

1. PHD PROJECT DESCRIPTION (4000 characters max., including the aims and work plan)

Project title: *Spectroscopy of quantum systems by means of exact and fast localization of optical cavity resonances*

1.1. Project goals

Main goals:

- development of new cavity-enhanced (CE) spectroscopy methods, for accurate and dynamic sensing of quantum systems, based on a novel idea of exact and fast localization of optical cavity resonances solely by means of frequency measurement,
- application of developed techniques for accurate measurements of molecular spectral line shapes relevant for atmospheric research of Earth and other planets.

Secondary goals:

- studying of rapid processes in the optical cavity associated with dynamic changes in absorption,
- testing of new solutions supporting the pressure metrology based on optical properties of molecules.

1.2. Outline

Accuracy in spectral line shape metrology is becoming an increasingly important determinant of progress in areas such as optical metrology of trace gases¹, Earth's atmosphere remote sensing^{2,3}, quantum chemistry⁴, astrochemistry⁵, and tests of fundamental physics in basic molecular systems^{6,7}. Modern high-resolution molecular spectroscopy benefiting from ongoing development in optical frequency metrology provides more and more accurate data on molecular transition energies. Nowadays, spectral line positions can be determined with relative accuracies better than 10^{-11} ^{8,9}. However, in case of measurement of the shape of the spectral line the situation is completely different. Here, obtaining the line profile that is systematically undistorted at the accuracy level of 10^{-3} is a serious challenge. The main reason for this is seen in a non-linear response of a detection system to light intensity measurement¹⁰. Presented PhD project addresses this very actual problem and focuses on development of new spectroscopic techniques that will be insensitive to the measurement of light intensity. The project will be realized in collaboration with optical measurement group from NIST (USA), one of the world-leading metrological institute, from which we initially gained experience in building of cavity ring-down spectrometers (CRDS).

The highest accuracy of the measured molecular spectrum can be provided by the purely frequency measurement, which in case of using an optical cavity is measurement of the dispersive shift of cavity modes in the presence of the absorber. This technique, called cavity mode-dispersion spectroscopy (CMDS)¹¹, has its origin at NCU and is still actively developing here. Its superior accuracy has been already demonstrated. First in 2019, when CMDS¹² together with NIST-calibrated CRDS¹³ set a new record in the accuracy of spectral line shape measurements at the level of 10^{-4} , and recently as a technique providing sub-promille agreement of an experiment with calculations for CO (3-0) overtone line intensities¹⁴. The CMDS was also extended to its high-speed variant, cavity buildup spectroscopy (CBS), thanks to measuring laser-cavity field interferences resulting in fast localization of cavity resonances¹⁵ as well as was adopted to broadband direct optical frequency comb spectroscopy¹⁶. This PhD project assumes further optimization of the CBS speed and accuracy, construction of the dual-comb laser system for the broadband CBS and accurate spectroscopy of CO or/and D₂ weak transitions. State-of-the art dual-comb broadband CRDS with extraordinary spectral resolution was recently developed by us¹⁷. Additionally, observation of non-stationary conditions in the

cavity and precise metrology of pressure can be included in the project.

1.3. Work plan

- Design and construction of optical frequency combs based on phase and intensity modulation in an electro-optic modulator (simulation of the comb spectral range, optimization of comb parameters).
- Construction, software and tests of the dual-comb cavity-enhanced spectrometer (optimization of the CBS method).
- Simulations of stationary and non-stationary conditions in the cavity.
- High-accuracy measurements followed by analysis of spectral line shapes of CO and D₂ molecules.
- Observation of saturation effects in the optical cavity.
- Pressure measurement on the basis of recorded cavity dispersion shift and line intensity.

1.4. Literature

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- [2] K. R. Gurney et al., Nature 415, 626 (2002).
- [3] D. R. Thomson et al., J. Quant. Spectrosc. Radiat. Transf. 113, 2265 (2012).
- [4] O. L. Polyansky et al., J. Quant. Spectrosc. Radiat. Transf. 210, 127 (2018).
- [5] P. F. Bernath, Phil. Trans. R. Soc. A 372, 20130087 (2014).
- [6] J. Komasa et al., Phys. Rev. A 100, 032519 (2019).
- [7] M. Zaborowski et al., Opt. Lett. 45, 1603 (2020).
- [8] K. Bielska et al., J. Quant. Spectrosc. Radiat. T 201, 156 (2017).
- [9] J. Wang et al., J. Chem. Phys. 147, 091103 (2017).
- [10] S. Wójtewicz et al., Phys. Rev. A 84, 032511 (2011).
- [11] A. Cygan et al., Opt. Express 23, 14472 (2015).
- [12] A. Cygan et al., Opt. Express 27, 21810 (2019).
- [13] A. J. Fleisher et al., Phys. Rev. Lett. 123, 043001 (2019).
- [14] K. Bielska et al., Phys. Rev. Lett., in review (2022).
- [15] A. Cygan et al., Comm. Phys. 4, 1 (2021).
- [16] D. Charczun at al., Measurement 188, 110519 (2022).
- [17] D. Lisak et al., Sci. Rep. 12, 1 (2022).

1.5. Required initial knowledge and skills of the PhD candidate

Good knowledge of optics, spectroscopy, atomic and molecular physics. Skills and experience in numerical methods and programming (knowledge of Labview, Mathematica, Fortran, C++ is welcome). Math skills in theoretical calculations. Experience in laboratory work, especially in construction of optical systems, is welcome. Great commitment to work and excellent problem-solving skills. Written and verbal communication skills as well as presentation skills. Teamwork ability.

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

Knowledge, skills and experience in laboratory work: construction of optical systems (especially cavity-enhanced spectrometers), techniques of phase and intensity modulation of light, techniques of laser light stabilization and narrowing, generation of optical frequency combs, experimental techniques of molecular spectroscopy. Good knowledge of molecular spectroscopy, theoretical skills in description of interactions of light with the optical cavity. Skills and experience in computer coding and numerical methods. Presentation skills.