Spectroscopy is a key tool to study properties of matter and its interaction with radiation. Among many available spectroscopic methods the cavity enhanced spectroscopies are the most sensitive due to a very long optical path length of interaction between radiation and quantum system in the cavity. Therefore these methods have become important tools used in basic research concerning atomic and molecular physics, and quantum optics.

The aim of this project is development of new spectroscopic method based entirely on frequency measurement. Contrary to any absorption spectroscopic techniques, the proposed method of the cavity mode dispersion spectroscopy is one-dimensional – both axes of the spectrum are created from the same frequencies of the electromagnetic cavity modes. This is a huge advantage since frequency can be nowadays measured with relative uncertainties of even 10^{-17}, which is far beyond the reach of any light intensity measurements. Considering this advantage the new method may become a future standard of ultra-accurate spectroscopy and a tool to provide reference data on transition frequencies and line intensities for testing fundamental interaction of matter and searching physic beyond standard model. Such a reference data are also crucial for testing ab initio methods of spectra calculations and for the most demanding applications in the atmospheric research or gas metrology. Moreover we plan to develop a complex refractive index spectroscopy which combines well established absorption technique of cavity ring-down spectroscopy with new dispersion spectroscopy. The cavity ring-down spectroscopy in which the information on absorption coefficient of the medium is obtained from the measurement of the decay time of radiation from the cavity after switching off the laser pumping the cavity mode. Exponential decay of radiation, resulting from the Beer-Lambert law, allows to determine the absorption coefficient of the medium from the inverse of the time constant. This method has become very attractive because of the insensitivity to fluctuations in laser power resulting from the measurement of time, not the amplitude of the radiation decay, but this method requires a highly linear light detector. Study of the complex spectra gives unique possibility to verify consistency between absorption and dispersion spectra and eliminate potential systematic error in experimental data. Therefore this new system will be used to study spectra of molecules particularly important in physics of atmosphere and test the advanced theories of spectral line shapes.

Accurate measurement of resonant dispersion associated with weak molecular transitions in gas medium requires very high spectral resolution. By measuring the frequencies of the electromagnetic modes of a high-finesse optical cavity one can provide extremely accurate information on dispersive spectrum of gas inside the cavity. Such method, however, requires use of laser with ultra-narrow line width and elimination of the drift and acoustic noise in the cavity. Both these requirements can be fulfilled by phase-locking the laser to the cavity mode and isolating the cavity from surrounding environment with properly designed temperature-stabilized vacuum chamber and locking the cavity optical path length to an optical frequency reference. Such system with frequency reference linked to the primary frequency standard, or strontium optical lattice clock (project POZA), available in KL-FAMO lab will allow for measurement of the mode frequency shifts with sub-Hertz accuracy.

Comparison of absorption and dispersion line shape in the linear absorption regime can be done with the Kramers-Kronig relations. The complex line shape function models will be used to validate the absorptive and dispersive spectra. Experimental study will be supported by development of algorithms for spectral data analysis. In particular, software for multi-spectrum complex line-shape fitting will be developed. As was demonstrated on absorption spectra simultaneous fitting of experimental data corresponding to different physical conditions (gas pressure, temperature) allows to study higher order collisional effects on molecular spectra.

Several laboratories have recently demonstrated absorption spectra with very high signal-to-noise ratio and emphasized their advantages in the basic research and metrology, but nonlinearity of absorption measurement was a dominant source of systematic error in those spectra. We can measure frequencies of atomic or molecular lines with great accuracy, but achieving agreement of line intensities from two laboratories at level better than 10^{-3} is a great challenge. Development of the proposed frequency-based spectroscopic method has a chance to change this by increase of accuracy and by traceability of measurements to the primary frequency standard, which is crucial for reliability of the reference laboratory data in the international scientific community. Recent ab initio calculations of the molecular spectral line intensities achieve accuracy at level of 10^{-3}. The experimental verification, and thus further improvement of these methods encounter difficulties due to disagreement of experimental results from different laboratories. The accuracy of measurement of the spectral line profile is also particularly important in metrology applications such as Doppler-width based optical thermometry and determination of the Boltzmann constant, and optical metrology of gas concentration, for example. hygrometry. Spectroscopy based on pure frequency measurement also in these applications will be very attractive.

Tests of the new method will be conducted in the spectra of CO and CO_2 molecules of great importance in the study atmosphere and climate changes, in the spectral range 1.5 - 1.65 um, and therefore together with development of the new methods of spectroscopy, we expect to analyze the impact of physical effects of spectral lines in these systems and we will provide accurate reference data on the intensity, transition frequencies and line-shape parameters. These reference spectroscopic data will be provided in a form compatible with recently recommended for the next generation databases line-shape model.