Molecular spectroscopy with a multimode THz quantum-cascade laser

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Abstract — **A terahertz absorption spectrometer for high-resolution molecular spectroscopy is realized. The spectrometer is based on a multimode quantum-cascade laser. The design and performance of the spectrometer are presented. Three aspects are discussed: sensitivity, frequency calibration, and frequency multiplexing.**

I. INTRODUCTION AND BACKGROUND

HIGH-resolution molecular spectroscopy is a powerful tool for investigations of the structure and energy levels of molecules and atoms. While in the low THz region many different methods have been developed, spectroscopy above 2 THz is hampered by the lack of frequency-tunable, continuous-wave, high-power, and narrow-linewidth radiation sources. THz quantum-cascade lasers (QCLs) are promising radiation sources for this type of spectroscopy, which was demonstrated with a distributed feedback (DFB) device [1]. Using a QCL with a Fabry-Pérot resonator instead of a DFB laser leads to a much more complex spectrum. When tuning the driving current of the QCL and thereby its frequency, all modes generate spectra simultaneously. Their superposition is difficult to interpret [2]. We will discuss the recent progress toward a THz spectrometer based on a multimode QCL.

In the QCL used for these experiments, efficient injection into the upper laser level is achieved by an intersubband transition resonant to the energy of the longitudinal optical phonon [3]. The laser has a single-plasmon waveguide and a Fabry-Pérot cavity with both facets uncoated. The laser is mounted in a compact air-cooled cryocooler (RICOR K535, for details see [2]). The emission spectrum of the QCL has several modes with a spacing of 31 GHz. The output power of the laser is 4 mW at a driving current of 600 mA and an operating temperature of 45 K. A small AC signal is superimposed onto the DC driving current. The beam emitted by the QCL is focused with a TPX lens onto the entrance slit of a monochromator. A 27-cm long absorption cell is placed between the lens and the monochromator. At the output of the monochromator, the first ($1f$) and second harmonic ($2f$) signal are detected with a Ge:Ga detector and a lock-in amplifier. Frequency tuning is achieved by adjusting the detection wavelength of the monochromator so that one particular mode is selected by the detector, while changing the QCL driving current of the QCL allows for fine tuning of the frequency. In order to increase the speed and frequency coverage of a QCL-based spectrometer, all modes emitted by the QCL should be detected simultaneously. Therefore, additional experiments with a microbolometer array, which replaces the Ge:Ga detector, are performed.

II. RESULTS

An example of a spectrum of $^{12}CH_3OH$ at a pressure of 1 hPa is shown in Fig. 1. The 4.5-GHz wide portion of the spectrum was measured with the Ge:Ga detector by tuning the laser driving current in steps of 0.25 mA, which corresponds to frequency steps of 5 MHz. The integration time for each spectral resolution element was 40 μs, resulting in a total integration time of only 36 ms for the whole spectrum. The sensitivity of the spectrometer can be determined by measuring an absorption line using amplitude modulation. This allows for a direct measurement of the absolute absorption. With an integration time of 1 s for each spectral resolution element, the signal-to-noise ratio of 20 to 100 for the absorption features in Fig. 1 corresponds to a fractional absorption (i.e. the fraction of absorbed power relative to the laser power) of approximately 10^{-5}.
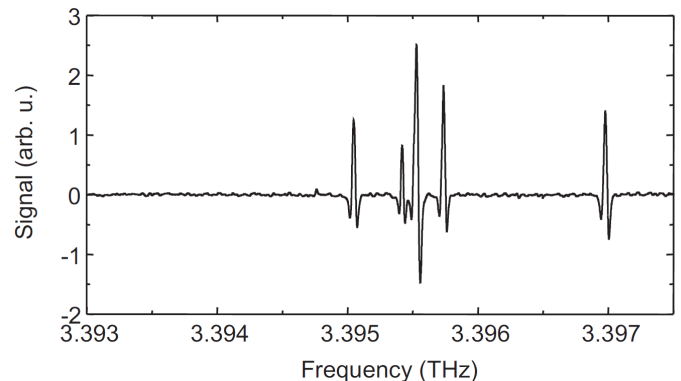

Fig. 1: Absorption signal ($2f$) of $^{12}CH_3OH$ at 1 hPa measured within 36 ms.

Using a microbolometer array, all modes emitted by the QCL can be detected simultaneously. This leads to a frequency multiplexing scheme as known from grating spectroscopy in the visible part of the electromagnetic spectrum, where the exit slit of the grating spectrometer is replaced by a linear array detector. Additional advantages are that such a spectrometer has no moving parts and that fast electrical frequency tuning can be implemented. To demonstrate the feasibility of this approach, a similar setup as described above is used. The main difference is that the Ge:Ga

detector is replaced by an infrared camera (InfraTec GmbH VarioCAM [4]). The core of the camera is a microbolometer array with 640×480 pixels. The size of the array is 16×12 mm^2 and the pixel pitch is 25 μm. The pixels are made from amorphous silicon. This camera is optimized for the mid-infrared spectral region of 8−14 μm. In order to use it at THz frequencies, the Ge lens was removed so that the THz radiation impinges directly on the array. The exit slit of the monochromator was removed as well. The resulting exit opening was 17 mm. This allows for the simultaneous imaging of five laser modes without any additional optical element between the monochromator and the microbolometer camera as shown in Fig. 2, which demonstrates that frequency multiplexing with a microbolometer array is feasible.

The experiments demonstrate the feasibility of frequency multiplexing using a microbolometer array. With an optimized monochromator, a dedicated optics, which images the output of the monochromator onto the microbolometer array, and an optimized QCL, it appears to be practicable to monitor several ten emission modes of a QCL simultaneously. Assuming a frequency tunability of approximately 10 GHz per mode by varying the QCL driving current, an instantaneous frequency coverage on the order of several 100 GHz may be achievable.
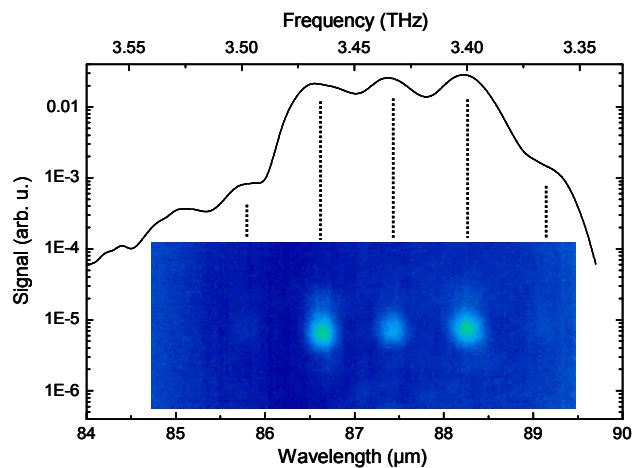
Fig. 2. Emission modes of the QCL measured with a grating monochromator and a Golay cell detector (solid line) and imaged with a microbolometer array (inset) at the output of the monochromator with its exit slit removed. Three strong and two weak modes are visible.

An example of a spectrum of $^{12}CH_3OH$ at a pressure of 1 hPa is shown in Fig. 3. The spectrum was obtained by integrating the intensity of one mode of the QCL detected by the microbolometer array. A lock-in amplifier, which is implemented into the data acquisition software of the array, detects the 1f signal of the integrated intensity. Obviously, the signal-to-noise ratio is less than that obtained with the Ge:Ga detector. This reflects the smaller sensitivity of the microbolometer array as compared to the Ge:Ga detector.
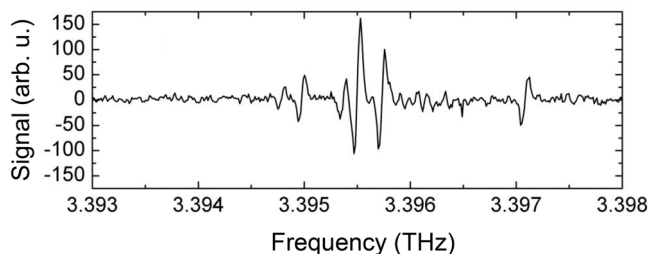
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Fig. 3: Absorption signal (1f) of $^{12}CH_3OH$ at 1 hPa measured with the microbolometer array.