High-Frequency Modulation Spectroscopy with a THz Quantum-Cascade Laser

R. Eichholz[a], H. Richter[a], M. Wienold[b], L. Schrottke[b], H. T. Grahn[b] and H.-W. Hübers[a,c]
[a]German Aerospace Center (DLR), Berlin, 12489, Germany
[b]Paul-Drude-Institut für Festkörperphysik, Berlin, 10117, Germany
[c]Technische Universität Berlin, Berlin,10623, Germany

***Abstract*—A terahertz absorption spectrometer with a quantum-cascade laser (QCL) for high-resolution molecular spectroscopy is realized. The spectrometer is based on high-frequency (up to 50 MHz) modulation of the QCL frequency. This allows for the determination of the absorption coefficient and dispersion of the absorbing medium along with a very precise measurement of the line shape of the absorption feature. The design and performance of the spectrometer are presented, and its sensitivity and frequency calibration are discussed.**

I. Introduction

Many physical phenomena have characteristic energies in the THz frequency range. In this frequency range, high resolution spectroscopy allows for the investigation of the structure and the energy levels of molecules and atoms. THz quantum-cascade lasers (QCLs) are promising radiation sources for such type of spectroscopy, because they are frequency tunable as well as powerful and exhibit a narrow line width. So far, absorption spectroscopy with QCLs employed modulation of the QCL frequency on the order of kHz and phase-sensitive detection [1,2]. We describe a spectrometer based on a QCL using frequency modulation (FM) spectroscopy with frequencies up to 50 MHz. This type of spectroscopy allows for the measurement of absorption and dispersion of a gas and is potentially very sensitive, because the modulation frequency is well above the frequencies of the most important noise sources [3].

II. Experimental Details

The QCL used in these experiments has a single-plasmon waveguide and a Fabry-Pérot cavity with both facets uncoated and is optimized for low electrical pump power. The laser is mounted in a commercially available, compact air-cooled Stirling cryocooler (model K535 from Ricor [4]). It operates on one laser-mode at 3.1 THz. The output power of the laser is 1 mW at a current level of 550 mA and a temperature of 45 K. Frequency tuning was achieved by varying the driving current of the QCL. The FM was achieved by superimposing an AC current of about 1–2 mA with a frequency up to 50 MHz onto the QCL driving current. The AC amplitude results in a modulation of the QCL emission frequency with a magnitude corresponding to the full width at half maximum of the spectral feature of interest of about 16 MHz at a pressure of 100 Pa. The condition for FM spectroscopy is achieved when the modulation frequency of the AC signal ω_m is large compared with the spectral feature of interest and only one sideband probes the spectral feature [3]. In this case, we obtain the maximum signal.

The used setup for these experiments is shown in Fig. 1. The beam is collimated with a TPX lens and guided through a 30 cm long absorption cell and focused onto a Schottky diode. The absorption is measured as a function of the laser driving current using a lock-in amplifier with a bandwidth of 50 MHz.

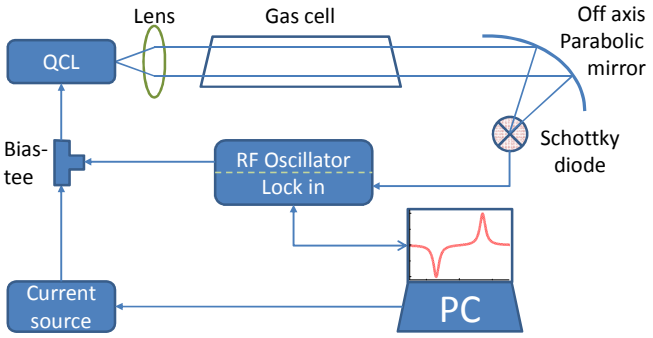

Figure 1: Block diagram of the experimental setup.

III. Results

An example of a line shape of the CH_3OH molecule at a pressure of 1 hPa measured with FM spectroscopy at 50 MHz is shown in Fig. 2. The experimental and calculated in-phase signals are displayed. The experimental curve was obtained by scanning the sidebands at ±50 MHz of the QCL frequency through the resonance via tuning the QCL current in steps of 0.5 mA (corresponding to 3 MHz) across the spectral feature. While the frequency of the QCL is increasing, the upper FM sideband probes the absorption line first (feature at +50 MHz in Fig. 2), while the second feature at −50 MHz occurs when the lower FM sideband passes the absorption line. The spacing between the two features corresponds to $2\omega_m$, which can be used to calibrate the frequency-current dependence of the QCL at a single spectral feature with high resolution.

A limitation is given by the line width of the molecular transition. This is illustrated in Fig. 3, which shows the experimentally determined line shapes for different modulation frequencies ranging from 1 up to 50 MHz. Almost no change of the position of the feature occurs in the lower modulation frequency range up to about 10 MHz.

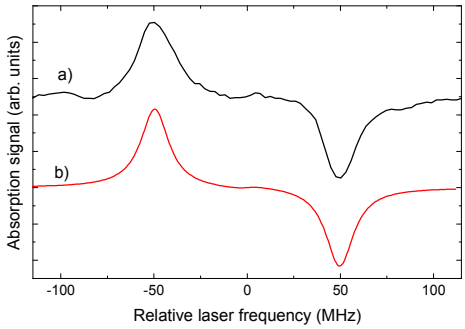

Figure 2: (a) Experimental and (b) calculated in-phase line shape for FM spectroscopy.

The reason for this limitation is the pressure broadening of the CH_3OH line which is approximately 15 MHz. This can be overcome by decreasing the pressure of the gas, at the expense of a reduced absorption signal. Once the modulation frequency is significantly larger than the line width, the separation of the two features is equal $2\omega_m$.

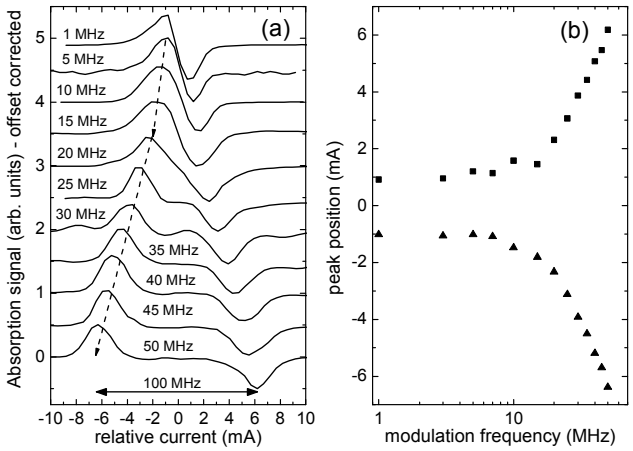

Figure 3: (a) Experimental measured line shapes (offset for clarity) and (b) the center position of the spectral feature for different modulation frequencies. In (a), the numbers indicate the modulation frequencies and the arrow the spacing between the two spectral features.

IV. Summary

In summary, the method allows for the determination of the absorption coefficient and the dispersion as well as the relative strength of the absorption and frequency modulation, which is a characteristic feature of the QCL. The technique also provides an accurate method for measuring line profiles, which is particularly important for pressure broadening and pressure shift measurements.

V. Acknowledgement

This work was supported in part by the European Commission through the ProFIT program of the Investitionsbank Berlin. R.E. acknowledges support by the Helmholtz Research School on Security Technologies.

References

[1] H.-W. Hübers et al., "High-resolution gas phase spectroscopy with a distributed feedback terahertz quantum cascade laser," *Appl. Phys. Lett.*, vol. 89, pp. 061115-1−3, Aug. 2006.

[2] R. Eichholz et al., "Multi-channel terahertz grating spectrometer with quantum-cascade laser and microbolometer array," *Appl. Phys. Lett.,* vol. 99, pp. 141112-1−3, Oct. 2011.

[3] G.C. Bjorklund, "Frequency-modulation spectroscopy: a new method for measuring weak absorptions and dispersions," *Opt. Lett.*, vol. 5, no.1, pp. 15−17, 1980.

[4] H. Richter et al., "A compact, continuous-wave terahertz source based on a quantum-cascade laser and a miniature cryocooler," *Opt. Express*, vol. 18, pp. 10177–10187, May 2010.