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Poster presentation

Comb-locked cavity ring-down spectroscopy: combining high precision with ultra-high detection sensitivity in molecular interrogation

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In the last decade, the technology of optical frequency combs (OFCs) has been successfully integrated with cavity-enhanced spectroscopic techniques in a variety of different ways, leading to interesting developments in the field of molecular spectroscopy.¹ More specifically, it has been possible to achieve high detection sensitivity in conjunction with high precision and accuracy in optical frequency measurements. Consequently, metrological-grade qualities have been added to molecular spectra, with relevant implications in various fields of fundamental and applied research.

Here, we report on a few examples of comb-locked cavity ring-down spectroscopy (CL-CRDS) performed in the spectral regions around 1.4 and 2 μm . A first example deals with residual water concentration measurements in ultra-high-purity gases that are needed in manufacturing processes of the semiconductor industry to meet the quality requirements.² The CL-CRDS system was based on a hemispherical optical resonator with a finesse as high as 507000, which gave an empty-cavity ring-down time of about 285 μs . Taking advantage of the frequency stability guaranteed by the optical frequency comb, it was possible to implement the strategy of long-term spectra averaging to remove efficiently the effect of mechanical, acoustic, and thermal noises. As a result, we could achieve a minimum detectable absorption coefficient as low as $3.7 \times 10^{-13} \text{ cm}^{-1}$, which corresponds to a limit of detection for H_2O in N_2 of nine parts per trillion and a H_2O partial pressure of $2 \times 10^{-8} \text{ Pa}$ (or $2 \times 10^{-10} \text{ mbar}$), numbers which set a benchmark for CRDS-based trace water detection.³ The potential of our approach was further demonstrated by recording the absorption features of rare water isotopologues, such as HD^{16}O and HD^{18}O , in ambient air.

A similar setup was developed for Lamb-dip spectroscopy of several P, Q, and R components of weak vibrational bands of C_2H_2 .⁴ Selection of the lines was based on the theory of spectroscopic networks (SN), in order to ensure that a large number of transitions, measured previously by precision-spectroscopy investigations, could be connected to the *para* and *ortho* principal components. The analysis of sub-Doppler profiles and the extrapolation of absolute frequencies to zero pressure in each case produced a combined average uncertainty of the measured line-center positions of about 7 kHz with a 1- σ confidence level.⁵

Finally, we report on preliminary results obtained by means of a new CL-CRDS spectrometer, expressly developed to perform accurate tests of quantum chemistry calculations for N_2O and CO_2 in the spectral region of 2 μm .⁶ An example spectrum is shown in Figure 1, together with the results of line-fitting of a vibro-rotational CO_2 transition at 4980 cm^{-1} in a sample of ambient air.

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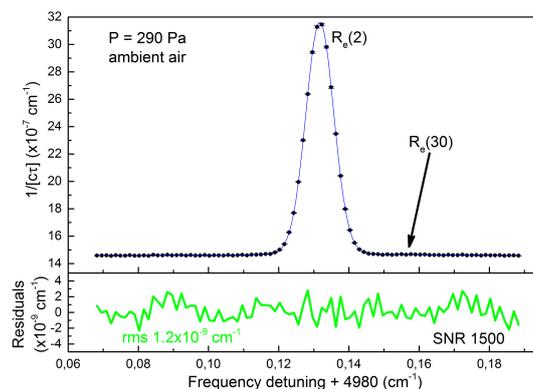


Figure 1: CO_2 spectrum, as recorded by means of the 2- μm comb-locked CRDS spectrometer.

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