Quantum Cascade Lasers Smell Success

Iain Howieson

Erwan Normand

Michael T. McCulloch

The development of the Quantum Cascade Laser, innovative spectroscopic techniques such as Intra Pulse Spectroscopy and recent advances in spectrometer hardware promise to deliver a major step change in the sensitivity, speed of operation, fingerprinting capability, size and cost of tuneable diode laser absorption gas sensors.

History

Since they were first demonstrated at Bell Laboratories in 1994, Quantum Cascade Lasers (QCL's) have been gaining acceptance as the mid-infrared (IR) source of choice. Their shift out of the laboratory into real world applications has been accelerated by the step change in performance that these devices can deliver in fields as diverse as range finding, electronic counter measures, Free Space optical telecoms and chemical sniffing. It is in this last field, chemical sniffing, that perhaps the biggest opportunities can be found as the combination of QC lasers and recent gas sensor developments promise to deliver levels of spectroscopic performance in terms of detection and selectivity that will open up huge markets in environmental monitoring, health and safety, security, defence and medical diagnostics.

Conventional semiconductor lasers, such as the lead salt devices commonly used in the mid-IR, rely on electron hole recombination across the doped semiconductor bandgap to emit photons. The quantum cascade laser, which is about the size of a pin head, operates on a fundamentally different principle whereby electrons cascade down a series of quantum wells, which result from the growth of very thin layers of semiconductor material. Whereas a single electron-hole recombination can only ever produce a single photon, the quantum cascade laser electron can cascade down between 20 and 100 quantum wells producing a photon at each step. This electronic waterfall provides a step change in performance in terms of lasing efficiency enabling QC lasers to emit several watts of peak power in pulsed operation and tens of milliwatts CW.

The lasing wavelength for QCL's is determined not by the choice of semiconductor material as with conventional lasers, but by adjusting the physical thickness of the semiconductor layers themselves. This removes the material barriers commonly associated with conventional semiconductor laser technology and opens up the possibility of near-infrared through to THz spectral coverage. For the first time an infrared spectroscopic laser source, which has no need for cryogenic cooling, high output powers, large spectral coverage, excellent spectral quality and good tuneability has become a reality.

Inter and Intra Pulse Spectroscopy

The practical implementation of QCL's in spectroscopy started in earnest in the late 1990's with researchers eager to harness the power of a spectroscopic source spanning the full spectrum of the technologically significant mid-IR wavelengths (3 - 25 μm). Two methods of direct absorption spectroscopy have resulted from this research. Known as inter and intra pulse spectroscopy respectively they have been developed to maximise the performance of the QC laser as a spectroscopic tool.

Inter pulse spectroscopy [1] uses the QC laser in pulsed mode to facilitate its use at or close to room temperature. The optical transmission is recorded by combining ultra short current pulses to the laser with a slowly varying current or temperature ramp superimposed to tune the laser wavelength through the spectroscopic transition of interest. However, it was found that pulsing the laser in this way resulted in a frequency chirp and consequently a broadening of the laser linewidth and a reduction in spectral resolution. To help overcome this effect it was necessary to limit the pulse width to less than a few tens of ns whilst keeping the pulse amplitude close to the lasing threshold. The typical tuning range for this technique is of the order of 1 to 2 cm^{-1} with repetition rates ranging from tens of Hz through to kHz.

Inter pulse has been employed with considerable success in spectroscopy. However, the threshold current limitation, the introduction of noise due to ultra short pulse to pulse variability and the lower duty cycles attainable have prevented inter pulse spectroscopy from achieving the very highest level of sensitivity currently available to other spectroscopic techniques.

Intra pulse spectroscopy [2], like inter pulse, uses the laser in pulsed mode to effect room temperature operation. However, rather than trying to minimise the frequency chirp brought about by pulsing the QCL, the chirp is instead harnessed to provide a near instantaneous frequency sweep through the spectroscopic features of interest. Pulse widths up to several micro seconds are employed with pulse amplitudes several amps above lasing threshold to produce a top hat current pulse that causes localised heating within the laser and consequently a frequency downchirp, which is typically between 4 and $6cm^{-1}$ wide. The spectral resolution in this case is defined by the instantaneous linewidth of the laser as it sweeps in wavelength. This is simply given by:

where $d\nu/dt$ is the chirp rate and k is a form factor defined by the pulse shape [3]. Typical QCL frequency downchirps will normally have better than 0.01cm-1 spectral resolution. This is better than the inter pulse technique for the same chirp rate. Repetition rates of up to 100 kHz can be used giving high duty cycles and the resulting spectra averaged to provide excellent S/N levels.

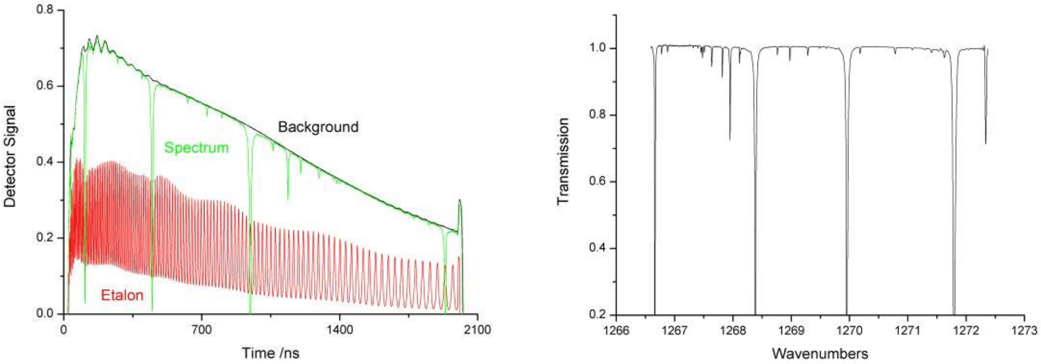

Figure 1. *Raw Data, Background and Transmission spectra of room air recorded using a 1270 cm^{-1} QCL with Intra Pulse Spectroscopy. A 2000 ns pulse is applied to the laser resulting in a frequency chirp, which sweeps the laser through the spectroscopic transitions of interest. A 0.048 cm^{-1} Ge etalon signal confirms greater than 6 cm^{-1} single mode tuning.*

Operating the laser in this quasi CW intra pulse regime provides another less obvious but significant spectroscopic advantage. The fast chirp rate can be used in conjunction with careful optical design to ensure incoherent optical feedback [4]. This is used to prevent laser feedback noise and optical fringing, which tend to be the common noise floors for most practical implementations of optical spectrometer design. The removal of this noise floor, without the need of complex fringe removal techniques such as Brewster Plate spoilers or expensive optical isolators, enables the laboratory performance of this technology to be easily transferred to real world applications.

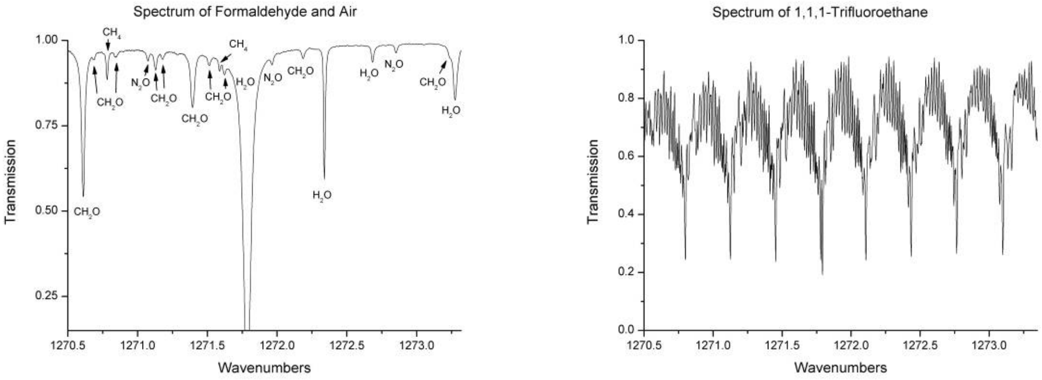

Figure 2. *Recorded spectra of Formaldehyde and 1,1,1 Trifluoroethane. These spectra highlight the excellent S/N and selectivity that can be achieved with the QCL and Intra Pulse Spectroscopy. Key features such as intrinsically fringe free operation and better than 0.01 cm^{-1} spectral resolution provide a powerful fingerprinting capability.*

Applications

The applications for laser spectroscopy, which have been opened up by the advent of the QC laser and new techniques such as intra pulse spectroscopy, are huge. For example the wide spectral tuneability, which is typically an order of magnitude greater than lead salt laser systems, raises the possibility of being able to observe anywhere up to five or six gases with a single laser. This will give access to volume markets in Health and Safety and Environmental monitoring, which would have been inaccessible to laser absorption spectroscopy in the past due to the cost and complexity associated with multiple laser systems. This tuneability and excellent selectivity can also be combined with multiple laser spectrometer designs to give broadband spectral coverage, which can potentially be applied to the identification and quantification of complex heavy molecules such as those found in toxic chemicals, explosives and drugs. Key instrumentation features such as large dynamic range, excellent sensitivity and failsafe operation combined with the high reliability associated with solid state technology will eradicate many of the technological problems associated with existing technology in these markets.

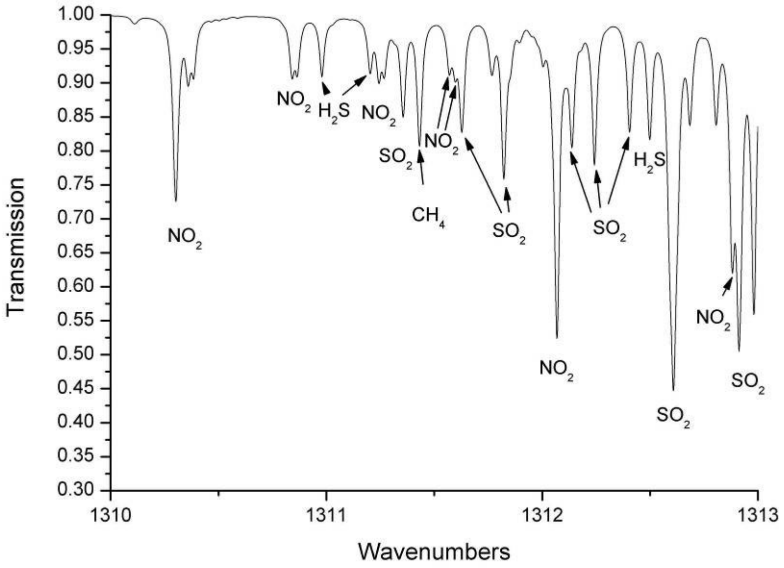

Figure 3. *A QCL transmission spectrum, which demonstrates simultaneous measurement of gases including* NO_2*,* SO_2*,* H_2S *and* CH_4*. Multiple gas measurement opens up the possibility of QC laser spectrometers entering volume markets such as Environmental monitoring and Health and Safety.*

Perhaps one of the biggest steps towards achieving significant penetration in any of these markets will come from recent developments in spectrometer hardware. The development of novel QC laser systems capable of operating in both inter and intra pulse mode, which exploit recent technological advances such as miniaturized integrated electronic systems, plug and play interfaces and micro optics, will banish the unwieldy, fragile and expensive instrumentation of the past. For example the design of innovative mid-infrared optics, which capitalise on the recent developments in both the QCL and optics industries is seen as a key step towards ruggedised industrial instrumentation. Specifically tailored to provide ultra high optical performance combined with suitability for use in harsh industrial environments the integration of micro optics can significantly reduce mechanical misalignment/drift over time. At the same time the use of integrated pulse circuitry, which minimises mismatch and enhances rise/fall time ensures excellent laser stability while a USB 'plug and play' interface can give instant access to all spectrometer control and engineering data.

Figure 4. *The QC laser system, which is capable of operating in both intra and inter pulse spectroscopic regimes. The development of compact and robust spectroscopic hardware is seen as a key step towards greater exploitation of laser absorption spectroscopy in real world industrial applications.*

These recent advances in both QCL laser technology and spectrometer hardware when combined with novel spectroscopic techniques such as intra pulse spectroscopy offers not simply a small improvement on other methods of gas detection but provides a major step change in sensitivity, speed of operation, fingerprinting capability, size and cost. These advantages will open up significant markets in environmental monitoring, health and safety, security, defence and medical diagnostics.

References

1. Namjou et al; Opt Lett 23, 219, 1998

2. Normand et al; Opt Lett 28,16, 2003

3. Michael T. McCulloch, Erwan L. Normand, Nigel Langford, Geoffrey Duxbury, D. A. Newnham; JOSA B, Vol. 20 Issue 8 Page 1761 (August 2003)

4. Normand et al; International Patent No. PCT/GB2003/001510

Iain Howieson is Director of R&D, Erwan Normand is Technical Director and Michael McCulloch is a Laser Spectroscopist at Cascade Technologies, 141 St. James Road, Glasgow, G4 0LT, Scotland, UK; e-mail: i.howieson@cascade-technologies.com, e.normand@cascade-technologies.com, m.mcculluch@casacde-technologies.com.