Doppler-Broadened NICE-OHMS Beyond the Cavity-Limited Weak Absorption Condition

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NICE-OHMS is a laser-based technique that has demonstrated an extraordinary high sensitivity for detection of molecules in gas phase. Its Doppler-broadened (Db) response is usually described by an expression that is restricted to the conventional cavity-limited weak absorption (CCLWA) condition, i.e. when the single pass absorbance is much smaller than the empty cavity losses. However, with the realization of NICE-OHMS in the Mid-IR range, in which the transition line strengths can be huge, often around 10^{-18} cm^{-1}/(molecule cm^{-2}), and with the use of high finesse cavities, it has become more and more common that assessments and calibrations are no longer performed under cases for which this condition is valid. So far, no analytical expression for the Db NICE-OHMS signal for the situations when the CCLWA condition is not fulfilled has been derived. Nor is the principle of Db NICE-OHMS under such conditions well known. We have therefore scrutinized in detail the principles of Db NICE-OHMS. Based on this, we have been able to provide a description of the technique that is based solely upon a few general assumptions and thus not limited to the CCLWA condition (termed the FULL description). Unfortunately, this description constitutes a set of equations to which there is no closed form solution. Hence, it needs to be solved numerically. To circumvent this, two analytical expressions have been derived; one based on an extended locking & extended transmission (ELET) description, and another on an extended locking & full transmission (ELFT) description, valid under close-to CCLWA conditions. By use of simulations and experiments, the extended descriptions have been verified and their ranges of applicability have been assessed. Access to these increases considerably the dynamic range of the technique, by at least two orders of magnitude, and facilitates calibration using certified reference gases, which significantly broadens the applicability of the Db NICE-OHMS technique.