Saturation Dynamics and Working Regimes of Saturated Absorption Cavity Ringdown Spectroscopy (Sat.-CRDS)

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In contrast to the well-established CRDS procedures in the linear absorption regime, no mature implementation for trace gas measurements in the presence of absorption saturation has been introduced yet. Modeling the saturation process is complex: it depends on the characteristics of the absorbing species, its interaction with the surrounding gas as well as on the temporal and spectral characteristics of the laser beam. Recently, a new approach of CRDS has been developed that relies on saturating the gas absorption using narrow linewidth continuous-wave (cw) laser sources[1]. Sat.-CRDS allows one to extract both the gas absorption and the cavity loss from a single ringdown signal simultaneously, resulting in lower detection limits. This is possible because the degree of sample saturation changes during the ringdown event causing non-exponential behavior. In this work, a new cw-CRDS experiment based on a high power cw-IR-OPO system has been setup. The dynamics of the saturation process, which is controlled by the degree of saturation as well as the relaxation rate relative to the empty cavity decay rate, has been investigated in detail. The transition from the highly saturated absorption regime to a lower degree of saturation has been studied. Both the intracavity intensity as well as the saturation intensity have been varied independently. Moreover, the saturation intensity was tuned either by changing the relaxation rate or by selecting absorption lines with different line strengths. A statistical analysis that provides the ultimate working conditions of Sat.-CRDS method will be introduced. The optimal values for the degree of saturation, which strongly controls the standard error as well as the sensitivity of the method, have been determined. Finally, the validity of the Sat.-CRDS model to retrieve the gas absorption and the empty cavity loss at different concentrations of the absorbing species has been tested.

References
[1] Giusfredi et al., Phys. Rev. Lett. 104 (2010)