Supporting Information of Multiple Gas Detection by Cavity-enhanced Raman Spectroscopy with Sub-ppm Sensitivity

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A 24-hour CERS Measurement of H_2

To test the capability for long-term measurement of this CERS instrument, we carried out a continuous 24-hour measurement. The sample gas was pure hydrogen at the pressure of 20 Pa, and the vibrational band of H_2 near 4155 cm^{-1} was measured. The Raman scattering spectrum of H_2 was measured every minute with an exposure time of 10 s. A sample spectrum is shown in Fig.S1 (A), and spectra with different averaging times are shown in Figs.S1 (B)-(E). We can see the noise level decreases when the averaging time increases.

The input laser power was monitored before the cavity (I_{in}), and the laser power transmitted from the cavity (I_{out}) was also measured, which are shown in Figs.S2 (A) & (B). We can see the input laser power was rather stable with a relative drift within ±3%. However, the output laser decreased from 17 mW to about 8 mW in the first four hours and then remained in the range of 6 - 10 mW in the following 20 hours. It might come from the drift of environmental conditions, such as room temperature drift and mechanical relaxations. We can see a periodic fluctuation in the output laser power, which is consistent with the fluctuation of the room temperature. The height (H) of the Raman peak $Q_1(1)$ at 4155 cm^{-1} of each spectrum was also recorded, as shown in Fig.S2 (C), which drifted with the laser power transmitted from the cavity. We normalized the Raman peak height with I_{out}, and the results are shown in Fig.S2 (D). We can see that after normalization, the Raman signal is quite stable, which satisfies quantitative measurements. The Allan standard deviation of the normalized Raman peak height (H_{norm}) is shown in Fig.S2 (E). The deviation decreases to about 5% of the peak height at an average time of 20 min. Meanwhile, the noise level of the Raman spectrum decreases as the number of averages increases. As shown in Fig.S2 (E), the noise level decreases to about 0.033 after an average time of about 1 hour. Therefore, we can effectively improve the detection sensitivity by increasing the averaging time.

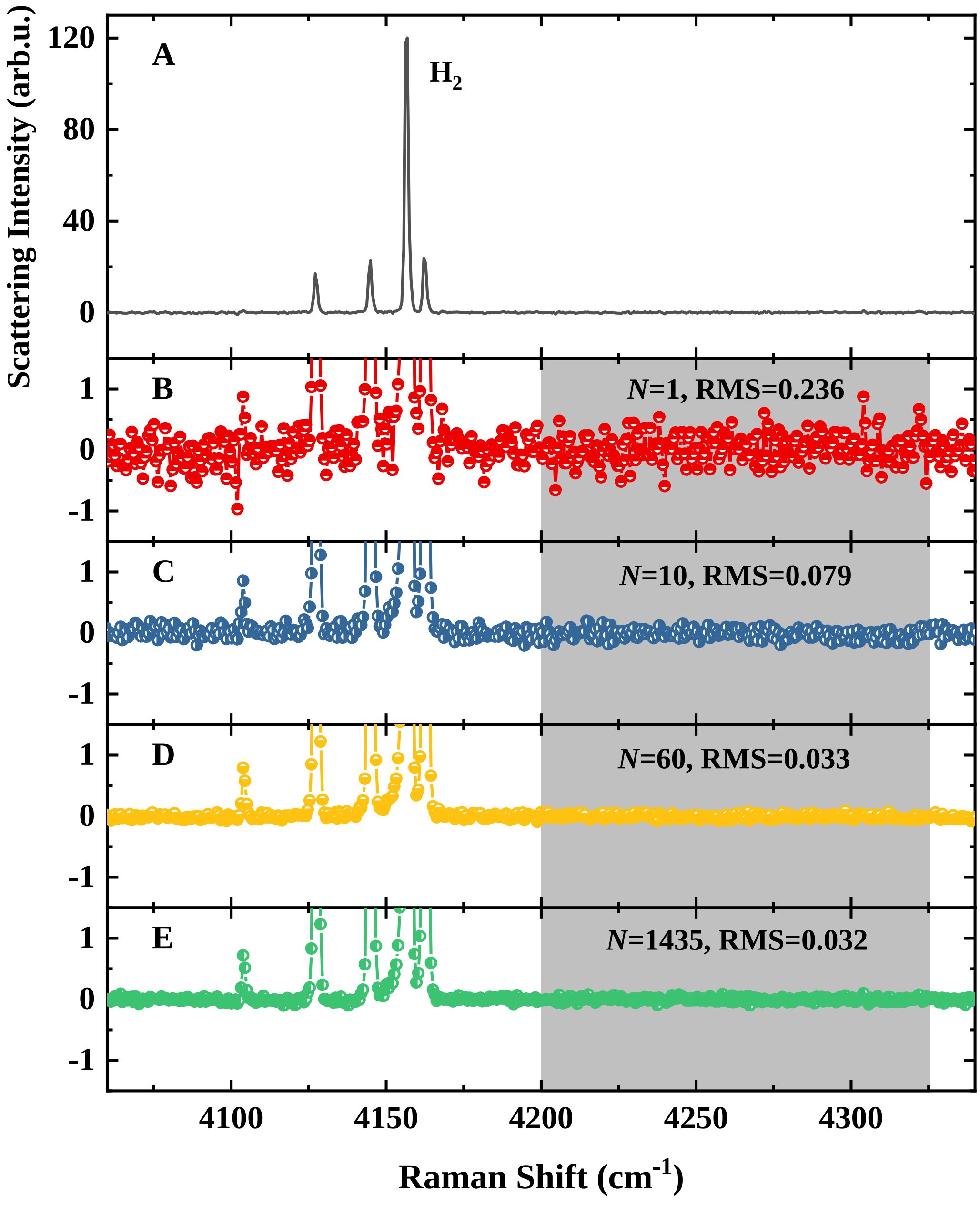

Fig. S1: A continuous measurement of the Raman spectrum of pure hydrogen near 4155 cm^{-1}. Spectra were recorded every minute at the pressure of 20 Pa with an exposure time of 10 s. The zoom-in plot for averaged spectra of 1, 10, 60, and 1435 scans are shown in (B), (C), (D), and (E), respectively.

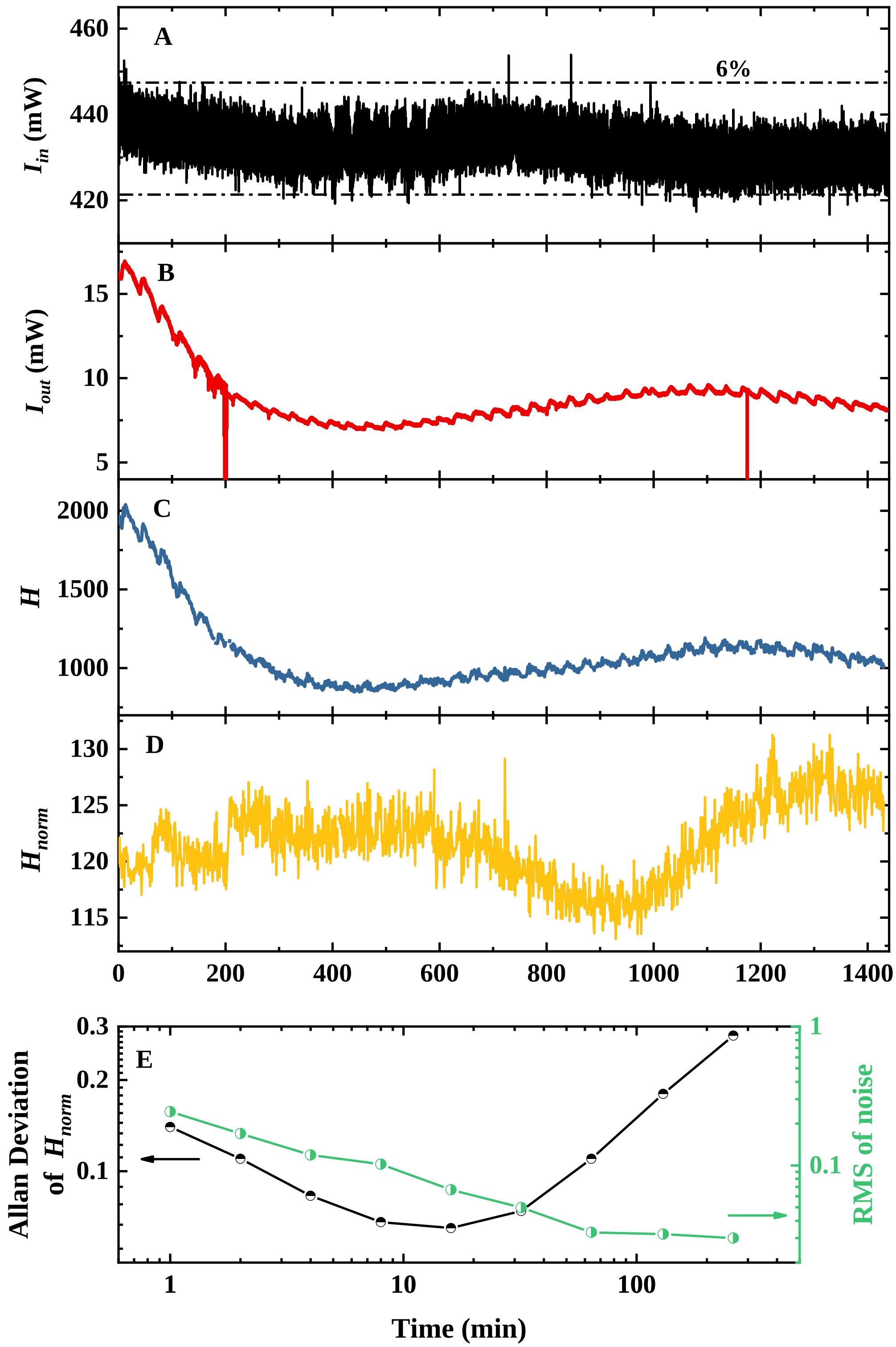

Fig. S2: A 24-hour CERS measurement of: (A) the input laser power coupled to the cavity, I_{in}; (B) the laser power transmitted from the cavity, I_{out}; (C) the height of the Raman peak of H_2 at 4155 cm^{-1}, H; (D) the normalized height of the Raman peak, H/I_{out}; and (E) the Allan deviation of H_{norm} (black) and the noise level (green) of the Raman spectrum versus the averaging time.