Current Status of Quantum Cascade Lasers for UFLS at 7.7, 11.8 and 16 μm

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- Motivation: Laser Absorption Spectroscopy of Uranium Hexafluoride (UF₆)
- High performance QC Laser Source
- Widely tunable External Cavity (EC) -QCLs
  - Daylight Solutions, Technical University Zuerich, Harvard
  - Rice University
- Future Directions and Conclusions
UF$_6$ Mid-Infrared Absorption Bands

Absorption spectrum of gas mixture under investigation and observed spectral features identification.


A. Nadezhdinskii et al, GPI, Moscow, March 2008
Needs and Methods in IR Laser Monitoring

**Requirements:** Sensitivity, specificity, multi-gas species, rapid data acquisition, cost, portability, low electrical power consumption and autonomous operation

**Optimum Molecular Absorbing Transition**
- Overtone or Combination Bands (NIR)
- Fundamental Absorption Bands (MID-IR)

**Long Optical Pathlengths**
- Multipass Absorption Cell
- Cavity Enhanced, Cavity Ringdown & Intracavity Spectroscopy
- Open Path Monitoring (with and without retro-reflector)
- Evanescent Field Monitoring (fibers & waveguides)

**Photoacoustic Spectroscopy**

**Spectroscopic Detection Schemes**
- Frequency or Wavelength Modulation
- Balanced Detection
- Zero-air Subtraction
Quantum Cascade Lasers

Advantages of QCLs:
- Traditional material systems (e.g., InGaAs/AlInAs/InP)
- Gain position by design
- Multi-wavelength and broadband emission possible
- Hundreds of milliwatts of CW output in mid-IR at RT
- Work in mid-IR and THz spectral range
Mid-IR QCLs: present status

"2-phonon" design or "b2c" design

Thermal management
\textit{InP re-growth and/or electroplating}

RT operation $\sim$3-12 $\mu$m

Wallplug efficiency up to $\sim$10%

Hundreds of mW CW output at RT ($\lambda\approx 3.3 \, \mu m$)

\textit{InGaAs/AlInAs system}

\textit{InAs/AlSb system}

Devenson et al., APL 2007
Motivation for High Wall Plug Efficiency

\[ \eta_{wp} = \frac{P_{\text{optical}}}{P_{\text{electrical}}} \]

- High WPE essential for applications limited by:
  - Electrical power
  - Temperature
  - Size
  - Weight

- CW, RT QC laser state of the art:
  - 2006: \(~1\%\)
  - 2008: \(~12\%\)
Quantum cascade laser

- High power
- High temperature operation
- Multi-wavelength emission
- High wall-plug efficiency

Optimization through design

Optimization through processing and packaging

HR coating
AR coating
Strategies for Improving WPE

**Voltage Efficiency:**
- Lower voltage defect
- Reduce contact resistance

**Extraction Efficiency:**
- Implement high reflection (HR) and anti-reflection (AR) coatings
- Employ low loss waveguides
- Optimize cavity length

**Internal Efficiency:**
- Reduce thermionic emission
- Improve heat removal from active core
- Optimize injector barriers

**Current Efficiency:**
- Design high gain, low loss structures
- Lower threshold currents

*Optimization of heat dissipation*  
*Epi-down mounting*
Quantum Cascade Gain medium

- Efficient injection in upper state
- Efficient extraction from lower state
  \[ \Rightarrow \text{Suitable for CW operation above RT} \]

- Transition from bound state to miniband
- Several lower states
- Distributed oscillator strengths
  \[ \Rightarrow \text{Broad and quasi-homogeneous gain medium} \]

\[ \Delta \nu = 274 \text{ cm}^{-1} \text{ (FWHM)} \]
\[ \Delta \nu/\nu_0 = 21 \% \]

Lubos Hvozdara et al, IPS Greifswald July 23, 2008
Faist et al., APL 78, 147 (2001)
Wavelength Selection

Feedback
- 25 different DFB grating periods
- Defined in a single photolithography step
- MOVPE regrowth

Processing of lasers
- Standard lithography processes
- Wet etching to 11-17 μm-wide ridges
- Cleaved in 1.5 mm-long bars
- HR back facet coating
7.8μm DFB QCL Characteristics

- High cw output power = 80 mW at 15°C
- High temp. operation > 40°C
- Large temp. tunability
- 30 dB side mode suppression ratio

9.6 μm DFB QCL Characteristics

- High cw output power (100mW @25°C)
- High temp. operation > 50°C
- Large temp. tuneability

10.65 μm CW QCL (Buried ridge) Characteristics (288K-358K)

- Max. CW operating temp > 350K
- Max. CW power @ 25°C = 143mW
- Emission wavelength @ 25°C = 10.65μm
- Far-field FWHM=43°×67°
- Epi-up bonding

11.5 μm CW QCL (Buried ridge) Characteristics (288K-343K)

Max. CW operating temp > 340K
Max. CW power @ 25°C = 115mW
Emission wavelength @ 25°C = 11.54 μm

CW DFB QCL at 7.8μm: performances

j_{th} = 2.62 kA/cm² (+30°C)
T_0 = 113K
T_{max} = 35°C
λ ~ 1206 cm⁻¹ (8.3μm)

j_{th} = 1.87 kA/cm² (+30°C)
T_0 = 109K
T_{max} = 60°C
λ ~ 1256 cm⁻¹ (8.0μm)

j_{th} = 2.45 kA/cm² (+30°C)
T_0 = 98K
T_{max} = 45°C
λ ~ 1302 cm⁻¹ (7.7μm)
CW DFB QCL at 7.8\,\mu m: Spectra

Single-mode emission with SMSR > 25 dB
(resolution limited by FTIR)

Wavelength coverage:
One laser: $\Delta \nu \sim 10$-15 cm$^{-1}$
In total: $\Delta \nu > 100$ cm$^{-1}$
(7.7 - 8.3 \, \mu m)

Average $R_{th} \sim 12.4$ K/W
Average tuning coefficient $\beta \sim -8.88 \times 10^{-5}$ K$^{-1}$
CW DFB QCL at 7.8μm: wavelength spreading

Average emission wavelength
At T = 0°C
Gain versus threshold current density

- Lowest threshold current density at gain center
- No trend with ridge width
A QCL on Mars ...

QCL for Tunable Laser Spectrometer integrated in the Sample Analysis at Mars Instrument Suite (SAM), a component of the Mars Science Laboratory.

MSL goal: chemical composition of different gases present at the surface of Mars

-> “suitable information to determine if the conditions of life have been previously assembled on the planet”

QCL mission: measurements of isotopic ratio at 7.79 μm
<table>
<thead>
<tr>
<th>Specification</th>
<th>Value</th>
</tr>
</thead>
<tbody>
<tr>
<td>Wavelength</td>
<td>1283.6 cm⁻¹ (7.79 μm)</td>
</tr>
<tr>
<td>Tuning range</td>
<td>1 cm⁻¹ (1283.1 - 1284.1 cm⁻¹)</td>
</tr>
<tr>
<td>Spectral linewidth</td>
<td>&lt; 20 MHz (RT-CW)</td>
</tr>
<tr>
<td>Heatsink temperature</td>
<td>305 K</td>
</tr>
<tr>
<td>Laser temperature</td>
<td>20-25°C</td>
</tr>
<tr>
<td>Max. laser I / V</td>
<td>600 mA / 10 V (6 W max.)</td>
</tr>
<tr>
<td>Output power</td>
<td>&gt; 2 mW</td>
</tr>
<tr>
<td>Max. cooler I / V</td>
<td>2.1 A / 4.5 V (9.45 W max.)</td>
</tr>
</tbody>
</table>

Shall survive the trip!

-> MIL specifications:

5000 hours of operation, radiation, heating, thermal cycles (-40 to +85°C), mechanical shocks, acceleration (up to 20 G),...
Final QCL Module

Size: 47.2 x 25.4 x 11.2 mm
Lifetime measurements

Ageing module #A14, cw, 10°C

Power [mW] vs Time [h]
High Performance 4.6 μm CW, RT QC Laser - 2008

Performance of Adtech – Princeton MIRTHE CW RT FP QC Lasers

June 23, 2008

October 30, 2008
Key Characteristics of mid-IR QCLs and ICL Sources - 2008

- **Band – structure engineered devices**
  (emission wavelength is determined by layer thickness – MBE or MOCVD; mid-infrared QCLs operate from 3 to 24 µm (AlInAs/GaInAs)

- Compact, reliable, stable, long lifetime, and commercial availability

- Fabry-Perot (FP), single mode (DFB) and multi-wavelength

- **Spectral tuning range in the mid-IR**
  (4-24 µm for QCLs and 3-5 µm for ICLs)
  - 1.5 cm⁻¹ using injection current control
  - 10-20 cm⁻¹ using temperature control
  - >200 cm⁻¹ using an external grating element and heterogeneous cascade active region design; also QC laser array (Harvard)

- **Narrow spectral linewidth**
  - CW: 0.1 - 3 MHz & <10Khz with frequency stabilization (0.0004 cm⁻¹)
  - Pulsed: ~ 300 MHz (chirp from heating)

- **High pulsed and cw powers of QCLs at TEC/RT temperatures**
  - Pulsed and CW powers of ~ 1.5 W; high temperature operation ~300 °C
  - >50 mW, TEC CW DFB @ 5 and 10 µm
  - >600 mW (CW FP) @ RT & wall plug efficiency of ~12. % at 4.6 to 45 nm

Why are long wavelength QCLs challenging?

- Optical absorption increases with wavelength
- Upper laser level lifetime > lower state lifetime

![Graph showing waveguide loss vs. wavelength for different carrier densities](image1)

![Graph showing transition time vs. wavelength](image2)
15 μm excited state QC laser

- $E_{ul} = 83.6$ meV
- $z_{ul} = 50.3$ Å
- $\tau_5 = 0.586$ ps
- $\tau_4 = 0.215$ ps
- $\tau_{54} = 1.9$ ps
- FoM = 1313 ps Å²
- $z^2\tau_5 = 1483$ ps Å²
- $z^2\tau_5E = 124$ ps Å² eV

E = 31 kV/cm

Energy (meV)

Position (Å)
Widely Tunable, CW, TEC Quantum Cascade Lasers
FP QCL suitability for an external cavity platform

- Power,
- CW operation,
- Broad gain,
- Far field pattern,
- Life-time,
- Components need to be compatible with mid-IR radiation
- Micro-optic components required for compact lasers
- QC Gain Media needs to be AR coated and single mode
- QC attach needs to be robust and high thermal conductivity
- Filter tuning needs to be wavelength and phase controlled
Key technology and components

QC gain media
AR coatings
Micro-Optic lenses
Wavelength tuning and filtering
Instrument control
• EPI up or down can be accommodated
  – Buried Heterostructures or Ridges
  – EPI down mounting for most efficient heat removal
  – Solder attach for high temperature operation

• Multiple commercial suppliers
  – Adtech
  – Hamamatsu
  – Alpes
  – Maxion
  – Laser Components
  – Nanoplus
  – III-V Labs
- MID IR coating materials required
- Broad band performance required (thick coatings)
- Reflectivity's below 0.001 for optimum performance
- Long lifetime, high stress, high heat coatings
Micro-Optic Lenses

- Diffraction limited performance required
- High N.A. (>0.8)
- Low wave-front error manufacturing
- MID IR materials (ZnSe, Ge, Chacolgenide..)
- Aspheric surfaces required
Wavelength filter and filter tuning

- Diffraction gratings provide high feedback (>80%)
- Disk drive or MEMS tuning are options
- Pulsed lasers do not need MHF (phase continuous) tuning
- High resolution tuning requires wavelength and phase control
- Digital encoders for wavelength readout
Instrument control

- Temperature control to 10mK
- Current control to \(~2A\)
- High compliance voltages (up to 20V)
- Wavelength control to $1:10^5$
- Pulsed and CW operation
- Computer interface
Tunable external cavity QCL based spectrometer

- Fine wavelength tuning
  - PZT controlled EC-length
  - PZT controlled grating angle
  - QCL current control
- Motorized coarse grating angle tuning
- Vacuum tight QCL enclosure with build-in 3D lens positioner (TEC laser cooling + optional 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
- Power output is ~ 11mW
- Access to Q(3/2) transition of NO at 1875.8 cm⁻¹ for LMR spectroscopy

Performance of 8.4 μm cw EC-QCL Spectroscopic Source

T = 243K, I_{QCL} = 420mA

CW mode

Tunability 182 cm\(^{-1}\) @ 8.4 μm; (1100 to 1280 cm\(^{-1}\)); λ_c = 15%

AR coating:

R_{AR} \approx 2 \times 10^{-4}

P_{EC-opt} : \sim 50 \text{ mW} @ -30 \text{ C}, \text{ also }\sim 100 \text{ mW} & 201 \text{ cm}^{-1} @ 8.8 \text{ μm} \& 30 \text{ C with 2008 QCL technology}
EC-QCL Noise

- EC-QCL outperforms DFB-QCLs in terms of RF noise figure
- EC feedback reduces laser excess noise
  - $\approx \frac{L_{\text{QCL}}}{L_{\text{EC}}}$ reduction of injection current fluctuations impact
  - Strong feedback gives $>30$dB 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
Commercial widely tunable CW EC-QCL

Mid-IR Lasers From Daylight Solutions

- CW, Mode-Hop Free
- Linewidth < 0.001 cm⁻¹
- Center wavelengths from 4 to 12 μm
- Broad tuning range up to 10%
- No cryogenic cooling
- Average power up to 50 mW
- Tuning speed: full range < 2s
- Superb wavelength accuracy
- Shipping in September 2007

Ammonia HITRAN Database

Daylight Solutions' Laser

CW Power vs Wavenumber

P-I curves vs Wavelength

DAYLIGHT SOLUTIONS
Broadband QCL Spectrometer on a Chip

Schematics of QCL spectrometer

Technical Approach

- Broadband mid-IR quantum cascade lasers
- Distributed feedback (DFB) laser array with wavelengths spanning more than 250 cm⁻¹
- Temperature tuning for continuous spectral coverage
- Computer Control

DFB QCL array performance

Emission spectrum of a 32 DFB-QCL array
Pulsed operation (80kHz, 50ns) at room temperature

Lee, Belkin, et al., APL 2007
Absorption spectroscopy

Methanol Absorption

Isopropanol Absorption

Acetone Absorption

Ammonia Gas Absorption

Lee, Belkin, et al., APL (2007)
Trace Gas Sensors Areas Explored at Rice

- Methods employed
  - Extended pathlengths (Multpass Gas Cells & Retroreflector)
  - Cavity Ringdown
  - Off-Axis Integrated Cavity Output Spectroscopy
  - Faraday Rotation Spectroscopy
  - Wavelength Modulation
  - Pulse-to-pulse fluctuation removal by comparing the same pulse on the same or another detector
  - Quartz Tuning fork based photoacoustic spectroscopy (QEPAS)
- 16 gases detected: \( \text{NH}_3, \text{CH}_4, \text{H}_2\text{S}, \text{N}_2\text{O}, \text{CO}_2, \text{CO}, \text{NO}, \text{C}_2\text{H}_2 \text{H}_2\text{O}, \text{OCS}, \text{C}_2\text{H}_4, \text{SO}_2, \text{C}_2\text{H}_5\text{OH}, \text{C}_2\text{HF}_5, \text{H}_2\text{CO}, \text{C}_2\text{H}_6, \text{HCN} \) and isotopic species of C, O and N
- Practical applications
  - Crew Health Maintenance & Life Support - \( \text{H}_2\text{CO}, \text{NH}_3 \)
  - Fire and Post Fire Detection
  - Radioactive site remediation
  - Medical breath analysis - \( \text{NO}, \text{NH}_3, \text{CO}_2, \text{CH}_3\text{COCH}_3, \text{OCS} \)
  - Industry catalyst poison - \( \text{CO} \)
  - Urban air smog - \( \text{H}_2\text{CO} \)
High resolution spectroscopy with a 5.3μm EC-QCL

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

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

External Amplitude Modulation:

- QTF is used as a mechanical chopper at $f=\sim32kHz$
- No chirp associated with the laser current modulation
- High resolution mode-hop-free tuning is possible
Applications

High Resolution Spectroscopy (NO)

- Step control of center wavelength over full tuning range phase continuously
- Fine tuning with PZT to achieve inter-step continuous tuning
- Doublets can be easily resolved with laser linewidths below 10MHZ
EC-QCL based Magnetic Rotation Spectroscopy
Magnetic Rotation Spectroscopy of Nitric Oxide

- 96 ppb NO on N₂
- $I_{qd}=850 \text{ mA}$, $P=40 \text{ Torr}$
- $TC=1 \text{ s}$, $SEN=2 \text{ mV}$
- $freq_{\text{mod}}=950 \text{ Hz}$, $B=110 \text{ Gauss}$
- $\Phi=3^\circ$ from crossed analyzer
- LN cooled InSb detector

SNR=253
MDL(1σ)=380 ppt

$1\sigma=4.325 \mu\text{V}$

Wavenumber [cm⁻¹]

1875.66 1875.72 1875.78 1875.84 1875.90
National Stadium, Beijing, July-Sept. 2008
Monitoring of broadband absorbers

- Freon 125 ($C_2HF_5$)
  - Refrigerant (leak detection)
  - Safe simulant for toxic chemicals, e.g., chemical warfare agents
- Acetone ($CH_3COCH_3$)
  - Recognized biomarker for diabetes
- TATP, Acetone Peroxide ($C_6H_{12}O_4$)
  - Highly Explosive
8.4 μm RT CW EC-QCL based QEPAS trace gas sensor

QEPAS Characteristics:

- High sensitivity (ppm to ppb)
- Excellent dynamic range
- Immune to environmental noise
- Ultra-small sample volume (< 1 mm³)
- Sensitivity is limited by the fundamental thermal TF noise
- Compact, rugged and low cost
- Potential for trace gas sensor networks

Medium power: narrow linewidth: high volume packaging

- Single ended output powers > 0.85W at wavelengths of 4.5-5.5um and T > 15C
- Long lifetime, high reliability, robust packaging with high wavelength stability
- Linewidths < 0.01cm\(^{-1}\) using external cavity
- Manufacturable wavelength setting to 1:1000 Independent of temperature
Integrated molecular detection subsystems

- Fully integrated microprocessor for spectral de-convolution
- Broad tuning ranges
- Multi-species identification
New design of fast broadly tunable EC-QCLs (2009)

- New optical configuration
  *Folded cavity (configuration #1)*
- Fast tuning capabilities:
  - Coarse Broadband Scanning
    \((\sim 55 \, cm^{-1} \, \text{at} \, 5 \, \mu m) \text{ up to } 5 \, kHz\)
    (compared to available technologies <10Hz)
  - High resolution mode-hop free tuning
    \((\sim 3.2 \, cm^{-1} \, \text{at} \, 5 \, \mu m) \text{ up to } 5 \, kHz\)
    (compared to available technology 100-200 Hz)

Conceptual Design of Ultra-compact QCL Trace Gas Sensor

Aerodyne, Inc, Barry McManus et al, 2008
PHOTONS v4.0 - 2.7μm CO2 Direct Absorption Based Sensor

- Small size
- Relatively low cost
- High efficiency switching power supplies
- PWM Peltier cooler driver
- 0.2W control system power consumption
- Detection sensitivity of CO₂ 1 ppm with 1sec. lock-in time constant
- Over 100x improvement in sensitivity is possible @ 4.2μm
Ultra-compact Diode Laser based Trace Gas Sensor
Future Directions of Mid-IR Sensor Technology

- Major QCL improvements in areas such as wall plug efficiency, temperature performance, beam shape and price reduction
- Improvements of the existing sensing technologies using novel, thermoelectrically cooled, cw, high power, and broadly wavelength tunable mid-IR interband and intersubband quantum cascade lasers.
- New applications enabled by novel broadly wavelength tunable quantum cascade lasers (especially sensitive concentration measurements of broadband absorbers, in particular VOCs and HCs)
- Development of optically multiplexed gas sensor networks based on LAS or QEPAS
- Wearable sensors for medical & biomedical diagnostics (NO, CO, COS, CO$_2$, NH$_3$, C$_2$H$_4$)