Opto-Electronic Advances

Supplementary information

November 2024, Vol. 7, No. 11

DOI: 10.29026/oea.2024.240077

Agile cavity ringdown spectroscopy enabled by moderate optical feedback to a quantum cascade laser

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Supplementary information for this paper is available at https://doi.org/10.29026/oea.2024.240077

Section 1. Laser linewidth measurement

Using the experimental setup shown in Fig. 2(a), we measured the converted intensity noise by a broad bandwidth (800 MHz) photodetector, which was analyzed by an electrical spectrum analyzer. Figure S1 shows the representative results of frequency noise power spectral density (FNPSD) measured in the span of 5 kHz to 500 MHz. The total linewidth of the QCL at different feedback rates can be obtained with the help of a β-separation line, above which the linewidth is determined by the frequency noise[S1]. It is clear that with the increase of feedback rate, the frequency noise spectrum first moves down and intersects the β-separation line at lower frequencies, i.e., narrower laser linewidth. Further increasing the feedback rate, the QCL maintains a narrow linewidth until the multi-mode emission is observed at −11.6 dB. The intensity noise of the QCL was also measured, which is always much smaller than the frequency noise at both the free-running and optical-feedback states. This indicates the intensity noise does not limit the frequency noise measurement and the sensitivity of the frequency discriminator is high enough.

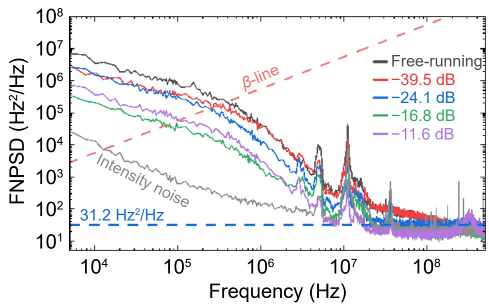

Fig. S1 | Frequency noise power spectral density of the QCL at various feedback ratios. Red-dashed line: β-separation line; blue-dashed line: white noise level of around 31.2 Hz²/Hz.

Section 2. Characterization of ringdown time, mirror reflectivity and gas concentration

In continuous-wave CRDS shown in Fig. S2, the incident low-intensity light builds up in the optical cavity when the light resonates with one of the longitudinal modes of the cavity. The incident radiation is then interrupted to generate the ringdown event using an optical modulator. The cavity-transmitted light intensity (I) decays with time (t) and can be determined by[S2]:

$$I(t) = I_0 e^{\left[-\frac{tc}{L}(1-R+\alpha_\nu L)\right]}\ , \tag{S1}$$

where I_0 is the light intensity observed when the optical build-up process is interrupted, c is the speed of light, L is the inter-mirror distance, R is the mirror reflectivity, and α_ν [cm⁻¹] is the absorption coefficient of the gas at the optical frequency ν. The ringdown time (τ) is defined as the time taken for the light intensity to fall to 1/e of the initial intensity I_0. When the optical cavity is empty (no gas absorption, $\alpha = 0$), the ringdown time τ_0 is determined by:

$$\tau_0 = \frac{L}{c(1-R)}\ . \tag{S2}$$

Hence, the reflectivity of the cavity mirror can be obtained from the measured ringdown time of the empty cavity. When gas absorption exists in the optical cavity, the ringdown time τ_1 can be expressed as:

$$\tau_1 = \frac{L}{c(1-R+\alpha_\nu L)}\ . \tag{S3}$$

Based on Eq. (S2) and Eq. (S3), the absorption coefficient of the gas can be determined from Eq. (1) shown in the main text. Hence, the CRDS-determined volume fraction X of the target species can be obtained based on Beer's law[S2]:

$$X = \frac{\alpha_\nu}{S(T)P\phi_\nu}\ , \tag{S4}$$

where $S(T)$ [cm⁻² atm⁻¹] is the line-strength, which is provided in the HITRAN database[S3]; P [atm] is the total gas pressure, and ϕ_ν [cm] is the line-shape function.

Fig. S2 | Schematic of cavity ringdown spectroscopy. *R*, reflectivity of the cavity mirror; *L*, physical distance between two cavity mirrors; *I*, intensity of the cavity-transmitted light detected by photodetector; I_0, cavity-transmitted light intensity at the moment when the incident light is cut off; τ_0, ringdown time of the cavity without gas absorption; τ_1, ringdown time of the cavity with gas absorption.

Section 3. Tuning characteristics of the continuous-wave DFB-QCL

The emission power and wavelength tuning characteristics of the QCL are illustrated in Fig. S3. The laser wavelength, characterized by an optical spectrum analyzer, was varied by adjusting the injection current with a step size of 10 mA at each laser temperature. The simulated absorption spectrum of N_2O is added in the same plot, along with the main interference species including CO_2, H_2O and CO in the air. The N_2O absorption line centered at 2207.62 cm^{-1} has minimal spectral interference, which is selected as the target line in the study.

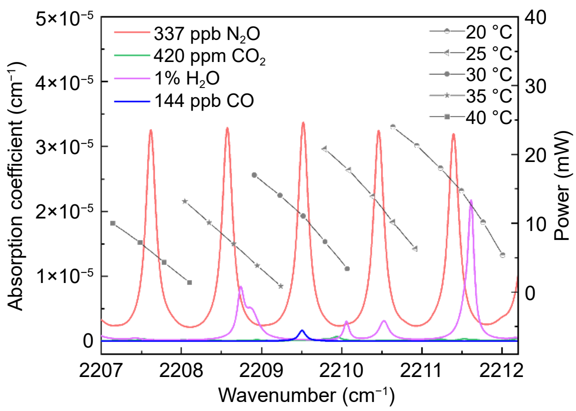

Fig. S3 | Emission power and wavelength tuning characteristics of the QCL. The absorption spectrum of a gas mixture including 337 ppb N_2O, 144 ppb CO, 420 ppm CO_2, and 1% H_2O is made in the same plot.

Section 4. Impurity in commercial N_2 gas cylinder

Commercially available pure N_2 is normally produced by the cryogenic fractional distillation of liquefied air, separation of gaseous air by adsorption, or permeation through membranes. Trace amounts of gas components such as N_2O and CO in the air may still exist as impurities in the N_2 gas cylinder. In our experiments with high precision and high sensitivity, it is essential to verify the gas concentration and interference species in the N_2 gas cylinder before using it as the balance gas, which can be investigated using our CRDS system. We initially evaluated the gas provided from a pure N_2 cylinder (N5.0, Linde HKO Limited) using the high-finesse (~52000) cavity, but the gas absorption was so strong that the measurement was beyond the linear dynamic range of the spectrometer. Instead, another pair of cavity mirrors with a lower reflectivity (99.975%) were used to quantify the impurity. Figure S4 demonstrates the CRDS measurement results and the spectral simulation based on the HITRAN database[S3], indicating the existence of 74 ppb N_2O and 85 ppb CO in the N_2 gas cylinder. However, we did not observe any impurity in the helium gas cylinder based on the CRDS measurement over the same spectral range.

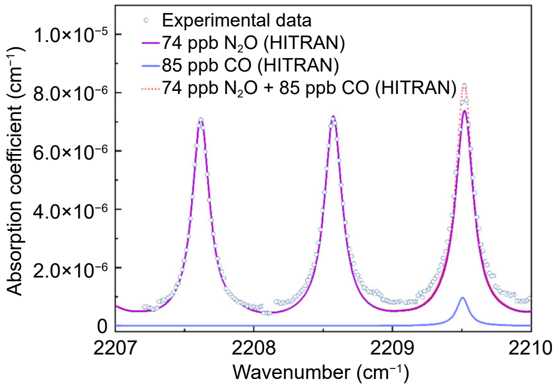

Fig. S4 | CRDS measurement of gases in a pure N_2 cylinder (N5.0, Linde HKO Limited).

References

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S3. Gordon IE, Rothman LS, Hargreaves RJ et al. The HITRAN2020 molecular spectroscopic database. *J Quant Spectrosc Radiat Transfer* **277**, 107949 (2022).