PHD PROJECT DESCRIPTION
(4000 characters max., including the aims and work plan to de published online)

Project title: Electro-Optic Frequency Combs for High-Accuracy Broadband Molecular Spectroscopy in Optical Cavities

Project goals

The goal of this PhD project is to improve the accuracy and capability of high-resolution, broadband molecular spectroscopy of atmospheric gases by (i) developing electro-optic optical frequency combs based on continuous-wave lasers with high power per comb tooth and precise controllability, (ii) implementing advanced cavity-enhanced, frequency-based spectroscopic techniques such as Cavity Mode Width Spectroscopy (CMWS) [1], Cavity Mode Dispersion Spectroscopy (CMDS) [2,3], and dual-comb Cavity Ring-Down Spectroscopy (dual-comb CRDS) [4,5], and (iii) performing simultaneous, high-accuracy measurements of multiple spectral lines of key atmospheric molecules such as CO, CO_2, and CH_4, enabling improved analysis of dense spectra including the influence of neighboring transitions.

The combination of electro-optic comb sources with frequency-based spectroscopic methods developed in our laboratory will enable efficient comb-to-cavity coupling, high measurement precision, and broadband acquisition with resolution limited by cavity mode linewidths. This approach will provide improved determination of line positions and widths, contributing to more accurate spectroscopic databases and more reliable molecular identification. The project is directly aimed at advancing spectroscopic tools for atmospheric sensing and precise characterization of greenhouse gases.

1.1. Outline

Optical frequency combs (OFCs) have become a key tool in precision spectroscopy, enabling simultaneous measurement of many spectral lines with high absolute frequency accuracy. They play a particularly important role in atmospheric research and trace gas detection, where both broadband coverage and high resolution are required. Most OFC-based systems rely on femtosecond mode-locked lasers, which provide very wide spectral spans but are relatively complex and offer limited flexibility in controlling comb parameters, making their integration with cavity-enhanced techniques challenging [6].

An alternative approach is the generation of OFCs based on continuous-wave (CW) lasers combined with electro-optic modulation (EOM) [7]. Although such combs typically exhibit a narrower spectral bandwidth, they offer significantly higher optical power per comb tooth and, importantly, straightforward and precise control over key parameters such as repetition rate and offset frequency. This makes them particularly attractive for efficient matching and stabilization to optical cavity modes. Previous studies have demonstrated direct coupling of femtosecond-based combs to cavity modes, although these implementations required complex and demanding stabilization schemes. Electro-optic combs offer a more flexible and potentially robust route toward comb–cavity matching.

High-resolution spectroscopy of atmospheric molecules such as CO, CO_2, and especially CH_4 requires not only narrow linewidths but also the ability to measure multiple transitions simultaneously. This is particularly important for dense spectra, where neighboring lines significantly influence observed line shapes and retrieved spectroscopic parameters. Simultaneous multi-line acquisition under identical conditions enables more accurate modeling of these effects, improving both molecular identification and quantitative analysis. Additional gains in accuracy can be achieved using frequency-based techniques such as CMWS [1] and CMDS [2,3], which rely on precise measurements of cavity mode widths and positions rather than optical intensity.

The present project aims to develop and apply electro-optic frequency combs as direct light sources for advanced cavity-enhanced spectroscopic techniques, including CMWS, CMDS, and dual-comb CRDS [4,5]. By leveraging the high power per comb tooth and precise controllability of EOM-based combs, the project seeks to achieve efficient comb-to-cavity coupling and stable operation over extended measurement times. The developed system will enable simultaneous, high-resolution, broadband measurements of molecular spectra, with a particular focus on methane, where dense spectral features provide a stringent test of the method.

Ultimately, this approach is expected to improve the accuracy of spectroscopic measurements by enabling comprehensive multi-line analysis, incorporating the influence of neighboring transitions, and utilizing frequency-based observables. The results of the project will contribute to advancing optical spectroscopy as a precise and reliable tool for atmospheric sensing and fundamental molecular studies, while establishing electro-optic frequency combs as a practical and versatile alternative to traditional femtosecond-based OFC systems.

1.2. Work plan

- **Development of the electro-optic frequency comb source**
  Design and construction of a CW-laser-based electro-optic frequency comb using phase and intensity modulators, with optimization of bandwidth, spectral flatness, power per comb tooth, and coherence properties.
- **Comb stabilization and comb–cavity coupling**
  Implementation of stabilization schemes enabling precise control of comb parameters and efficient matching to optical cavity modes, including demonstration of stable and robust comb-to-cavity locking.
- **Implementation of frequency-based cavity-enhanced spectroscopic techniques**
  Development and integration of frequency-based methods, including CMWS, CMDS, and dual-comb CRDS, for high-accuracy, broadband measurements.
- **High-resolution spectroscopy of atmospheric molecules**
  Broadband, simultaneous measurements of CO, CO_2, and CH_4 spectra, with particular focus on methane as a dense and spectroscopically demanding system.
- **Advanced data analysis and line-shape modeling**
  Development of analysis methods accounting for neighboring line effects and line mixing, enabling improved accuracy of retrieved spectroscopic parameters through simultaneous multi-line fitting.
- **Presentation of results including publications and conference presentations**
- **PhD thesis preparation**

1.3. Literature (max. 7 listed as a suggestion for a PhD candidate preliminary study)

[1] A. Cygan et al., Opt. Express 21, 29744 (2013).
[2] A. Cygan et al., Opt. Express 23, 14472 (2015).
[3] A. Cygan et al., Opt. Express 27, 21810 (2019).
[4] D. Lisak et al., Sci. Rep. 12, 1 (2022).
[5] A. Cygan et al., Sci. Adv. 11, eadp8556 (2025).
[6] A. Foltynowicz, et al., Phys. Rev. Lett. 107, 233002 (2011).
[7] A. J. Fleisher et al., Opt. Express 24, 10424 (2016).

1.4. Required initial knowledge and skills of the PhD candidate

Knowledge of optics, spectroscopy, atomic and molecular physics. Good skills and experience in numerical methods and programming (preferred Labview, Mathematica, Fortran, Python). Mathematical skills in theoretical calculations. Experience in laboratory work is desirable, especially in building and using optical laser-based systems. Independence at work is welcome. High commitment to work and excellent problem-solving skills. Written and verbal communication and presentation skills (including English). Teamwork skills.

1.5. Expected development of the PhD candidate's knowledge and skills

Knowledge, skills, and experience in laboratory work: building electro-optical systems (especially cavity-enhanced spectrometers), techniques of phase and intensity modulation of light, techniques of laser frequency stabilization, experimental techniques of molecular spectroscopy. Good knowledge of molecular spectroscopy, theoretical skills in description of interactions of light with molecules and with optical cavity. Skills in programming and numerical methods. Experience in finding solutions, making hypotheses and formulating final conclusions. Presentation skills, including research papers and conference presentations.