Advances in rapidly swept cavity ringdown spectroscopy for gas sensing

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Abstract: A high-performance continuous-wave cavity ringdown spectrometer, based on either a rapidly swept cavity or a widely tunable swept-frequency laser, has been developed as a compact versatile instrument for real-time efficient spectroscopic sensing of multiple species. We discuss various innovations, performance of the system, and several applications.

Cavity ringdown spectroscopy (CRDS) is an extremely sensitive technique for absolute absorption measurement by determining the decay rate of light in a high-finesse cavity containing a sample of interest. Absorption of light by the sample inside the cavity causes the decay rate to increase linearly with respect to its absorption coefficient, enabling either the sample’s concentration or its absorption strength to be measured accurately. We have implemented continuous-wave (cw) CRDS by simply sweeping either the resonance frequencies of the cavity (via its length) [1] or the laser frequency [2,3]. Coherent interaction between the laser radiation and the cavity enables build-up and subsequent decay of laser radiation in the cavity, without the need of an optical switch or high laser-frequency stability. CRDS signals can be collected either in reflection, with highly sensitive optical heterodyne detection (OHD) in a single-ended transmitter-receiver configuration or in transmission as in a more conventional way.

The layout of our rapidly swept cw-CRDS instrument [4] is depicted in Fig. 1. Typically, we work at optical-telecommunications wavelengths (1.5–1.6 μm) to take advantage of low-cost, monolithic, miniature external-cavity tunable diode lasers, as well as fibre-optical components and networks for remote, distributed-cavity sensors. We use optical-fiber and photonic components to direct light so that the ringdown cavity module (right-hand side in Fig. 1) can be located far away from the transmitter-receiver module (left-hand side in Fig 1) for remote applications via fiber-optical networks. A three-port optical circulator passes the laser radiation to the ringdown cavity and diverts the backward-propagating reflected and ringdown radiation to a photodetector (PD1) for OHD measurements. A second photodetector (PD2) provides an alternative, forward-propagating mode of CRDS measurement. The resulting OHD ringdown signals are processed by innovative analog circuits, for real-time generation of an analog voltage output that is proportional to the ringdown time [3]. This analog detection scheme registers a single data-point for each build-up and ringdown-decay event, without logging details of decay waveforms; this case demand on digitizer speed and reduces data-processing time.

This versatile system can be operated in three different configurations: with a single tunable laser [1], with a swept-frequency (SF) laser [2,3], and with multiple lasers set at characteristic on- and off-resonance wavelengths for gases of interest (such as CO, CO2, NH3, H2O, and various hydrocarbons) [1]. Fig. 2 shows a spectrum recorded by using a single tunable laser. The laser frequency is scanned in fine steps over a small wavelength range that spans absorption features of C2H2 and H2O. In this case, the ringdown cavity is dithered at high repetition rate and generates ringdown events at 4 kHz, limited by the time needed to allow each decay event to expire before the next one is generated. Baseline noise levels in our latest cw-CRDS experiments yield
a competitive noise-equivalent absorption limit of $-5 \times 10^{10}$ cm$^{-1}$ Hz$^{-1/2}$, independent of whatever molecules are to be detected. For instance, this allows sub-ppbv detection of acetylene ($\text{C}_2\text{H}_2$) in air [4].

Swept-cavity CRDS with several fixed-frequency cw diode lasers enables quasi-simultaneous specific multi-species detection [1]. For trace-level monitoring of particular gas-phase molecules, active scanning of the wavelength is usually not required. This reduces the required wavelength-tuning performance of laser sources. The multi-wavelength variant of our OHD cw-CRDS technique uses several fixed-wavelength TDLs, each pre-tuned to a characteristic on- or off-resonance feature of each gas molecule of interest, with their outputs combined into a single-mode optical fiber. Each TDL wavelength is coupled simultaneously into the ringdown cavity and attains CRDS resonance at a separate part of the cavity-sweep cycle. Alternatively, different wavelengths can be separated in space via dispersive optics or wavelength-selective filters, and monitored by individual photodetectors.

Use of a swept-frequency (SF) laser offers a great advantage in that survey spectra can be recorded rapidly over a broad wavelength range [2]. This is particularly helpful in achieving identification and real-time monitoring of gas-phase molecular species. During a wide range frequency sweep of the SF laser, successive occurring of build-up and ringdown decay events at cavity resonance frequencies register effectively the absorption spectrum of gas in the cavity at intervals defined precisely by the cavity’s free spectral range. A full absorption spectrum can be recorded in a single rapid sweep of the widely tunable laser frequency by innovative analog circuits [3]. One example is demonstrated in Fig. 3 for a gas mixture (2% CO$_2$ in N$_2$ with residual water vapor, $P = 125$ Torr; $T = 300$ K). This spectrum spans a 3.3-THz (110-cm$^{-1}$) range of SF laser frequencies, sampling $10^4$ SF cw-CRDS signal build-up and decay events within a time interval of 2 s, on a frequency grid defined exactly by the resonance frequencies of a fixed-length ringdown cavity.

Advantages of our form of rapidly swept cw-CRDS instrument are that it is intrinsically simple but sensitive. Moreover, our central transmitter-receiver cw-CRDS detection system is well suited to fibre-optical networks with CRDS sensor modules distributed around a site. Several diode lasers at pre-set wavelengths can be used to monitor multiple species simultaneously. Likewise, our rapidly scanned swept-frequency cw-CRDS method facilitates real-time, broad-spectrum sensing of multiple species.

In this presentation, we discuss various aspects of our compact, robust, high-performance instruments and prospects for real-time spectroscopic sensing of trace species.

References


