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Cavity ringdown spectroscopy of NO with a cw single frequency quantum cascade laser.

A.A. Kosterev, A.L. Malinovsky, F.K. Tittel, C. Gmachl,* F. Capasso,* D.L. Sivco,* J.N. Baillargeon,* A.L. Hutchinson,* A.Y. Cho,* Rice Quantum Institute, Rice University, Houston, TX 77251-1892; email: akoster@rice.edu; *Bell Laboratories, Lucent Technologies, 600 Mountain Avenue, Murray Hill, NJ 07974

Cavity ringdown spectroscopy (CRS) enables the measurements of very weak absorption introduced into a high-finesse optical cavity. This method is based on measuring a decay time of the cavity mode. An absorption coefficient of the intracavity medium per unit length is given by the expression $\alpha = (1/\tau_0 - 1/\tau_f)/c$, where τ_0 and τ_f are the ringdown times of the empty and filled cavity, respectively. Such an approach can lead to

an effective pathlength of several kilometers in a small volume, and the results are not influenced by laser power fluctuations. The first CRS experiments were reported in 1988,¹ but the application of CRS to the mid-IR spectral region only became possible recently due to the progress in low-loss high-reflectivity mirror technology.

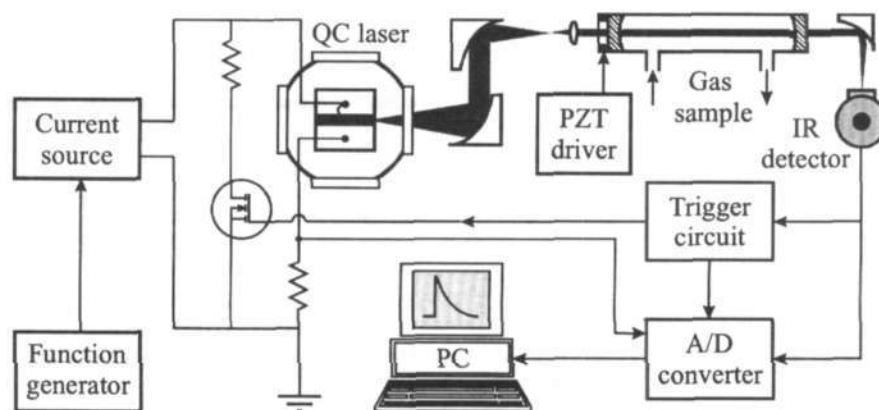
In this contribution we shall report the ultrasensitive detection of NO by its fundamental absorption at 5.2 μm with CRS technique. A continuous wave quantum cascade distributed feedback (QC-DFB) laser was used as the spectroscopic source. This kind of laser is ideally suited for CRS because it exhibits high power, stable single frequency operation, low sensitivity to optical feedback and intrinsically narrow linewidth.

The first demonstration of mid-IR ($\lambda = 8.5 \mu\text{m}$) CRS with a QC-DFB laser was reported in Ref.² The authors used an acousto-optic modulator for interrupting the laser radiation after the cavity buildup. This element adds complexity to the sensor architecture. In our experimental setup shown schematically in Fig. 1, the laser emission was interrupted by rapidly decreasing the laser current to a below-threshold value. The IR detector signal was acquired using a fast A/D board capable of up to a 50 MS/s conversion rate. The same comparator circuit triggered both the current control MOSFET and the data acquisition board when a sufficiently large buildup of radiation in the cavity occurred. The cavity ringdown time was measured by fitting the detector signal with an exponential function. The decay time measurements were repeated while the laser frequency was swept near the absorption line of interest.

With the available mirrors and the best alignment of the evacuated cavity, we found $\tau_0 = 3.48 \mu\text{s}$ (equivalent to an effective pathlength of 1,040 m) and a relative standard deviation of a single-measurement ringdown time, $\sigma/(\tau_0) = 2.24 \times 10^{-3}$. This yields a potential single-point sensitivity of $\alpha = 2 \times 10^{-8} \text{ l/cm}$, which can be further improved by means of averaging. We have detected NO in a pure nitrogen mixture (Fig. 2), in human exhaled air samples at the several ppb level (unresolved lines at 1921.599 and 1921.601 cm^{-1}), and also detected a weak CO_2 absorption at 1921.641 cm^{-1} in ambient air. The actual sensitivity to absorption lines in room and exhaled air samples (~ 60 Torr in the cavity) was reduced by the tails of absorption lines of the highly abundant species such as H_2O and CO_2 , which create a broadband background and decrease the effective pathlength. A comparison of the CRS technique to a multipass cell and cavity-enhanced spectroscopy will be made and potential future developments will be discussed.

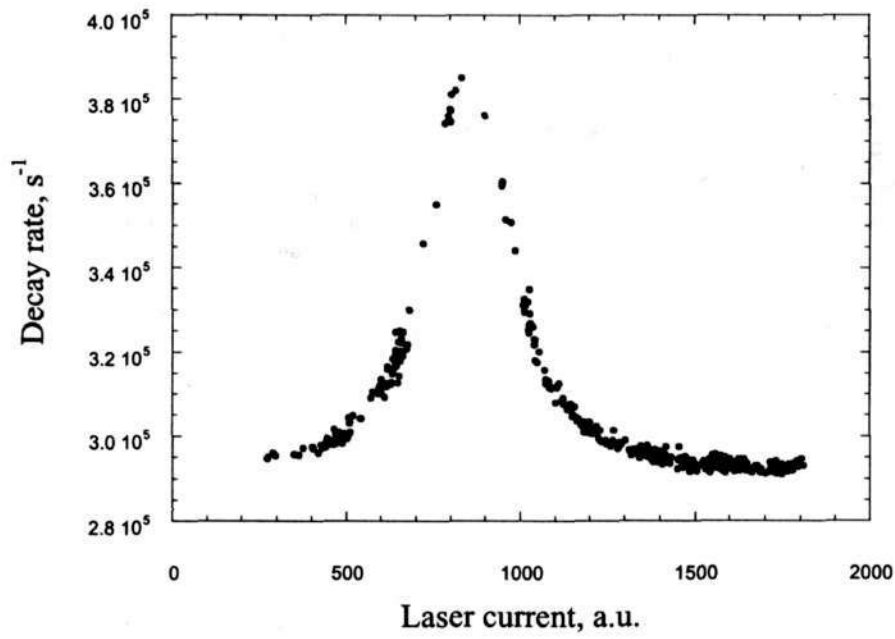
References

1. A. O'Keefe and D.A.G. Deacon, "Cavity ringdown optical spectrometer for absorption measurements using pulsed laser sources", *Rev. Sci. Instrum.* **59**, 2544-2551 (1998)
2. B.A. Paldus, C.C. Harb, T.G. Spence, R.N. Zare, C. Gmachl, F. Capasso, D.L. Sivco, J.N. Baillargeon, A.L. Hutchinson, and A.Y. Cho, "Cavity ringdown spectroscopy using mid-infrared quantum-cascade lasers", *Opt. Lett.* **25**, 666-668 (2000)



CThT2 Fig. 1. A CRS based NO sensor. The QC-DFB laser is mounted in a liquid nitrogen Dewar.

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CThT2 Fig. 2. NO Absorption at 1921.60 cm^{-1} measured in a mixture with N_2 . NO concentration is ~ 700 ppb. Each point corresponds to a single ringdown time measurement.