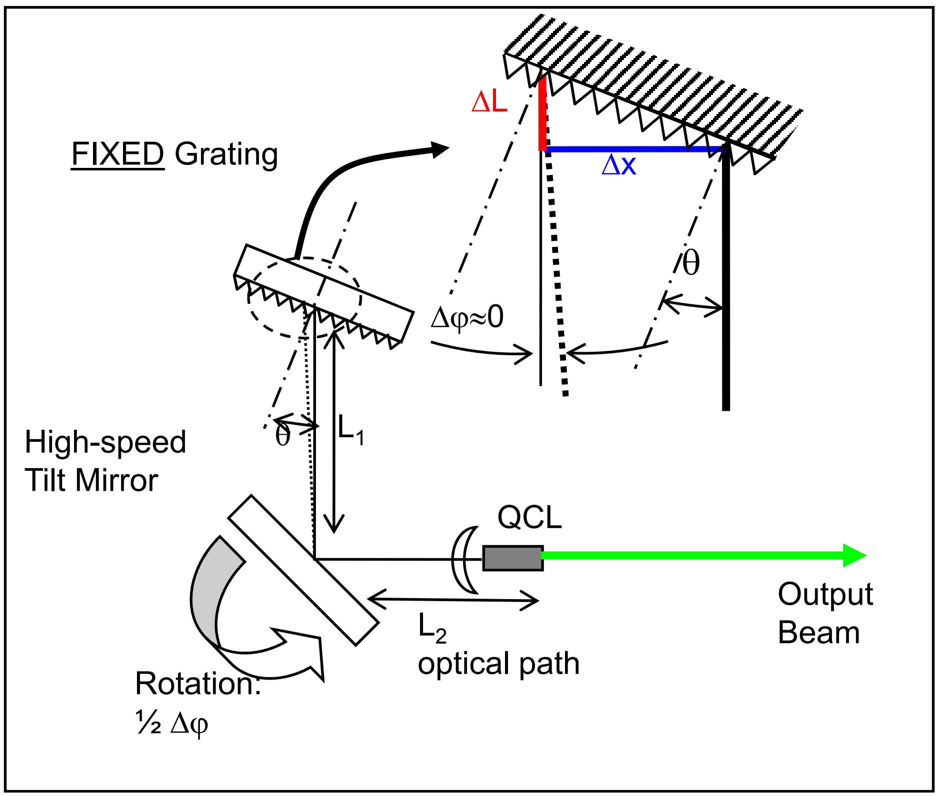
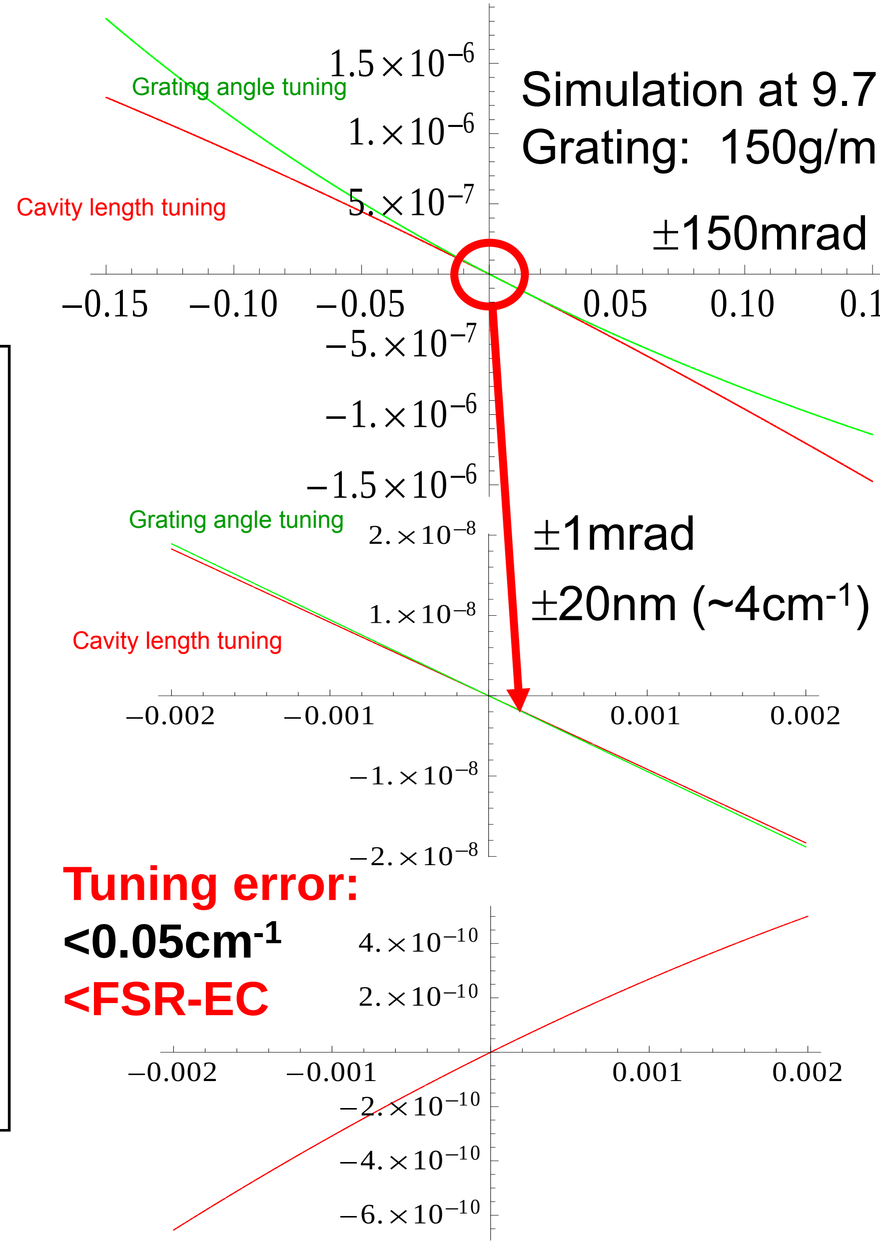


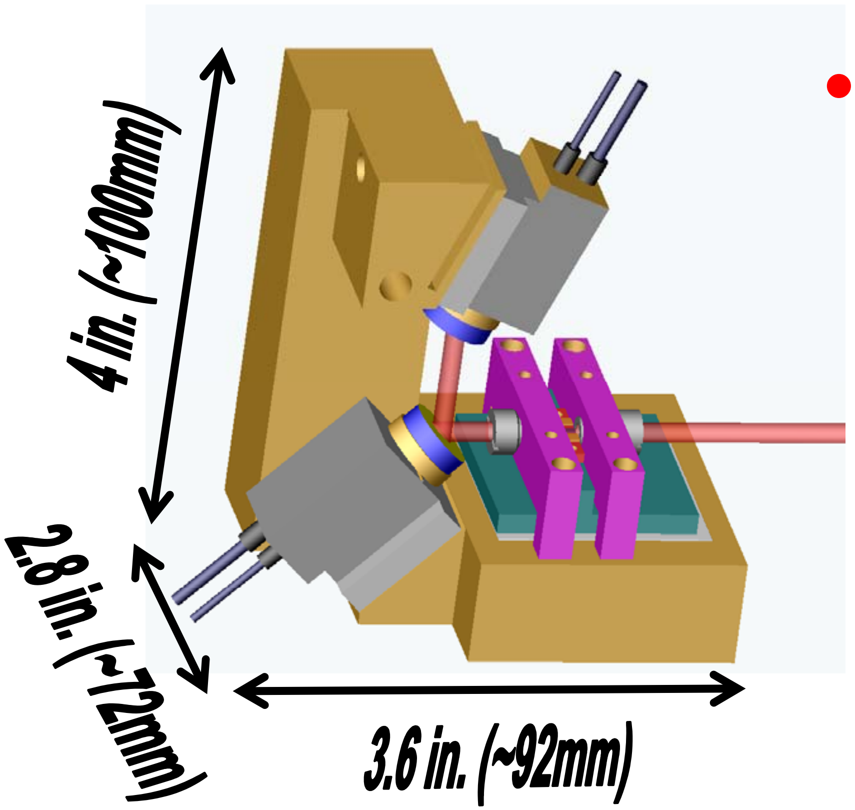
Figure : Schematic of the proposed new EC-QCL folded cavity tuning scheme

Simulation at 9.7μm
Grating: 150g/mm



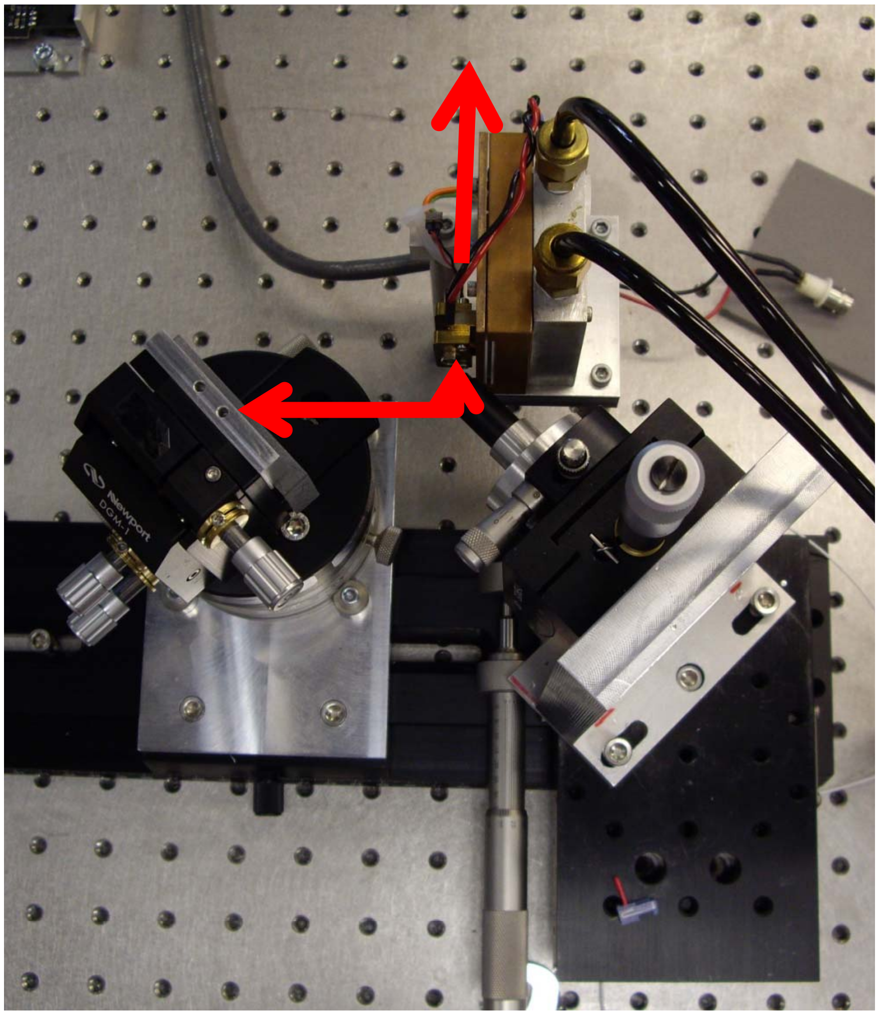
Tuning error:
<0.05cm⁻¹
<FSR-EC

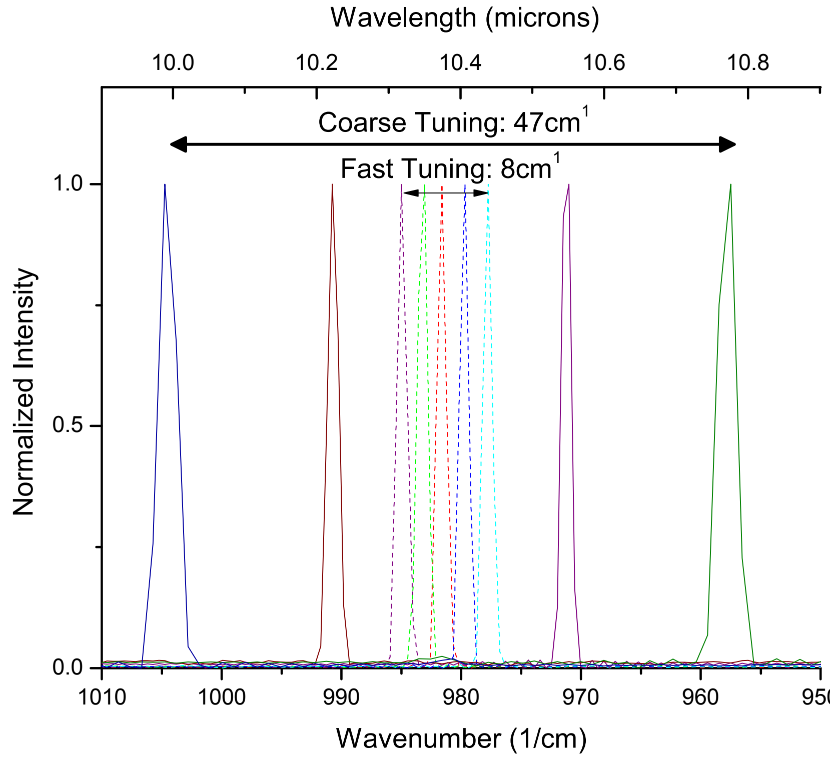
New design of fast broadly tunable EC-QCLs



- New optical configuration
  Folded cavity (configuration #1)
- Fast tuning capabilities:
  - Coarse Broadband Scanning (~*55 cm^{-1}* @5µm) ***up to 5 KHz***
    *(compared to available technologies **<10Hz**)*
  - High resolution mode-hop free tuning (~*3.2 cm^{-1}* @5µm) ***up to 5 KHz***
    *(compared to available technologies **100-200 Hz**)*

Fast broadly tunable EC-QCL @9.7μm under test





To-date:

- Manual coarse tuning 47cm^{-1}
- Fast coarse electrical tuning within 8cm^{-1} at KHz rates (PZT resonance at 6.5KHz)

NEXT STEPS:

- Mode-hop-free tuning
- Spectroscopic applications

Summary & Future Directions

- **Widely tunable, continuous wave and thermoelectrically cooled EC-QCLs** were demonstrated with:
    - **15% tunability,**
    - **>100 mW output power**
    - **high resolution (<0.001cm^{-1}) mode-hop-free tuning capabilities**

- Number of high sensitivity spectroscopic sensing applications are enabled by the EC-QCL technology. Examples given:
    - **Photoaccoustic detection**
    - **Faraday Modulation Spectroscopy**
    - **Heterodyne detection**
- First tests of a high-speed EC-QCL system were demonstrated with KHz scanning capabilities

Future directions:

- Fast mode-hop-free EC-QCL
- new applications in laser based trace gas sensing
    - Sensitive concentration measurements of broadband absorbers, in particular VOCs and HCs
    - Multi-species detection

Acknowledgements

- **Frank K. Tittel**
- **Robert F. Curl**
- **Damien Weidmann**

QC-lasers:

- **Jerome Faist**
- **Federico Capasso**
- **Claire Gmachl**

DoE STTR: Aerodyne Inc.

QCL/lens assembly: Daylight Solutions Inc.