

**CFA** 8:00 am–9:45 am  
Room: 101B

**Laser-Based Gas and  
Chemical Sensing**

Frank K. Tittel, Rice Univ., USA, Presider

**CFA1** 8:00 am

**Chemical Sensing with a Pulsed 16  
Microns QC-DFB Laser**

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A compact, tunable mid-IR laser source is the most critical opto-electronic component in a spectroscopic molecular sensor. Until recently, the choice of such sources was limited to cryogenically cooled Pb-salt diode lasers and low-power devices based on difference frequency generation (DFG). The development of quantum cascade (QC) lasers and especially single-frequency devices with distributed feedback (QC-DFB) provided an attractive new option for IR absorption spectroscopy. QC-DFB lasers operating at near-room temperature (i.e., temperature accessible with a thermoelectric module) are of particular interest for practical applications.

Presently, the thermoelectric temperature management can be achieved only in a pulsed operation mode of a QC-DFB laser. Successful applications of pulsed QC-DFB lasers operating in the 4  $\mu\text{m}$  to 10  $\mu\text{m}$  range to spectroscopic detection of various molecules<sup>1-4</sup> have been reported to date. We shall report the first spectroscopic quantification of CO<sub>2</sub> and other species using a long-wavelength ( $\lambda = 16 \mu\text{m}$ ) thermoelectrically cooled pulsed QC-DFB laser. Long-wavelength QC lasers make accessible a spectral region where some large molecules, such as benzene exhibit a rotationally resolved structure, thus facilitating their detection and quantification.

The laser was driven with  $\sim 20$  ns long current pulses at a repetition rate of 1 MHz. The lasing threshold varied from 7A to 10A in the heat sink temperature range of  $-40^\circ\text{C}$  to  $+30^\circ\text{C}$ . CO<sub>2</sub> and H<sub>2</sub>O absorption lines were used for precise calibration of the laser frequency (Fig. 1). From these data, the temperature tuning coefficient was found to be  $-0.048 \text{ cm}^{-1}/^\circ\text{C}$ . At optimized conditions that include current pulse duration, laser current, and electric coupling, the observed FWHM of the low-pressure CO<sub>2</sub> absorption line was 210 MHz, which assumes a FWHM laser linewidth of  $\sim 150$  MHz.

Different approaches to the laser frequency manipulation for spectroscopic data acquisition will be discussed, and the achieved gas-sensing performance characteristics reported.

**References**

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3. D.M. Sonnenfroh, W.T. Rawlins, M.G. Allen, C. Gmachl, F. Capasso, A.L. Hutchinson, D.L. Sivco, J.N. Baillargeon, and A.Y. Cho, "Application of balanced detection to absorption measurements of trace gases with room-temperature, quasi-cw quantum-cascade lasers," *Appl. Opt.*, vol. 40, pp. 812–820 (2001).
4. D. Hofstetter, M. Beck, J. Faist, M. Naegele and M.W. Sigrist, "Photoacoustic spectroscopy with quantum cascade distributed-feedback lasers," *Optics Letters*, vol. 26, pp. 887–889 (2001).

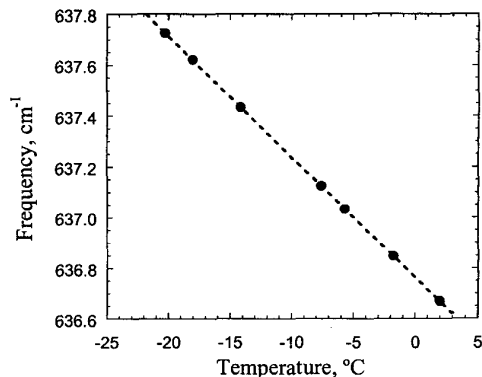
**CFA2** 8:15 am

**Sub-doppler NICE-OHMS Spectroscopy  
at 8.5 Microns Using a Quantum  
Cascade Laser**

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Quantum cascade lasers (QCLs)<sup>1,2</sup> have greatly increased the availability of tunable lasers emitting in the mid- to long-wave infrared spectral regions. Due to the abundance of spectroscopic signatures from compounds used in a vast array of chemical processes, high-resolution spectroscopy<sup>3,4</sup> and chemical sensing<sup>5</sup> are very important and topical applications for such laser sources. QCLs are intrinsically low-noise due to their near-zero alpha parameter,<sup>1,6,7,8</sup> have good control characteristics and lend themselves well to laser stabilization.<sup>9</sup> This makes them ideal for high performance cavity-based chemical detection schemes such as Noise-Immune Cavity-En-



**CFA1** Fig. 1. Long-wavelength QC-DFB laser temperature tuning. The dots indicate the positions of observed CO<sub>2</sub> and H<sub>2</sub>O absorption lines.

hanced Optical Heterodyne Molecular Spectroscopy (NICE-OHMS).<sup>10-14</sup>

NICE-OHMS is an extremely sensitive technique that has demonstrated a noise-equivalent integrated absorption of  $5.2 \times 10^{-13}$ , comparable only to that of mass spectrometry but with a selectivity found only in spectroscopy. This is understandably of much interest in the world of chemical sensing. A non-trivial technique however, it has only been performed in a few laboratories around the world. In this paper we present the detection of the Lamb-dip in the transition  $[0,0,0 \geq 0,2,0 (v_1, v_2, v_3), J = 6 \geq 7]$  of nitrous oxide (N<sub>2</sub>O) at 1174.05 wavenumbers, using both direct cavity-enhanced absorption, and NICE-OHMS.

A QCL operating at 8.5 microns was tightly locked using the Pound-Drever-Hall technique<sup>15-17</sup> to a 26.8 cm long optical cavity formed by two mirrors mounted inside a gas cell. This cell was evacuated, flushed, and then loaded with various pressures of N<sub>2</sub>O in the absence of buffer gases. The cavity length was scanned using a piezo element mounted on one of the mirrors. The cavity transmission was observed using a commercial 100 micron diameter mercury-cadmium-telluride (MCT) detector for a range of gas pressures. The Lamb-dip seen in figure 1.a) was observed at an optimum N<sub>2</sub>O pressure of 15 milliTorr. This feature, not yet well optimized for laser power and other parameters, shows significant homogeneous broadening. Nevertheless, while others have used QCLs to observe Lamb-dips in other molecules,<sup>3,18</sup> this is to our knowledge the first demonstration of a cavity-enhanced Lamb-dip measurement using a QCL.

The above-described system was then modified for the first demonstration of NICE-OHMS signal recovery in the mid-IR region. With the same operating conditions, additional sidebands at 559 MHz were added to the QCL via its injection current. This frequency matches the free-spectral range of the optical cavity and so these sidebands pass through the cavity equally with the optical carrier, allowing them to beat on the MCT transmission detector. Noise caused by errors in the lock between the laser and cavity causes identical effects on both the carrier and sidebands, and thus yields no contribution to the demodulated signal from the cavity transmission detector. Figure 1.b) shows this demodulated sig-