Spectroscopy of quantum systems by means of exact and fast localization of optical cavity resonances

In many contemporary studies, including in the field of accurate optical metrology of trace gases, precise monitoring of the Earth's atmosphere, quantum chemistry, astrochemistry or testing quantum-electrodynamic calculations and searching for physics going beyond the Standard Model in the basic molecular systems, it is important to accurately measure the shape of the spectral line. Registering a line profile with an accuracy of 10^{-3} is considered as incredible progress in these areas, and although it is more than 8 orders of magnitude worse than the accuracy associated with current measurements of molecular transition energy, it is a serious challenge for the most experimental groups. The problem is the measurement of light intensity present in almost all spectroscopic techniques, but unfortunately very susceptible to systematic distortion from the detection system. Its solution is either a professional calibration of the detector linearity, however, requiring cooperation with the best metrology institutes in the world, or the search for alternative spectroscopic methods, by definition independent of measuring light intensity. In our team since 2015 the CMDS (cavity mode-dispersion spectroscopy) spectroscopic technique is being developed, which is based on molecular spectrum retrieval from purely frequency measurement of optical cavity resonances. Its huge potential results from the fact that frequency is a quantity that we can measure even with relative accuracy of 10^{-18}. It also gives the easy way of linking both molecular spectrum axes to the atomic frequency standard. In 2019 CMDS technique together with the NIST-calibrated absorption method set a new record for the accuracy of spectroscopic measurements at the level of 10^{-4}. Unfortunately, the CMDS method, compared to other cavity-enhanced methods, is not so fast, making it more susceptible to various drifts of physical quantities and not applicable to measurements of fast processes.

The goal of this project is development of CBS (cavity buildup spectroscopy) frequency methods, based on the innovative idea of the exact and fast localization of the cavity resonance based on the measurement of the beating signal frequency between a non-resonant laser field exciting the cavity mode and the resonant field built in the cavity. This approach will provide many new benefits for experimental spectroscopy. It will allow achieving a fundamental limit on the spectrum measurement speed, previously unattainable for CMDS methods, and related to the cavity photon lifetime. Applied to broadband spectroscopy using a double optical frequency comb, it will allow the world's first real-time measurements to be carried out with an accuracy guaranteed by the best frequency standards. Such system will be also a promising high-resolution alternative for current comb-based broadband CRDS systems having limited resolution and for cavity-enhanced dual-comb spectroscopy relaying on intensity measurements. Non-stationary conditions of the field in the cavity, accompanying the registration of the beating signal in the CBS method, will also give the opportunity to study rapid processes associated with dynamic changes in absorption in the optical cavity occurring on a time scale much shorter than the cavity photon lifetime. The dynamic CBS technique developed as part of the project can provide a time resolution and measurement sensitivity higher than in case of other current techniques. The CBS method will be also used to support development of the new-generation metrology standard of pressure based on optical properties of molecules.

Accurate measurements of spectral line shapes of CO and CO_2 molecules, important from the point of view of atmospheric studies of the Earth and other planets, as well as the D_2 molecule, which plays an important role in the basic physics research, are planned in the project. Simultaneous measurement of absorption and dispersion spectra possible in CBS method will enable development of highly accurate complex refractive index spectroscopy with many potential applications, including thorough testing of molecular collision theory.