Supplementary Information for

Doubly resonant sub-ppt photoacoustic gas detection with eight decades dynamic range

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**Supplementary Note 1: Characterization of mirror reflectivity**

To characterize the mirror reflectivity using ring-down measurement, a longer optical resonator of 600 mm is designed with two high-reflectivity mirrors which have a radius of curvature (ROC) of 1500 mm. Note that all the mirrors used in this study are fabricated in the same coating process and have the same reflectivity. An optical switch with 85 ns fall time is used to swiftly generate the ring-down event, which is monitored by a high-bandwidth (150 MHz) photodetector. The ring-down signal, illustrated in Supplementary Fig. 1, is acquired by a high-speed data acquisition card with a 100 MHz sample rate and a 16-bit analog-to-digital converter.



**Supplementary Figure 1.** Characterization of mirror reflectivity using cavity ring-down measurement. The averaged ring-down time (average of 84 ring-down events) is determined to be 2.596 μs, corresponding to a reflectivity of 99.923%.

**Supplementary Note 2: Optical coupling efficiency**

It is important to couple as much light as possible into the optical resonator. To evaluate the coupling efficiency, the reflected light from the optical resonator is detected by a photodetector. By scanning the resonator length using a piezo transducer (PZT), the fundamental resonator mode shown in Supplementary Fig. 2 is used to investigate the coupling efficiency. Based on the ground voltage (4.451 V) and valley voltage (0.719 V), a coupling efficiency of 84% is determined in this work.



**Supplementary Figure 2.** Measured intensity of the reflected light from the optical resonator. A coupling efficiency of 84% is determined from the ground voltage (4.451 V) and valley voltage (0.719 V).

**Supplementary Note 3: Sensor performance with a microphone**



**Supplementary Figure 3.** Principle of the opto-acoustic resonance for PAS using a microphone. ASW: acoustic standing wave; OSW: optical standing wave. The amplitude of the OSW is modulated at the resonant frequency of the acoustic resonator. Two buffers are designed on both sides to form the acoustic standing wave with an antinode located at the center of the resonator.

The principle of the opto-acoustic resonance for PAS with a microphone as the acoustic transducer is illustrated in Supplementary Fig. 3. The cylindrical PAS cell is designed to have a total length of 70 mm, which is placed inside an optical resonator with a length of 80 mm. In particular, the acoustic resonator has a length of 35 mm and an inner diameter of 3 mm, and the two buffers on both sides of the acoustic resonator have a length of 17.5 mm and an inner diameter of 12 mm. The photoacoustic cell is characterized to have a resonant frequency of 3.5 kHz and a Q-factor of 25 at 760 Torr. An electret microphone is mounted at the center of the photoacoustic cell for acoustic detection, which corresponds to the antinode of the standing acoustic wave. The representative PAS-1f signals measured at different C2H2 concentrations (0.1-10 ppm) with an incident power of 12 mW are shown in Supplementary Fig. 4.



**Supplementary Figure 4.** Representative PAS-1f signals of C2H2 measured at different concentrations along with the spectral fitting curves at 760 Torr. The target C2H2 peak is observed at 1531.59 nm. The detection bandwidth of the lock-in amplifier is 1 Hz.

The linear response of the sensor is studied by measuring C2H2 of different concentrations shown in Supplementary Fig. 5. The sensor shows an excellent linear response from 1 ppb to 30 ppm with an R-squire value of 0.999. The Allan–Werle deviation analysis is plotted in Supplementary Fig. 6 by measuring nitrogen for about 1.5 hours at 760 Torr. The detection bandwidth of the lock-in amplifier is 1 Hz. As a result, the sensor can achieve a noise equivalent concentration (NEC) of 1.75 ppt at an integration time of 180 s, corresponding to a noise equivalent absorption (NEA) of 1.9×10-12 cm-1. In this case, the linear dynamic range of the sensor is about seven orders of magnitude.



**Supplementary Figure 5.** Background-subtracted PAS-1f amplitude as a function of C2H2 concentration. The linear fitting yields an R-square value of 0.999 from 1 ppb to 30 ppm. The incident laser power is 12 mW and the detection bandwidth of the lock-in amplifier is 1 Hz.



**Supplementary Figure 6.** Allan–Werle deviation analysis for the microphone-based sensor. The laser wavelength is tuned to the peak of the C2H2 absorption line with the chamber filled with nitrogen. As a function of integration time, the analysis shows a NEC of 1.75 ppt at 180 s. The detection bandwidth is 1 Hz, the same as signal measurement.

**Supplementary Note 4: Continuous measurement**

The sensor is tested by continuously filling 100 ppb C2H2 into the gas chamber. The flow rate is varied (200, 300, and 400 mL/min) at 60 s and 120 s, respectively, during the continuous measurement. As shown in Supplementary Fig. 7, the sensor can work continuously without losing lock due to the robust locking between laser and optical resonator.



**Supplementary Figure 7.** Continuous monitoring of 100 ppb C2H2 at varied flow rates (200, 300, and 400 mL/min). The flow rate is changed at 60 s and 120 s, respectively, during the continuous measurement.