

Linear cavity optical-feedback cavity-enhanced absorption spectroscopy with a quantum cascade laser

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A cw distributed feedback quantum cascade laser (DFB-QCL) coupled to a two-mirror linear optical cavity has been used to successfully demonstrate optical-feedback cavity-enhanced absorption spectroscopy (OF-CEAS) at 5.5 μm . The noise-equivalent absorption coefficient, α_{min} , was $2.4 \times 10^{-8} \text{ cm}^{-1}$ for 1 s averaging, limited by etalon-fringing. The temporal stability of the instrument allows NO detection down to 5 ppb in 2 s. © 2013 Optical Society of America

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Optical-feedback cavity-enhanced absorption spectroscopy (OF-CEAS) is a resonant cavity technique, where optical-feedback from a cavity injection seeds a semiconductor laser. With an appropriate feedback rate and optical geometry [1,2], as the laser's frequency is continuously tuned by varying the injection current it locks to successive axial cavity mode frequencies [3]. Accordingly, the temporal cavity transmission consists of an envelope of peaks, whose frequency spacing corresponds to the free spectral range (FSR) of the optical cavity. When an absorber is introduced to the cavity, this yields a cavity-enhanced absorption spectrum with a well-defined frequency scale, making OF-CEAS well suited to sensitive detection of pressure-broadened trace gases, as discussed in [3,4].

Distributed feedback quantum cascade lasers (DFB-QCLs) are compact, single-mode, and tuneable mid-IR sources. This spectral region coincides with the intense fundamental vibration bands of many small molecules, and hence these sources are the focus of intensive research efforts into gas sensors capable of selective and sensitive measurements. Recently, three OF-CEAS instruments based on QCLs have been reported [4–6]. All followed the form of the first OF-CEAS experiment [3], which used a near-IR diode laser and a three-mirror V-shaped optical cavity to spatially decouple the optical-feedback beam from other reflections off the first cavity mirror. This Letter presents a novel QCL OF-CEAS instrument with a simpler arrangement. A two-mirror linear cavity is shown to provide viable OF-CEAS performance. With this arrangement, the field directly reflected from the mirror coating of the first cavity mirror is not found to affect the successful operation of the optical feedback locking scheme, developed by Morville *et al.* [3] for a diode laser. Given the power of the QCL, the unmodified feedback rate resulted in a frequency locking range smaller than one cavity FSR, so no feedback attenuation optics were required. Compared to a V-cavity, this simpler configuration requires one less cavity mirror and

thereby reduces cavity losses. Additionally, there is no alternation in the power buildup of odd and even modes, as all modes have a node at each mirror [3].

Figure 1 depicts the experimental setup. The DFB-QCL (Alpes Lasers) had a tuning range from 1818 to 1826 cm^{-1} and a maximum power output of 3 mW at an operating temperature of -10°C . The laser chip was mounted in a previously described custom mechanical housing [7], and driven by temperature and current controllers made in-house. The QCL beam was collimated by an aspheric lens (4 mm focal length, 0.56 NA), and a copropagating HeNe laser beam was overlapped to aid alignment. An intermediate beam waist was formed by an off-axis parabolic mirror (OAPM), where an iris was positioned to act as a spatial filter. Imaging this beam waist with a mid-IR microbolometer array (DataRay, WinCamD FIR), and fitting the image with a two-dimensional Gaussian profile, returned vertical and horizontal beam waists of 0.20 and $0.15 \pm 0.09 \text{ mm}$, respectively. This beam waist was mode matched to that of the cavity by an uncoated plano-convex CaF_2 lens with a focal length of 150 mm. The optical cavity was formed by two highly reflective mirrors

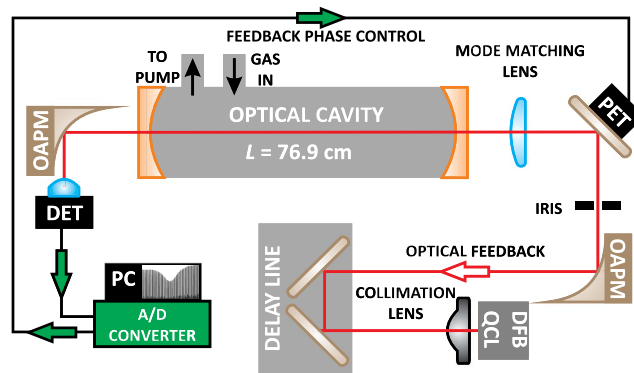


Fig. 1. Schematic of the setup. OAPMs are off-axis parabolic mirrors; PET is the piezoelectric transducer; ADC is the analog-to-digital converter; and DET is the detector.

(Los Gatos Research, 0.5 m radius of curvature), whose dielectric surfaces were deposited on 1 in. diameter ZnSe substrates. The cavity length, L , was chosen to be 76.9 cm to give a FSR (and therefore a spectral sampling resolution) of 195 MHz.

An OAPM redirected the beam exiting the cavity on to a thermoelectrically cooled HgCdTe detector (VIGO, PVI-2TE-6), with an effective active surface area of 1 mm². The cavity mirrors formed the windows of the cell and were held in bellows assemblies connected by a 1 in. diameter stainless steel tube. A rotary pump evacuated the cell, and analyte and inert broadening gases were added via 1/4 in. stainless steel tubing.

Following an analysis similar to that of Laurent *et al.* [2], if the distance between the QCL output facet and the first cavity mirror is equal to an integer multiple of L , then the phase of the emitted and feedback fields match at the QCL output facet. This causes the shape of the peaks in the cavity transmission to be symmetrical and encompass a single envelope within a laser scan. A delay line enabled coarse adjustment of this parameter. Fine phase adjustments on a μm scale were controlled actively. For this purpose, the turning mirror immediately before the cavity was mounted on a piezoelectric transducer (PET), and the cavity output signal was acquired by a digitizer card (National Instruments, PCI 6010). A LabVIEW program analyzed the phase-dependent peak asymmetry, from which an error signal was derived. This was transformed into a feedback voltage supplied to the PET with a response time equal to the duration of a single scan (0.2 s).

Linear cavity OF-CEAS spectra of the atmospherically important species N₂O and NO were recorded. The QCL was scanned over transitions of interest by applying a symmetrical triangular current ramp of 20 mA with a frequency of 4.95 Hz. Data were acquired by the digitizer card, and Fig. 2(a) shows one such cavity output signal exhibiting absorption by NO. Over the 100 ms of the up-ramp, the QCL locks to 60 successive cavity modes leading to power modulation of the detected signal. The peak-power increases as the QCL power increases over the course of the up-ramp. The magnified section in Fig. 2(b) demonstrates successful feedback locking to three suc-

cessive modes. The locking time, estimated using the mode FWHM is 120 μs , much longer than the expected ring-down time.

Frequency-calibrated transmission spectra were obtained from the cavity output signal by identification of the transmission maxima, $S(\nu)$, the positions of which were mapped on to a relative frequency scale, ν , defined by the FSR of the cavity. A global fitting routine was used based on the approach described in [8]. Line positions and other parameters used in the fitting routine came from the HITRAN database [9]. Molecular absorption lineshapes were modeled as Voigt profiles, while a linear baseline ($a + b\nu$) accounted for constant transmission losses and the power modulation of the QCL during frequency tuning. Laser power attenuation, due to intracavity absorption reduced optical feedback [4], was found to be negligible ($\ll 1\%$), so no correction was made for this effect. The relationship between the absorption coefficient, $\alpha(\nu)$, the mirror reflectivity R , and the ratio of the transmission of a linear cavity with and without absorbing losses is given by Eq. (1) and was used for the model:

$$\frac{(a + b\nu)}{S(\nu)} = \left[\frac{\alpha(\nu)L}{(1 - R)} + 1 \right]^2. \quad (1)$$

Equation (1) is an approximation in the limit of small $\alpha(\nu)$. In this work, the maximum values of $\alpha(\nu)$ were on the order of $1 \times 10^{-5} \text{ cm}^{-1}$, and comparison of Eq. (1) with the exact expression results in an overestimation of 0.1%. Note that Eq. (1) is the same as that derived for a symmetrical V cavity [10], which becomes a linear cavity in the limit where the angle between the arms tends to zero. The fit returned the ratio $\alpha(\nu)/(1 - R)$, the two coefficients (a and b) describing the linear baseline variation, and the 1σ uncertainty on these parameters.

Figure 3 shows transmission spectra for (a) N₂O and (b) NO, from single scans recorded in 0.2 s. The N₂O absorption comprised three unresolved transitions with a combined intensity of $2.3 \times 10^{-24} \text{ cm}^{-1}/(\text{molecules cm}^{-2})$ at 296 K. To obtain a pressure broadened absorption lineshape covering around 10 cavity FSRs, a total pressure of 0.28 atm of a $1.8 \pm 0.1\%$ N₂O in N₂ gas mix (CK Gas) was used. This yielded a mirror reflectivity, R , of $99.931 \pm 0.006\%$, where the error corresponds to estimates of the uncertainties in the mixture calibration, pressure measurement and data fitting, and is consistent with the manufacturer's specified value. From this spectrum, the minimum detectable signal can be defined as one standard deviation of the residual trace. Using Eq. (1), the corresponding noise-equivalent absorption coefficient, α_{min} , was $2.7 \times 10^{-8} \text{ cm}^{-1}$ for 1 s averaging. Figure 3(b) shows a spectrum of the unresolved Λ -doublet of the $P(14.5)$ transition in the $\nu = 1 \leftarrow 0$ band of NO, which lies in the wing of an intense water absorption and has an combined intensity of $5.5 \times 10^{-20} \text{ cm}^{-1}/(\text{molecules cm}^{-2})$ at 296 K. The spectrum was recorded with trace NO broadened by N₂ to 0.19 atm. The α_{min} was $2.4 \times 10^{-8} \text{ cm}^{-1}$ (1 s), in close agreement with that obtained for N₂O. The sensitivity is currently limited by etalon fringes on the transmission signal caused by reflections between the cavity output mirror

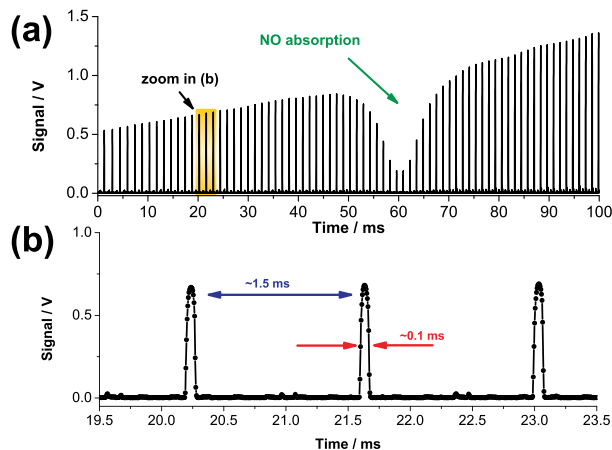


Fig. 2. Raw cavity transmission signal acquired for (a) one up-ramp of the QCL and (b) a magnification of this signal around 20 ms.

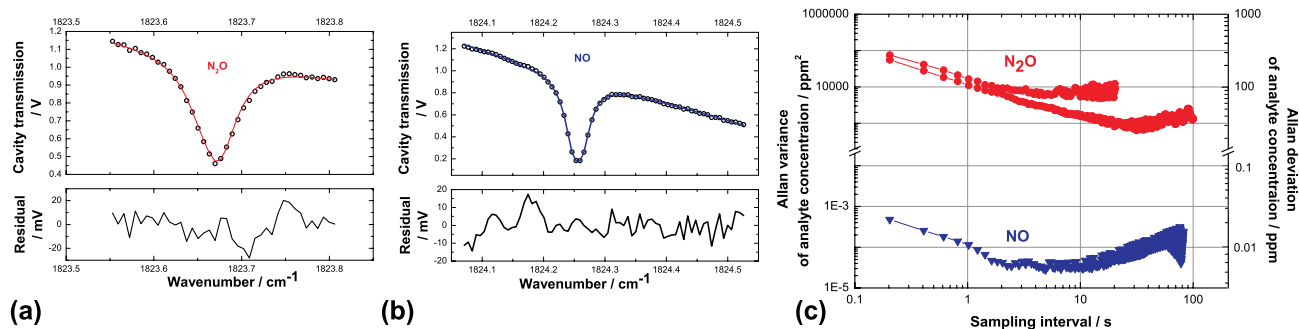


Fig. 3. (a) N₂O and (b) NO QCL OF-CEAS transmission spectra, showing experimental data (dots), fitted models (solid lines), and residuals (lower plots); (c) log-log Allan variance (left) and deviation (right) plots for returned concentration of N₂O (red circles) and NO (blue triangles).

and the immersion lens on the detector. Calculations indicate that the sensitivity of the present system could be increased to give a $\alpha_{\min}$ of $5.1 \times 10^{-9} \text{ cm}^{-1}$ in 1 s, corresponding to a system limited by the bit noise from the acquisition card (150 μV). The current sensitivity of this proof of principle device is two orders of magnitude lower than for state-of-the-art QCL OF-CEAS [6], based on a V cavity. While the development of the linear cavity QCL OF-CEAS instrument to a similar level is far from trivial, it is, in principle, possible as the major limiting factors are not related to cavity geometry.

The temporal stability was also investigated. Allan variance calculations were carried out on concentration time series, with a total duration of 200 s derived from 1000 spectra of the same analyte gas mixture sampled at 0.2 s intervals. Figure 3 shows log-log plots of the Allan variance (left) and deviation (right) for monitoring N₂O and NO. The typical optimal averaging period was 2 s as shown for the top Allan plot for N₂O (red circles) and NO (blue triangles). Temperature variations are the most likely limitation and efforts to mitigate these effects are on-going. On occasion, the system was stable enough to average for 35 s, evidenced by the lower Allan plot for N₂O (red circles). The Allan variance plots all have initial gradients close to -1 indicating white noise behavior. The best minimum detectable concentrations for the linear QCL OF-CEAS instrument are 32 ppm for N₂O (35 s) and 5 ppb for NO (2 s). To conclude, linear cavity QCL OF-CEAS in a relatively simple arrangement has been demonstrated

to be relevant to trace gas detection. The detection limit obtained for NO already indicates a potential application for monitoring in urban environments [11].

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