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Optical Frequency Comb Fourier Transform Spectroscopy

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Akademisk avhandling

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Abstract

Fourier transform spectroscopy (FTS) based on optical frequency combs is an excellent spectroscopic tool as it provides broadband molecular spectra with high spectral resolution and an absolutely calibrated frequency scale. Moreover, the equidistant comb mode structure enables efficient coupling of the comb to enhancement cavities, yielding high detection sensitivity. This thesis focuses on further advances in comb-based FTS to improve its performance and extend its capabilities for broadband precision spectroscopy, particularly in terms of i) spectral resolution, ii) accuracy and precision of molecular parameters as well as concentrations retrieved from fitting models to spectra, and iii) species selectivity.

To improve the spectral resolution we developed a new methodology to acquire and analyze comb-based FTS signals that yields spectra with a resolution limited by the comb linewidth rather than the optical path difference of the FTS, referred to as the sub-nominal resolution method. This method enables measurements of narrow features, e.g. low-pressure absorption spectra and modes of enhancement cavities, with frequency scale accuracy and precision provided by the comb. Using the technique we measured low-pressure spectra of the entire $3\nu_1 + \nu_3$ carbon dioxide (CO_2) band at 1575 and retrieved spectral line parameters for this CO_2 band using the speed-dependent Voigt profile. We measured the transmission modes of a Fabry-Perot cavity over 15 THz of bandwidth with kHz resolution. From the mode center frequencies, we retrieved group delay dispersion of cavity mirror coatings and intracavity gas with an unprecedented combination of spectral bandwidth and resolution. By measuring both the cavity mode broadening and frequency shift simultaneously we performed broadband cavity-enhanced complex refractive index spectroscopy (CE-CRIS), which allowed for simultaneous and calibration-free assessment of the absorption and dispersion spectra of three combination bands of CO_2 in the 1525 to 1620 nm range.

We improved the noise-immune cavity-enhanced optical frequency comb spectroscopy (NICE-OFCS) technique in terms of stability, sensitivity and modeling of the NICE-OFCS signal. We implemented a model of the NICE-OFCS signal with multiline fitting for assessment of gas concentration and identified the optimum operating conditions of the NICE-OFCS system.

To improve the species selectivity we combined comb-based FTS with Faraday rotation spectroscopy (FRS), a technique called optical frequency comb Faraday rotation spectroscopy (OFC-FRS), to measure background and interference-free spectra of the entire Q- and R-branches of the fundamental band of nitric oxide at 5.3 μm showing good agreement with the theoretical model.

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