Further development of NICE-OHMS
– an ultra-sensitive frequency-modulated cavity-enhanced laser-based spectroscopic technique for detection of molecules in gas phase

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Abstract

Noise-immune cavity-enhanced optical heterodyne molecular spectroscopy, NICE-OHMS, is a laser-based spectroscopic detection technique that comprises the concepts of frequency modulation (FM, for reduction of $1/f$-noise by detecting the signal at a high frequency) and cavity enhancement (CE, for a prolongation of the optical path length) in a unique way. Properly designed, this gives the technique an intrinsic immunity against the frequency-to-noise conversion that limits many other types of CE techniques. All this gives it an exceptionally high sensitivity for detection of molecular species. Although originally developed for frequency standard purposes in the late 1990s, soon thereafter development of the technique towards molecular spectroscopy and trace gas detection was initiated. This thesis focuses on the further development of Doppler-broadened NICE-OHMS towards an ultra-sensitive detection technique. A number of concepts have been addressed. A few of these are: *i*) The detection sensitivity of fiber-laser-based NICE-OHMS has been improved to the $10^{-12}\,\mathrm{cm}^{-1}$ range, which for detection of C_2H_2 corresponds to a few ppt (parts-per-trillion, 1:10^{12}) in gas phase, by improving the locking of the laser to a cavity mode by use of an acousto-optic modulator. *ii*) It is shown that the system can be realized with a more compact footprint by implementation of a fiber-optic circulator. *iii*) A systematic and thorough investigation of the experimental conditions that provide maximum signals, referred to as the optimum conditions, e.g. modulation and demodulation conditions and cavity length, has been performed. As a part of this, an expression for the NICE-OHMS line shape beyond the conventional triplet formalism has been proposed and verified. *iv*) To widen the applicability of NICE-OHMS for detection of pressure broadened signals, also a setup based upon a distributed-feedback (DFB) laser has been realized. *v*) In this regime, the Voigt profile cannot model signal with the accuracy that is needed for a proper assessment of analyte concentrations. Therefore, the thesis demonstrates the first implementations of line profiles encompassing Dicke narrowing and speed-dependent effects to NICE-OHMS. While such profiles are well-known for absorption, there were no expressions available for their dispersion counterparts. Such expressions have been derived and validated by accompanying experiments. *vi*) The applicability of the technique for elemental detection, then referred to as NICE-AAS, has been prophesied.

Keywords

NICE-OHMS, Frequency Modulation, Cavity Enhancement, Molecular Spectroscopy

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