

High-frequency modulation spectroscopy with a THz quantum-cascade laser

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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 a spectroscopy, because they are frequency tunable, 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 will 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 for 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 with a frequency up to 50 MHz to the QCL driving current. 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]. 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 lock-in amplifier with a bandwidth of 50 MHz.

III. RESULTS

An example of a line shape of the CH₃OH molecule at a pressure of 1 hPa measured with FM spectroscopy at 50 MHz is shown in Fig. 1. The experimental and theoretical 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 a +50 MHz in Fig. 1), 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. We have modeled the absorption by taking into account the simultaneous modulation of the amplitude and frequency modulation. This 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. This technique also provides a very accurate method for measuring line profiles, which is particularly important for pressure broadening and pressure shift measurements.

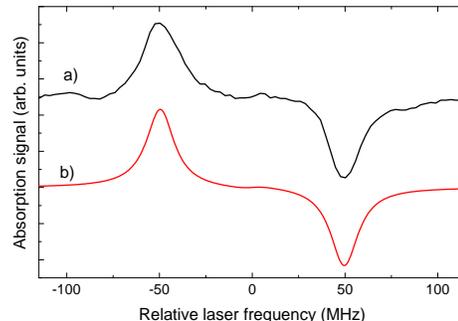


Fig. 1: (a) Experimental and (b) theoretical in-phase line shape for FM spectroscopy.

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