HIGH-RESOLUTION MOLECULAR SPECTROSCOPY WITH 2.5 TERAHERTZ QUANTUM CASCADE LASERS

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Abstract- Quantum cascade lasers operating at about 2.5 terahertz have been implemented in a spectrometer for high resolution molecular spectroscopy. Linewidth, frequency tunability and frequency stability of the lasers were investigated by mixing their radiation with radiation from a 2.5 terahertz gas laser. Two first laser characteristics were found to be sufficient for Doppler-limited spectroscopy. For a demonstration of the spectrometer, absorption at rotational transitions of methanol was measured. The cascade lasers served as radiation source. A gas laser was used to determine the absolute frequency simultaneously with the detection of absorption signal by mixing the gas laser radiation with a part of that from cascade lasers. Amplitude as well as frequency modulation of the output power of the cascade lasers was used. The pressure broadening and the pressure shift of a rotational transition of methanol at 2.519 terahertz were determined.

Keywords: terahertz cascade lasers, molecular spectroscopy.

INTRODUCTION

Since the realization of a terahertz (THz) quantum cascade laser (QCL) in 2002 [1] several applications of these devices have been exploited. Among most thoroughly investigated ones are imaging in close-by configuration [2] and at stand-off distances [3], first attempts for implementation of the THz QCLs as local oscillator in a heterodyne receiver [4,5]. Beside this, such attractive features of the QCLs as narrow intrinsic linewidth of less than 20 kHz [5, 6] and high output power [7], can be used for high resolution molecular spectroscopy.

The THz-range molecular spectroscopy has a grand scientific importance. This is because many absorption and emission lines of astrophysical and atmospheric important molecules and atoms occur in the THz spectral region. Up to now more than one hundred molecules, such as CO and also more complex ones, have been detected in space [8]. On the ground, laboratory spectroscopy requires at least a $\Delta\nu/\nu\approx10^{-6}$ resolution at THz frequencies for thorough investigations of the structure and energy levels of molecules and atoms. With this spectral resolution, information on the species itself, on Doppler and pressure broadening can be obtained. Spectroscopy at above 2 THz is still suffer from the lack of frequency tunable, continuous wave, powerful, and narrow linewidth radiation sources. For the future astronomical and planetary missions, compact semiconductors sources such as THz QCLs are a promising alternative to bulky and consuming optically pumped THz gas laser used at present in laboratory heterodyne spectrometers.

In this paper we describe a spectrometer for high-resolution molecular spectroscopy in the THz region based on 2.5 THz quantum cascade lasers and first experimental results obtained for methanol.

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Experimental

Two QCLs designed for a frequency around 2.5 THz have been tested for operation in the spectrometer (Fig.1). Both are based on a GaAs/AlGaAs superlattice and a plasmon-type waveguide. The design follows the bound-to-continuum approach with a uniformly chirped superlattice (for more details, see Ref. 9). One laser operates on several (up to 5) modes. The second laser has a similar active medium but a distributed feedback (DFB) on its top surface formed by a series of slits with half-wavelength period. Both lasers are mounted on the cold finger of a mechanical cryo-cooler (1 W @ 4 K). Frequency tunability of the QCLs in the range of 5-6 GHz is reached by varying the laser drive current (Fig.2) in the range of 500-1000 mA. The tunability is slightly nonlinear and instantaneous laser frequency has fluctuations in the order of 20-30 MHz when measured during 1 min.

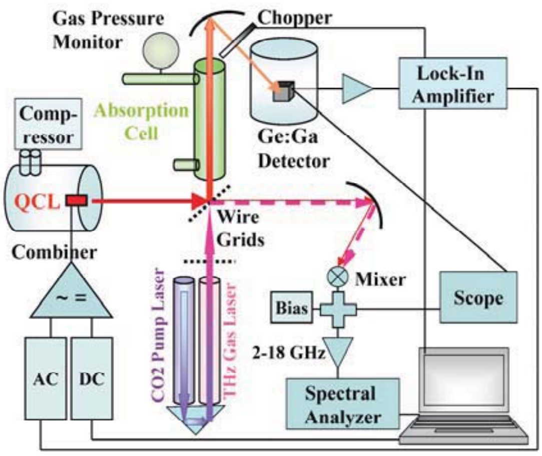

Fig.1. Sketch of a spectrometer with a THz QCL.

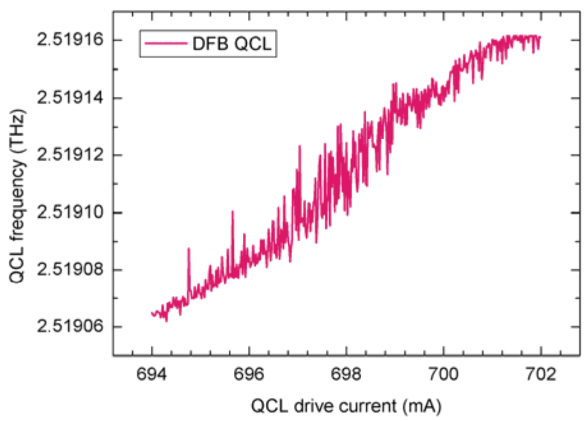

Fig.2. Current-induced frequency tunability of DFB QCL.

Since frequency band of the multi-mode Fabry-Perot QCL (~ 80 GHz) covers a few methanol transitions around 2.5 THz, analysis of the absorption spectra is not straight forward. Using a DFB single-frequency laser gives unambiguous identification of the transition frequency [10]. However, instantaneous determination of the QCL frequency performed by a mixing of part of the QCL radiation with a radiation of the 2.5227816 THz gas laser (Fig.1) is required. By these means the absorption spectra as a function of the QCL current (Fig.3a) can

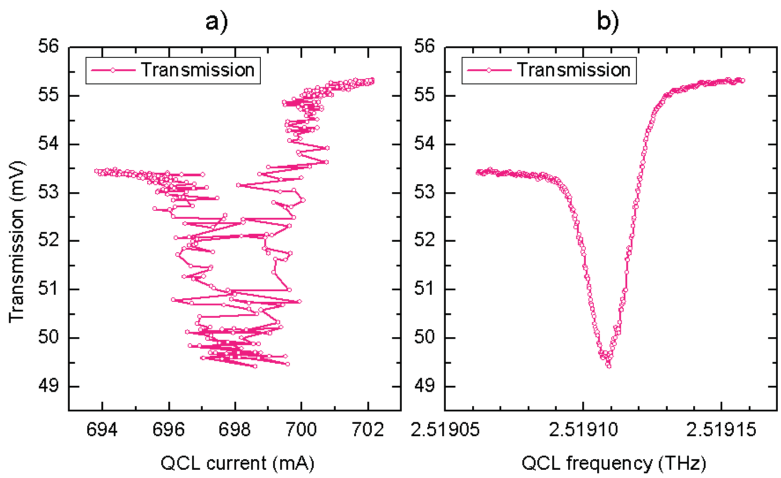

Fig.3. Typical absorption spectrum of methanol at 0.47 mbar pressure measured by a THz spectrometer: a) vs a QCL drive current, b) vs absolute QCL frequency determined simultaneously with a transmission signal.

be transformed in ones as a function of the QCL frequency. The center frequency of the absorption line was determined from least squares fits of a Voigt profile to the measured profile.

The self pressure broadening of 229(2) kHz/Pa of the methanol line was determined by measuring its profile at different pressures up to 1000 Pa and determining the full width at half maximum (FWHM). This is similar to pressure broadening coefficients of other methanol lines (e.g. 265.6(2) kHz/Pa at 76 GHz [11]). The pressure shift is determined as 10(2) kHz/Pa (6 kHz/Pa in Ref. [12]) and the zero pressure frequency is determined to be 2.519112(1) THz.

SUMMARY

We have demonstrated a THz spectrometer for high-resolution molecular spectroscopy based on QCLs as radiation source. Frequency calibration of the spectra is achieved by instantaneously measuring the frequency difference between the QCL and the THz gas laser. The frequency coverage of the spectrometer is limited by the tuning range of the QCL. The transition frequency, pressure broadening and pressure shift parameters of a methanol line at 2.5 THz measured using this spectrometer shows good agreement with other spectroscopic data.

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