INTRODUCTION

Highlights
- Photoacoustic spectroscopy combined with optical power enhancement in high finesse cavity
- Highly efficient coupling (> 80%) of mid-infrared light into the cavity using optical feedback locking based on Brewster window cavity design
- Effective power inside the cavity increased from 25 mW to 6 W after optical impedance matching via angle of Brewster window
- Trace detection of CO, limit of detection = 6 ppbHz (3σ)

Photoacoustic spectroscopy
Absorption can be detected by measuring the reduction of light’s intensity (direct absorption spectroscopy) or by measuring the energy dissipated in the sample upon absorption (indirect spectroscopy). In photoacoustic spectroscopy (PAS) the energy dissipated as sound is detected via a microphone or a quartz tuning fork (Quartz Enhanced PAS, QEPAS). As compared to direct absorption spectroscopy, PAS allows for the use of gas cells with small volume of a few cm³, yielding fast gas exchange and response time. Since the photoacoustic signal increases with optical power, high power quantum cascade lasers (QCL) are ideal sources for PAS and allow targeting fundamental molecular vibrational modes in the mid-infrared.

Cavity-enhanced spectroscopy
High finesse optical cavities can boost the sensitivity of laser absorption spectroscopy by increasing the effective optical path length. The achievable enhancement scales with the mirror reflectivity R as 1/(1-R) and reaches values of several 1000s for state-of-the-art mirrors. The mid-infrared spectral region is particularly attractive for spectroscopic trace gas sensing since strong fundamental absorption lines of most molecules fall in this region.

OPTICAL FEEDBACK LOCKING - THE BREWSTER WINDOW CAVITY

Power buildup in high finesse cavities
Ideally, the optical power buildup, i.e. the power circulating inside the cavity relative to the incident power, is equal to the path length enhancement given by 1/(1-R). Two effects can drastically reduce the buildup factor:
- Laser phase noise: The free running laser linewidth is typically larger than the widths of the cavities resonance profile, yielding noisy and incomplete buildups.
- Optical impedance mismatch: For maximum buildup to occur, the transmission T of the incoupling mirror must match the losses at all other elements of the cavity. For highly reflective mirrors, this is hardly achievable as T ≤ 1-R.

Employing the Brewster-window cavity design can overcome these limitations.

Optical feedback
Optical feedback can drastically reduce laser phase noise and achieve stable, complete buildups [3]. Here, light from the cavity is reflected back into the laser, forcing it to emit light at the re-injected light’s wavelength. When coupling into the cavity via the Brewster window (compare scheme), off-resonance wavelengths (grey) are transmitted through the window and do not return to the laser. Since only resonant light (red) is fed back, optical feedback locks the laser to a cavity resonance, given the back reflected light is in phase (DF = 0) with the emitted one. The latter is essentially adjusting the optical path length between laser and cavity via a mirror on a piezo.

When ramping the laser’s wavelength across a cavity resonance (see figure), optical feedback locking occurs, resulting in a buildup of power inside the cavity. As the laser is tuned away from the cavity resonance far enough, it returns to its free running, off-resonance wavelength. The duration and temporal profile of the buildups depends on the feedback phase. This is exploited to actively stabilise the feedback phase.

EXPERIMENTAL AND RESULTS

Experimental details
- Laser
 CW DFB-QCL, AdTech HLH, 41 mW (25 mW at cavity)
- Cavity
  L = 28 cm, r = 750 mm, F = 2500, waist radius = 0.3 mm, R₀ = 0.9992, RO = 15 cm, Layeret
  CaF₂/Brewster window (Ø = 10 mm)
- Intra-cavity power buildup (bidirectional) buildup factor: 300, coupling efficiency 81%, power 6 W
- Feedback ratio of optical feedback: 2%
- Custom quartz tuning fork (Thorlabs)
  f₀ = 15.9 MHz, Q = 100 G × µs, prong spacing: 1.5 mm

Spectra of ambient air
The cavity ring-down and QEPAS spectra of ambient air show absorption lines of CO, N₂O, and NO₂. With respect to H₂O, the QEPAS signal of N₂O and NO₂ are much weaker than expected from the cavity ring-down spectrum. Especially for CO, slow vibrational-translational (V-T) relaxation limits the sensitivity of QEPAS. Addition of water vapor strongly increases V-T relaxation rates and hence QEPAS signals of CO.

Calibration and LODs
Calibration was performed for CO in humidified N₂. The QEPAS signal was normalized to the back-reflection signal from the detector that is directly proportional to intracavity power. The offset (signal at 0 ppb) originates from water absorption. Using the noise floor determined for humidified N₂, a noise equivalent concentration (1σ) of 2 ppb is found, corresponding to a power normalized noise equivalent absorption (both side) of 1.1±0.1 cm⁻¹ Hz⁻¹/2 (4.6±0.5 cm⁻¹ Hz⁻¹/2).

OPTICAL IMPEDANCE MATCHING

Optical impedance matching
The efficiency of power buildup P build/Plum (bidirectional) inside the cavity depends critically on the reflective losses at the Brewster window R₀:

P build/Plum = 1 - exp(-2πL/(1-R₀)R₀)

For too large R₀ the finesse is reduced. If R₀ is too small, the losses through the cavity mirrors exceed the "gain" of light that is coupled into the cavity via reflection at the Brewster window. A maximum is found for

R₀ = (1 - 1/n₀)⁻¹

by changing the angle of the Brewster window’s angle, the reflection can be chosen to maximize the optical power in the cavity.

SUMMARY AND CONCLUSIONS

QEPAS measurements of CO inside a high finesse cavity were performed. The optical power from a DFB-QCL is increased inside the cavity by a factor of 245 to a level of 6 W. A coupling efficiency of 81% was measured, enabled by optical feedback locking employing the Brewster window cavity design.

The Brewster window design also allows maximizing the intracavity power by means of optical impedance matching by tilting the window inside the cavity. The power normalized noise equivalent absorption of 4.6±0.5 cm⁻¹ Hz⁻¹/2 corresponds to a concentration of CO of 2 ppbHz⁻¹/2, is limited by slow V-T relaxation of CO.

Cavity-enhanced photoacoustic spectroscopy combines the advantages of indirect spectroscopy and high finesse cavities. Photodetection allows using small volume gas cells (few cm³), ensuring rapid gas exchange and short response times, while cavity enhancement increases the sensitivity for trace gas sensing.

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REFERENCES

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