Recent advances in cavity ring-down spectroscopy with high-bandwidth locking and frequency scanning techniques

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Cavity ring-down spectroscopy (CRDS) is a powerful technique, providing high accuracy, sensitivity, spectral resolution, and specificity in a compact spectrometer setup. However, certain limitations, notably duty cycle and technical noise, have precluded realization of the technique's full potential. Here we survey efforts in our laboratories to reach and exploit fundamental measurement limits of CRDS. These studies include the implementation of high-bandwidth laser locking techniques that enable line parameter measurements of species such as O_2, CO, and CO_2 with relative uncertainties at the ≈0.3% level, spectra with signal-to-noise ratios exceeding 10^5:1 [1], and molecular line positions with kHz-level uncertainties based on optical frequency-comb-referenced spectra [2]. We also present frequency-agile, rapid scanning cavity ring-down spectroscopy (FARS-CRDS) [3], which uses microwave-tuned, fiber-waveguide phase modulators to rapidly tune the probe laser wavelength. This new method enables near-shot-noise-limited decay-time statistics [4], ring-down signal acquisition rates of 10 kHz, tunability – without detuning of the laser – over an optical frequency range of 70 GHz, and a noise-equivalent absorption coefficients of ≈10^{-12} cm^{-1} Hz$^{-1/2}$. Using comb-linked FARS-CRDS and a tunable diode laser, we have demonstrated spectral coverage of ≈2 THz with 10 kHz-level absolute frequency accuracy, and we have measured pressure-dependent shifting of cavity resonances caused by intra-cavity absorption. The FARS-CRDS technique can also yield cavity-enhanced absorption spectra in the frequency domain by measuring resonance line widths with an uncertainty of ≈5 Hz.

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