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# Broadband cavity ring-down Fourier-transform spectroscopy

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**Abstract.** We perform broadband cavity ring-down spectroscopy (CRDS) relying on the near-infrared frequency comb as the excitation source and a time-resolved mechanical Fourier transform spectrometer as detection device. The many decays corresponding to each spectral element are recorded simultaneously and sorted after Fourier transformation to yield the CRDS spectrum of CO in Ar contained in a 20'000-finesse cavity.

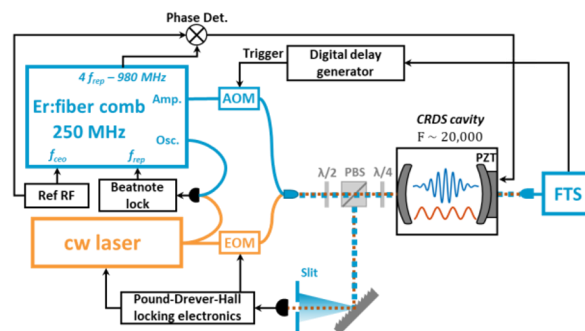
## 1 Introduction

Multiplexed cavity ring-down spectroscopy (CRDS) is challenging to achieve as it requires a detection scheme able to retrieve many undistorted exponential decays simultaneously [1]. We recently demonstrated Fourier transform CRDS (FT-CRDS) based on an optical frequency comb source and a time-resolved Fourier transform spectrometer (FTS) [2]. In this proof-of-concept experiment, the frequency comb was locked to a CRDS cavity using the Pound-Drever-Hall (PDH) technique, to ensure a quasi-continuous comb light transmission through the cavity. The decays were synchronized with the movement of the fast-scanning FTS, allowing to retrieve a 3D spectrum containing the untangled decays at each spectral element. The cavity was characterized by a moderate finesse of 2,000, far below standard reflectivity used for CRDS, and the comb mode absolute frequencies were not stabilized.

## 2 Experimental setup

Here, we revise the locking scheme, as depicted in Fig. 1: the offset frequency of the Erbium comb is stabilized to a reference radiofrequency while the repetition rate is phase-locked to a continuous wave laser emitting at 1550 nm, which in turn is locked to a cavity mode using the Pound-Drever-Hall technique. Absolute frequency stability is insured by actively stabilizing the repetition rate to a tunable reference acting on the piezo-actuator glued to one of the CRDS cavity mirrors. The cavity transmission is sent to the FTS where it propagates parallel to a frequency-stabilized HeNe laser whose interferogram is used for optical path difference (OPD) calibration. This reference is also used to trig the acousto-optic modulator switching off the comb light, allowing to retrieve cavity decays with a controlled OPD step. Fast Fourier transformation of the comb interferograms obtained at different delays after the triggers yields the

ring-down events in spectral domain. Finally, these exponential decays are fitted individually to retrieve the CRDS spectrum, which is directly proportional to the absorption coefficient of the analyte, without requiring knowledge of the cavity length or finesse. In addition, as the comb frequencies are stable in time, averaging of successive measurements is now possible.



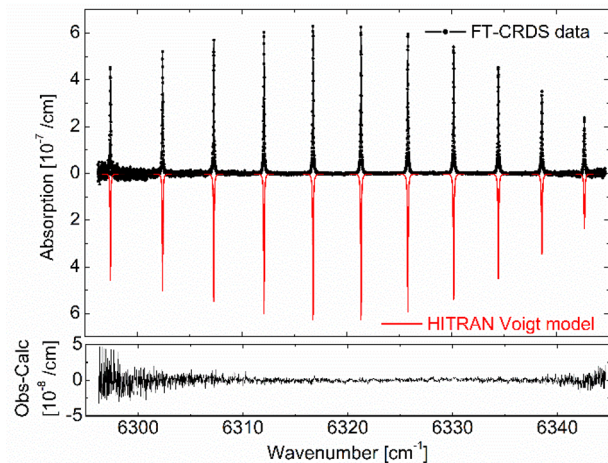
**Fig. 1.** Experimental setup: EOM, electro-optic modulator; AOM, acousto-optic modulator;  $\lambda/2,4$ , half, quarter waveplates; PBS, polarizing beam splitter; PZT, piezo transducer.

## 3 Spectroscopy of cold acetylene

Figure 2 shows the absorption spectrum obtained when the cavity is filled with 246.95(5) ppm of CO diluted in Ar at a pressure of 534 Torr and a temperature of 299 K. The baseline containing the broadband losses of the cavity mirrors and etalons have already been subtracted. The data shown is the average of 15 independent measurements acquired in 80 min with a spectral resolution of 250 MHz. The CO lines are compared to a model calculated using the linestrengths from the HITRAN database, the sample thermodynamic conditions and Voigt profiles where the centre frequency and pressure broadening coefficient of each transition are fitted (red trace, inverted). The residuum of the fit is

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shown in the lower panel and does not exhibit any residual line structure. The signal-to-noise ratio of the line at  $6316.7\text{ cm}^{-1}$  is above 330, with a minimal detectable absorption equal to  $1.9\times 10^{-9}\text{ cm}^{-1}$ . This sensitivity opens up for comb-resolved FT-CRDS studies using the sub-nominal resolution method [3].



**Fig. 2.** Resulting absorption spectrum of CO (upper panel, black trace) compared to the model based on the HITRAN linestrengths and Voigt profiles where the frequency centres and pressure broadening coefficients of each line have been fitted (red line, inverted for clarity). Lower panel: residuum of the fit.

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