Recent Results from Broadly Tunable External Cavity Quantum Cascade Lasers

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ABSTRACT

Broad tunability in the mid-IR (~3-12 µm) is desirable for a number of applications. We have built a number of External Cavity quantum cascade Lasers (ECqcL™) that maximize tuning range from the quantum cascade chip. As much as 525 cm^{-1} of pulsed and 121 cm^{-1} of cw, mode hop-free tuning has been obtained. Various mechanisms for the broadening of the gain of the QC chip are considered.

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1. INTRODUCTION

The mid-IR portion of the electromagnetic spectrum (~3-20 µm) is an extremely prolific spectroscopic region. Almost all molecules have fundamental vibrational transitions in this region. A vibrational mode is "infrared active", if the change of states causes a change in the dipole moment of the molecule. In this case, a strong absorption of light at this wavelength can be observed. The fundamental vibrations in this region of the spectrum are typically 30-1000 times stronger than "overtones" or "combination bands" which occur in the more easily accessible near infrared and visible portion of the spectrum. The increased absorption strength in the mid-IR means that sensitivities can be much higher and it has been well worth increasing access to this region.

The merit of access to the mid-IR is seen in Figure 1. A sampling of molecules is placed on the chart where their respective fundamental vibrations occur in the mid-IR. Molecules of interest to the atmospheric sciences, health, and Homeland Security communities are all displayed. For instance, NOx, SOx, ozone, and methane are all critical Greenhouse Gases. NO is a known marker for asthma, and glucose is, of course, the key analyte in diabetes. Nerve agents such as Sarin and VX are detectable in the mid-IR, as are many explosives, including TNT and TATP.

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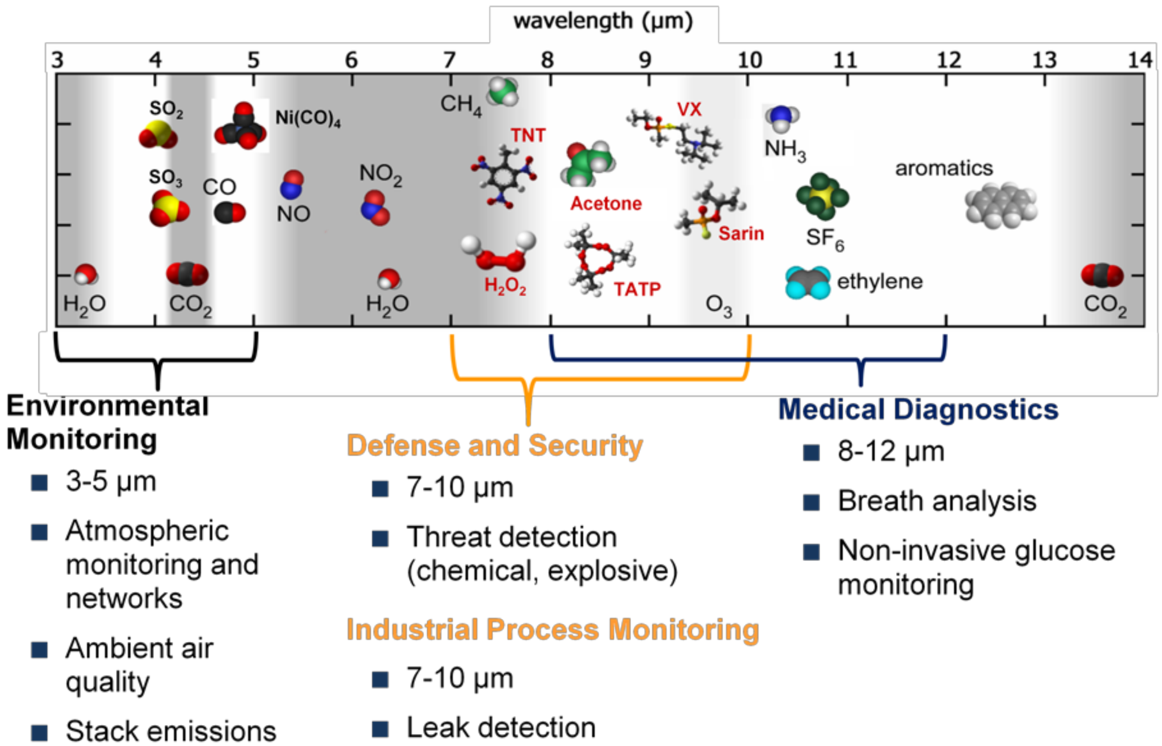

Figure 1. Molecules of interest accessible in the Mid-Infrared.

For 40 years the Fourier Transform Infrared Spectrometer (FTIR) has held the unquestionable and enviable position of the superlative commercial technology for obtaining an IR spectrum.[1] Non-linear devices such as Optical Parametric Oscillators (OPOs) and Difference Frequency Generation techniques (DFG) have for a number of years offered access also, but their very nature as non-linear devices has made them a challenge to operate and less robust than the simpler strategy of the FTIR. They have largely been the domain of those investigators willing to build and maintain a complex custom spectrometer.

The relatively new technology of Quantum Cascade Lasers, invented in 1994[2], has finally begun to chip away at the glowbar's preeminence as the mid-IR source. Glowbar sources have broad spectral emission but fundamentally poor beam quality and very low spectral brightness. Quantum Cascade Lasers (QCLs) are high spectral brightness, electrically pumped semiconductor lasers with diffraction-limited performance from 3.15 μm[3] to at least 63 μm (4.7 THz)[4]. QCLs have less gain bandwidth than a glowbar, but much has been done to increase the coverage available from single chips.

2. TUNING MECHANISMS

Most spectroscopic investigators lack the ability to control both the bandwidth and emission wavelength of QCL chips. QCLs operating in Fabry Perot mode can produce light at any and all wavelengths within its gain curve, but unpredictable gain nonlinearities result in fluctuations in intensity and variations in spectral width. Without feedback, the emission wavelength is determined largely by these inaccessible chip gain dynamics. Figure 2 is a series of lasing spectra obtained for a typical QCL operating in Fabry Perot mode from 820-950 mA. For changes in operating current of a few mA, the spectral emission can shift, broaden, and even collapse to relatively small values. For the 4.0 μm chip shown in the figure, different current regimes produce spectra as narrow as 10 cm^{-1} (16 nm) or as broad as 60 cm^{-1}(96 nm). Figure 3 shows the sub-threshold gain of a chip centered around 4.4 μm. The bandwidth of the gain exceeds 700 cm^{-1}. Describing methods for taking advantage of this broad gain is the goal of this paper.

One approach to controlling the linewidth and frequency of a QCL is to epitaxially grow a Distributed Bragg Reflector (DBR) onto the surface of the waveguide. By controlling the temperature of the gain chip, this Distributed Feedback

(DFB) system can be caused to tune with very narrow linewidth over a small frequency range (roughly 5-15 cm^{-1}). But a large portion of the gain bandwidth is sacrificed to this on-chip wavelength control. Furthermore, it has proven to be quite difficult to center the DBR at a specific wavelength making them difficult to depend on in a commercial setting. A less limiting tuning mechanism is needed to take advantage of the raw performance of some QCLs.

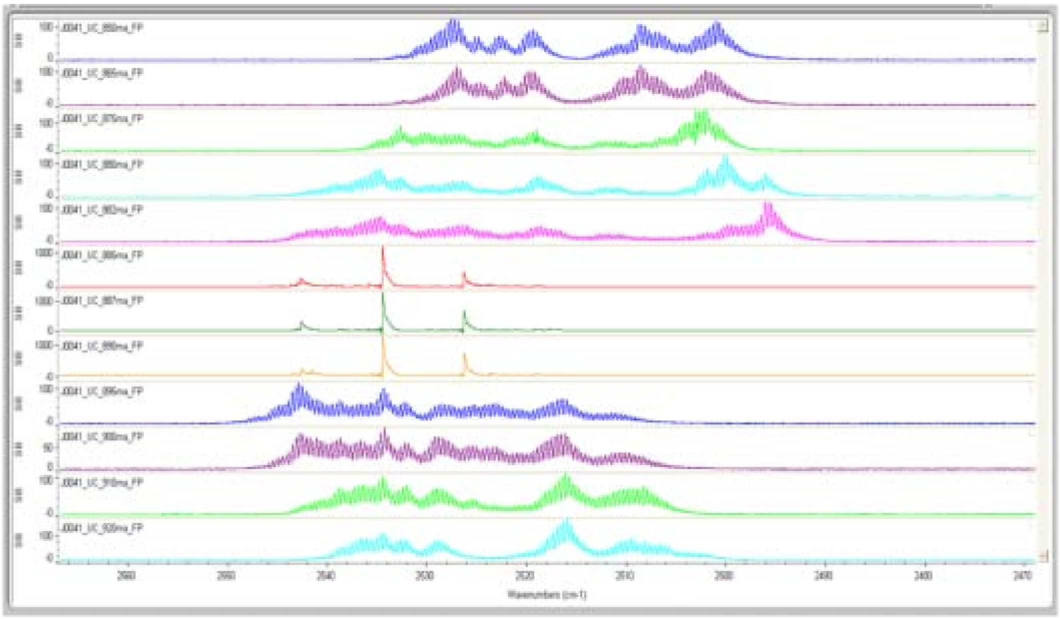

Figure 2. Spectra of a typical QCL at varying drive currents.

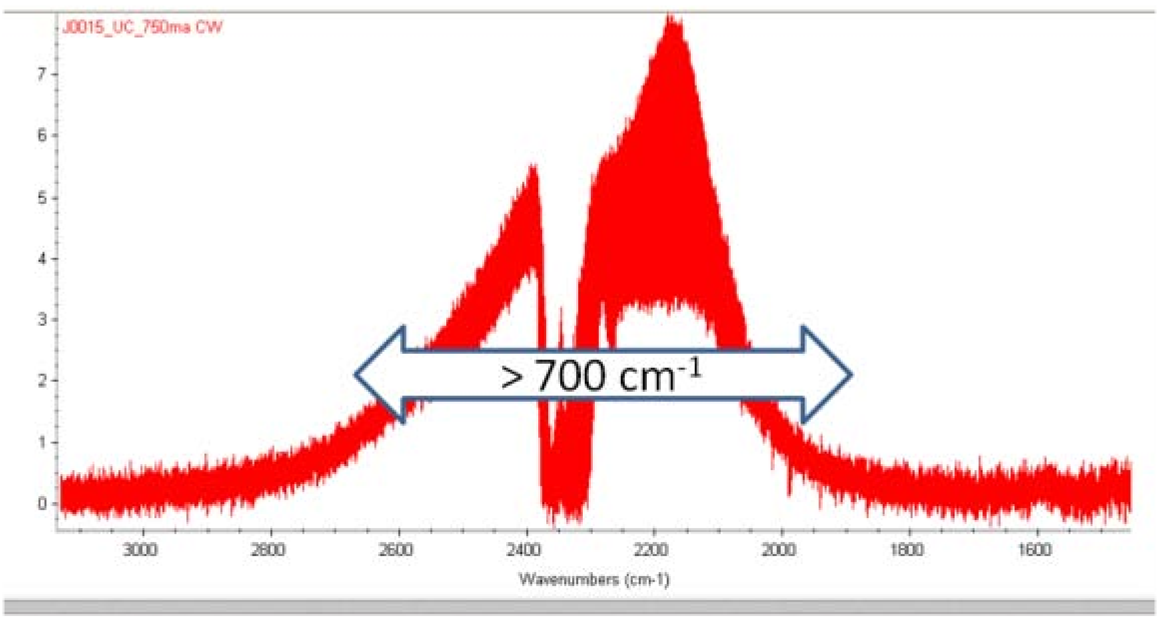

Figure 3. Sub-threshold gain of broad tuning chip. The dip in the gain curve is caused by ex-chip CO_2 absorption and is not a feature of the chip itself.

There are novel methods of tuning QCLs beyond the DFB configuration that invoke temperature tuning of a frequency-selective element in the cavity[5] or taking advantage of the time evolution of the spectral content of a short (200 ns) pulse.[6] But the most effective method for controlling both the linewidth and wavelength while accessing the entire gain bandwidth of the chip is by employing an External Cavity. In its simplest form, the External Cavity consists of the gain chip, collimating lenses and a rotatable diffraction grating (Figure 4). As we shall see, with proper mechanical and thermal design, the External Cavity architecture can provide for over 500 cm^{-1} of controlled tunability from a single gain chip while maintaining linewidths of less than 1 cm^{-1}.

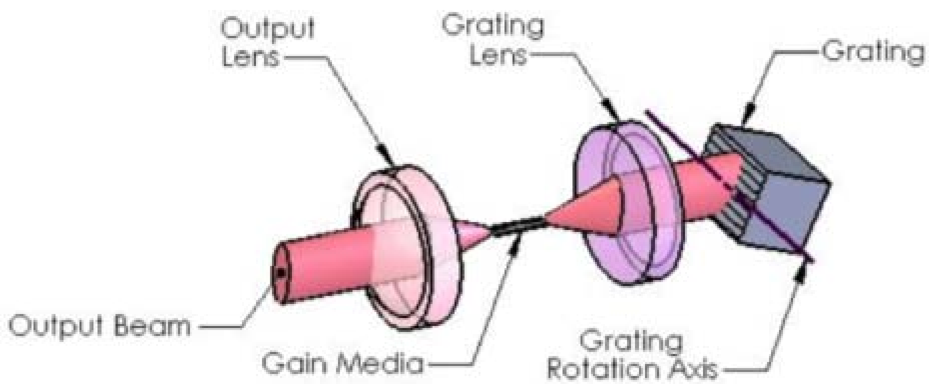

Figure 4. External Cavity geometry for controlling linewidth and wavelength output of a quantum cascade laser.

3. RESULTS

Various approaches have been employed to broaden the bandwidth of QCL chips. The results given below are all obtained in External Cavity configurations with well-controlled bandwidths. The broadest tuning results invariably come from pulsed operation of the gain chip due to the thermal load imposed on the chip when operated in cw mode. Therefore the results provided below are mostly for pulsed operation. Only the mode hop-free results touch upon cw operation.

3.1. Chip Design

The basic structure of the series of quantum wells that make up the cascade can be modified in a number of ways to increase the bandwidth of a given chip.

3.1.1. Bound-to-Bound

The simplest form of quantum well structure is that of Bound-to-Bound energy levels where within the well there are discreet upper and lower states in the lasing transition. The resulting emission is relatively narrow since both states are well-defined. Figure 5 is a visualization of this system where the resultant bandwidth of the laser light is shown to the right. Figure 6 displays the cw tuning range obtained from such a chip. On the order of 70-100 cm^{-1} can be obtained (5-10% of the center wavelength).

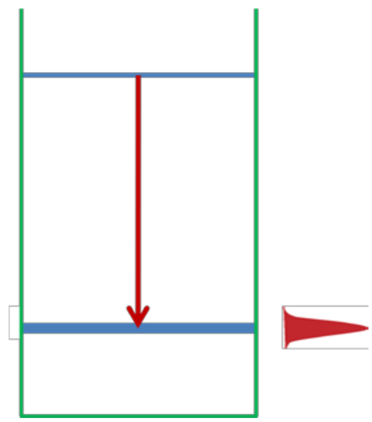

Figure 5. Cartoon of a bound-to-bound transition in a quantum well. The red arrow represents the lasing transition. Side image: depiction of bandwidth of resulting laser light.

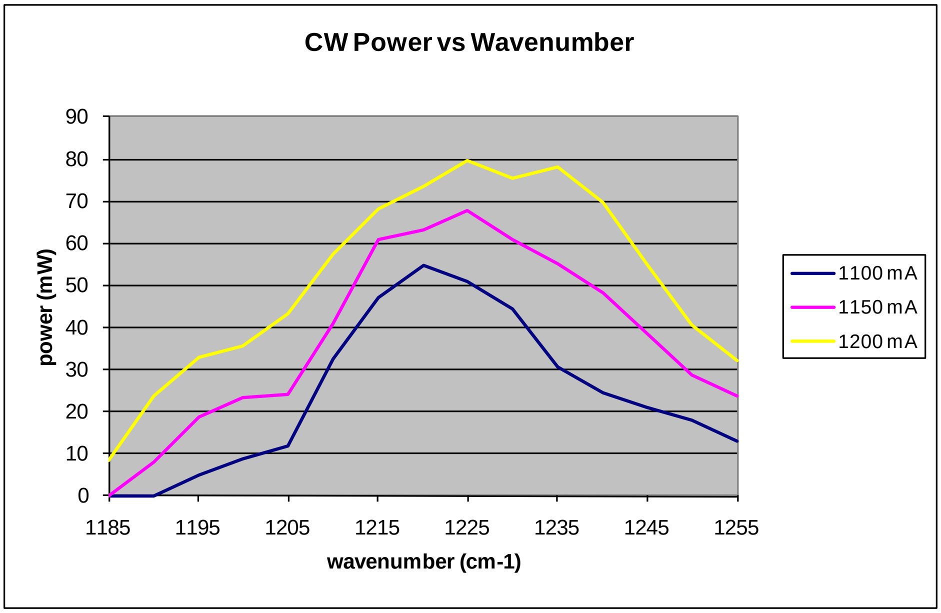

Figure 6. Typical tuning curve of a bound-to-bound chip morphology.

3.1.2. Continuum States

If either the upper or lower state (or both) of the lasing transition is replaced by a continuum of states, the spectrum of the laser is broadened substantially.[7,8] Figure 7 illustrates the concept of the bound-to-continuum and continuum-to-bound structures. Taking advantage of the continuum states provides for considerably more bandwidth. Figure 8 demonstrates that a bound-to-continuum energy level configuration can provide tuning over 26% of the center wavelength. 2.4 µm or 305 cm^{-1} or continuous tunability is demonstrated. A continuum-to-continuum structure has also been developed which presumably provides further broadening of the gain profile.[9]

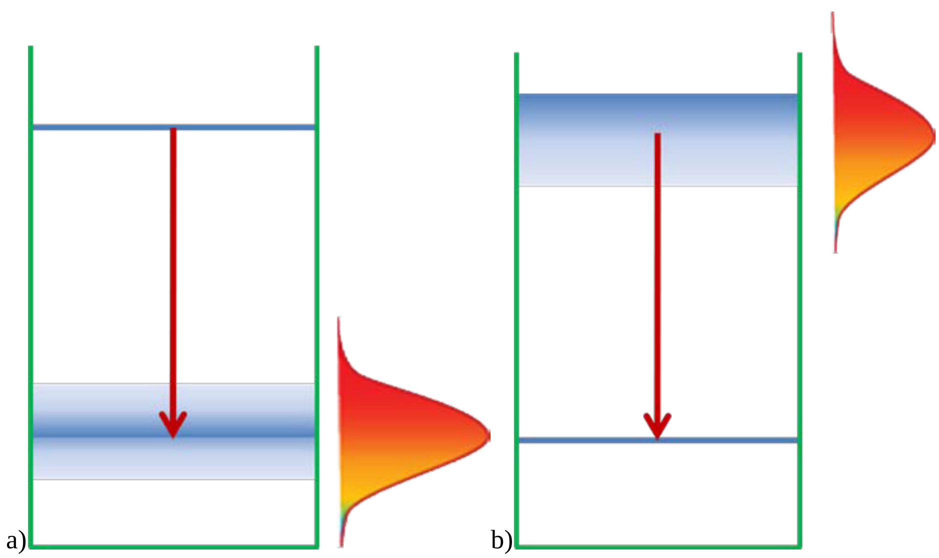

Figure 7. Cartoon of a) bound-to-continuum and b) continuum-to-bound energy level structures with their concomitant broadening illustrated to their left.

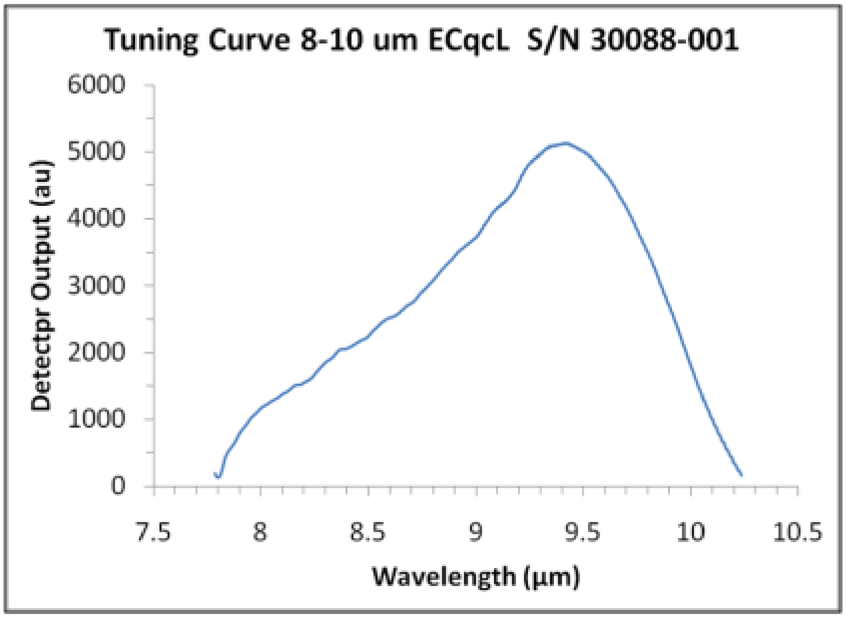

Figure 8. Tuning curve obtained for a bound-to-continuum energy level structure.

3.1.3. Heterogeneous Structures

Further gain broadening can be achieved by compositing within the same chip quantum wells with different structures. So-called heterogeneous structures compounded with continuum energy levels are currently the state-of-the-art in broadband QCL chips. Since the structure contains more than one well depth, it is to be expected that the gain profile would have more than a single maxima as can be seen in Figure 10. The tuning range for this chip is over 525 cm^{-1} which, while it represents our broadest tuning chip in wavenumbers, is only 22% of the gain center because this chip lases as a shorter wavelength (4.2 µm center).

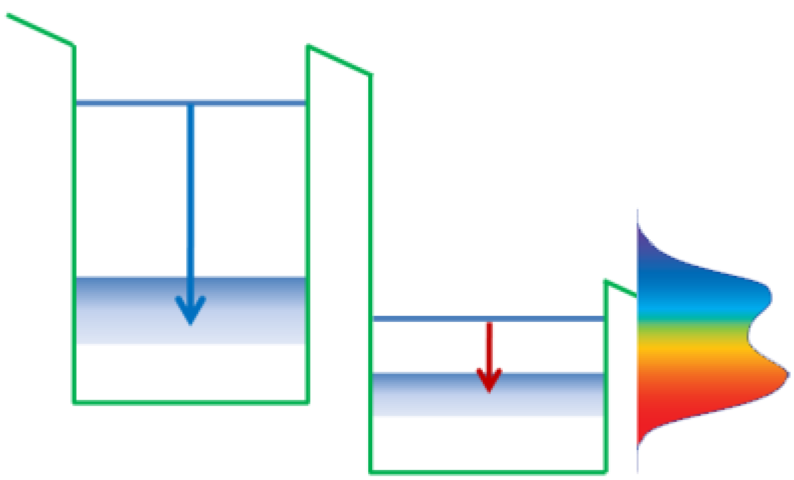

Figure 9. Cartoon of a heterogeneous quantum well design with continuum lower states. Notice that the well depth is exaggeratedly different in the two wells.

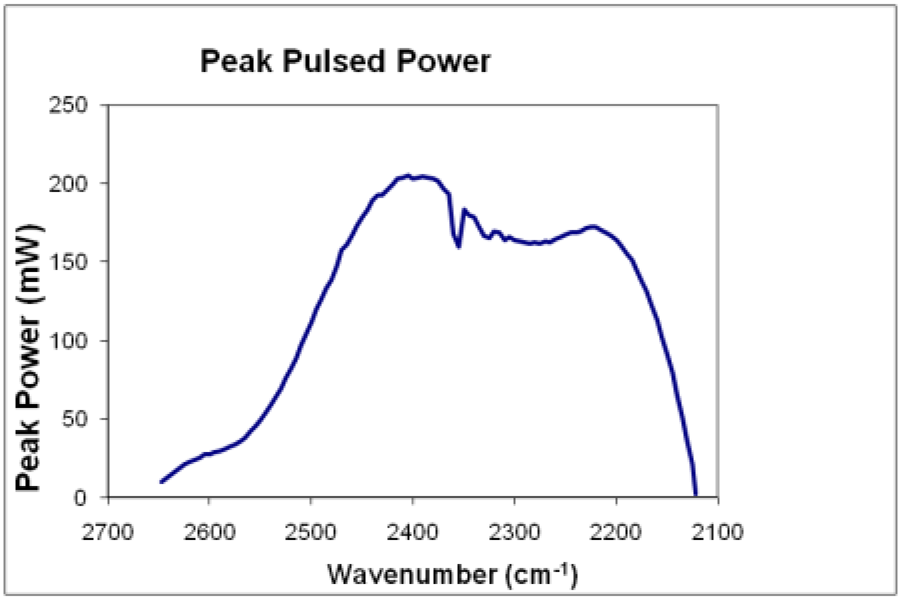

Figure 10. Tuning curve for a heterogeneous quantum well structure employing a bound-to-continuum energy level structure.

3.2. Broad cw Tuning

The broad tuning results presented thus far have focused on getting the most coverage from a single chip. These broad tuning ranges have all been obtained with pulsed laser sources. But another need is for cw performance and specifically for mode hop-free tuning. Here again, broad tuning can be quite desirable. In this instance, the performance is typically not limited by the gain of the chip, but by cavity engineering. The boundary condition for a system to tune without mode hops is that the change in cavity length must match the change in wavelength as the cavity is tuned. This then becomes an exercise in geometry and fine mechanical engineering.[10] These design constraints can be applied to quantum cascade systems to provide broad, truly mode hop-free tuning. Linewidths as narrow as 45 MHz (0.0015 cm^{-1}) have been achieved in our laboratories and Mukherjee *et al.* report a factor of two improvement over this.[11] In stabilized cavities, linewidths as narrow 5 kHz have been reported[12] and the natural linewidth is reported to be hundreds of Hertz.[13]

The gross features of the mode hop-free cavity do not differ dramatically from that shown in Figure 4. However the location of the pivot point about which the grating rotates becomes crucial as this is what allows for meeting the boundary condition necessary for mode hop-free performance. In addition, cavity length, collimation optics, and coatings must be optimized carefully. When all of these interdependent parameters have been optimized it is possible to tune the external cavity phase continuously. We have achieved as much as 121 cm^{-1} (from 4.37 to 4.62 µm; Figure 11) of mode hop-free tuning from a single laser system. Modulation of the power output caused by the residual reflectivity of the QC facets provides convenient verification of phase continuous performance of the system.

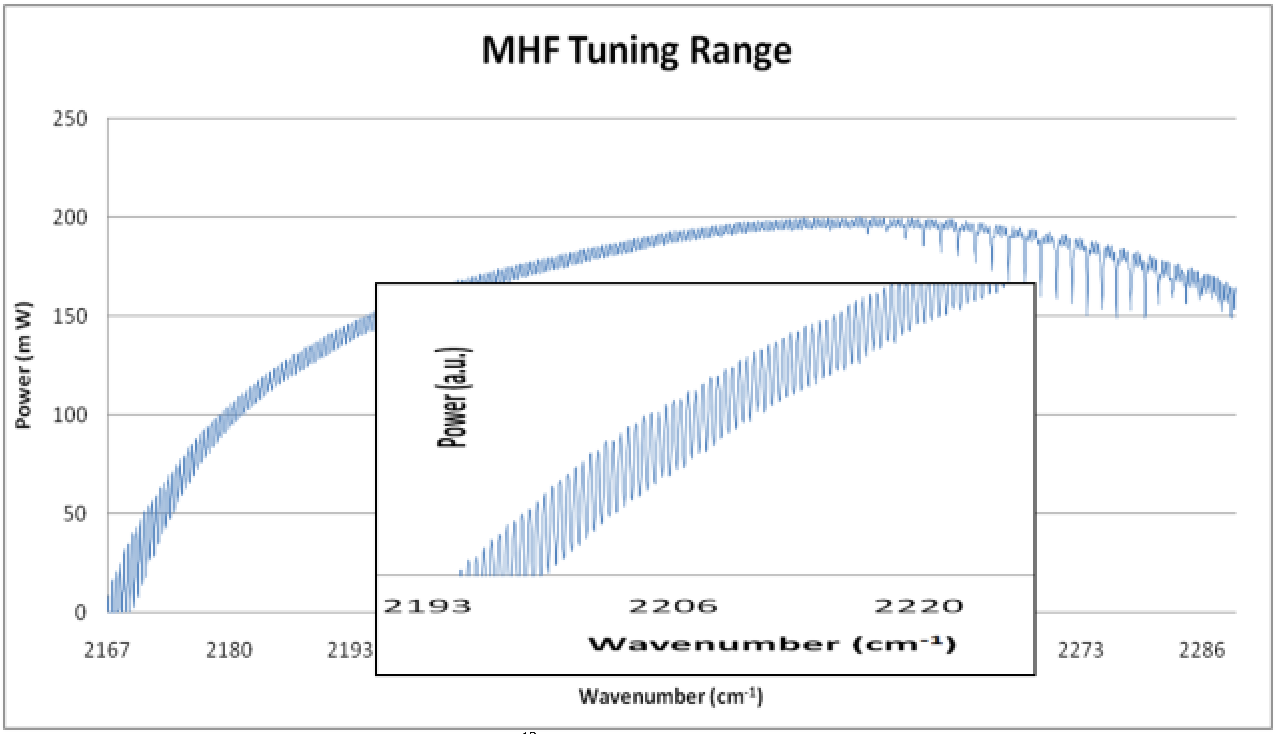

Figure 11. Wide-tuning mode hop-free performance. $^{13}CO_2$ absorption lines interrupt the tuning curve at high wavenumber. Inset: expanded view to show chip mode modulation.

3.3. Outlook

As chip designers and developers respond to the community's need for broad tuning, performance in this category continues to develop. Figure 12 is a chart of reported tunability of QCL systems over time. The upward trend is good news for those who wish to make use of the high-brightness, spectral purity, coherence, and low-divergence of packaged QCL systems, but need broad tuning for chemical analysis, sensing, and imaging experiments.

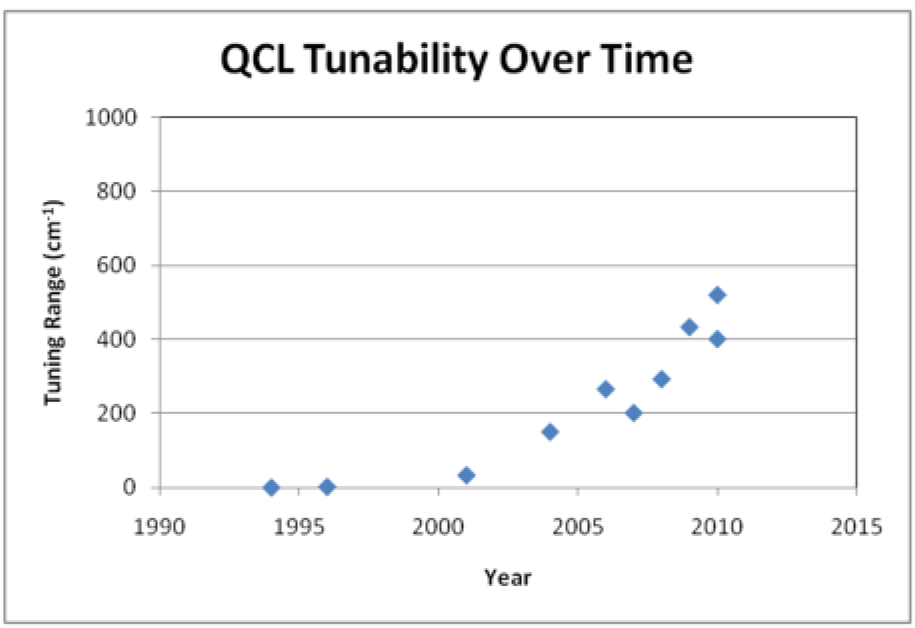

Figure 12. Chart of reported tunability vs. year reported. The trend to broader tunability will serve the research and commercial markets.[14,15,16,17, 18,19]

4. APPLICATIONS

Once the broad tuning of the chips discussed above is integrated into an external cavity, they can be put to use in many scenarios. The bound-to-continuum chip covering over 2 µm centered at 9 µm was built into a wavelength-sweeping spectrometer and used to collect the spectra of ethanol, methanol, and isopropyl alcohol, as well as other chemicals. Figure 13 shows the individual spectra compared to their respective reference spectra. Each spectra was obtained by averaging ten sweeps directed through a multipass flow cell with an effective path length of 1.22 m. Each sweep requires 10 ms and so this signal-to-noise is obtainable in 100 ms. Concentrations of the gases ranged from 500 to 900 ppm in approximately 1 atm of N_2. The spectrometer can be configured such that by comparison to reference spectra, it is able to identify and discriminate between a mixture of these and other analytes at the single-digit ppm level with a 1 second update rate.

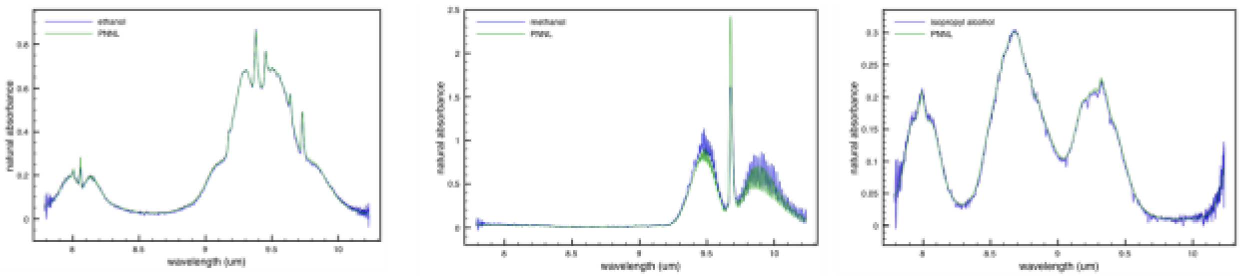

Figure 13. Spectra of ethanol, methanol, and isopropyl alcohol obtained with broad-tuning spectrometer (blue) and comparison to reference spectra (green) obtained from the PNNL database.[20]

Inspection of Figure 10 reveals a notch in the spectrum of the heterogeneous chip around 2370 cm^{-1}. This is the result of incomplete purging of CO_2 from the beam path. By passing the beam through the laboratory air over a path of about 20 cm and expanding the frequency axis, the full CO_2 spectrum, including the ^{13}C isotopologue becomes visible (Figure 14.) A device based on this chip would be useful for CO_2 monitoring and as the isotopes can be distinguished, information regarding the source of the CO_2 and the status of the probed environment can be obtained.[21]

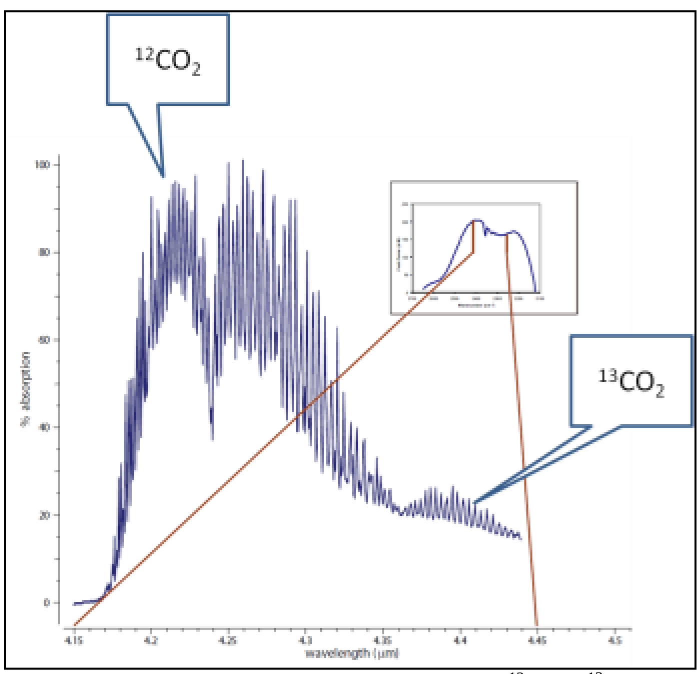

Figure 14. Single-pass spectrum of CO_2 without background subtraction. Both the ^{12}C and ^{13}C isotopologues are resolved. Inset: full tuning range of External Cavity Laser.

5. CONCLUSIONS

We have built several External Cavity quantum cascade Laser systems (ECqcL™) designed specifically to maximize the tuning range available. Using variously designed QCL chips these ECqcL provide up to 525 cm^{-1} of tuning from a single-chip resonator design. These systems will find application in imaging, remote sensing, sample analysis, and elsewhere when the ability to view a large spectral region provides a performance enhancement. Pulsed systems with linewidths on the order of 1 cm^{-1} and cw systems on the order of 0.0015 cm^{-1} have been demonstrated.

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