High Resolution Spectroscopy with a Quantum Cascade Laser at 2.5 THz

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Abstract—**The quantum cascade laser (QCL) is a powerful, narrow linewidth, and continuous wave source of terahertz radiation. We have characterized a 2.5 THz distributed feedback QCL with respect to linewidth and frequency tunability. Both were found to be sufficiently good for high resolution gas phase spectroscopy. The QCL was implemented in a spectrometer for high-resolution gas phase spectroscopy where the absolute frequency of the laser was determined by mixing its radiation with the radiation of a 2.5 THz optically pumped gas laser. The absolute frequency as well as the pressure broadening of a rotational transition of methanol at 2.519 THz was measured. The results demonstrate that THz QCLs are well suited for either direct absorption or heterodyne high resolution spectroscopy.**

***Index Terms*—quantum cascade laser, spectroscopy, terahertz**

I. INTRODUCTION

HIGH resolution gas phase spectroscopy at terahertz (THz) frequencies is a powerful tool for investigations of the structure and energy levels of molecules and atoms. Besides information on the species itself, important information on Doppler and pressure broadening can be obtained from THz spectra. These data are a prerequisite for the interpretation of spectra obtained from astronomical sources or planetary atmospheres including the Earth [1]. 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, powerful, and narrow linewidth radiation sources. The recently developed THz quantum cascade laser (QCL) [2] has attractive features for gas phase spectroscopy, namely, its intrinsic linewidth of less than 20 kHz [3, 4] and high output power [5]. The goal of the work described in this paper is to characterize a distributed feedback (DFB) THz QCL with respect to linewidth and frequency tunability and to implement it into a THz spectrometer for high resolution gas phase absorption spectroscopy. The experiment also allows assessing the performance of the QCL when used as local oscillator (LO) in a heterodyne spectrometer.

II. DESIGN OF THE QCL

A distributed feedback (DFB) QCL has been used for the experiments described in this article. It is designed for an operation frequency at about 2.5 THz. The active medium of the laser is based on a GaAs/AlGaAs superlattice. The design follows the so-called bound-to-continuum approach [6] with a rather uniformly chirped superlattice and no marked distinction between the injection and lasing regions. The active medium is formed by 110 repeat units of the superlattice (total thickness 15 μm) covered on top by a Cr/Au layer. Between the ~250 μm thick substrate and the active medium is a highly doped GaAs layer. This layer has two doping concentrations: 2.7×10^{18} cm^{-3} in the 530 nm next to the superlattice and 2.6×10^{17}cm^{-3} in the 500 nm close to the substrate. By these means the boundary conditions at the two sides of the buried doped layer can be controlled separately. The resonator is a mesa-etched, 240 μm wide ridge with a length of 2.5 mm defined by cleaving. The top layer of the QCL is patterned into a series of narrow slits with half-wavelength period to create the DFB structure [7]. The laser is soldered to a copper bar, wire bonded, and mounted on a copper holder thermally coupled to a 4 K stage of a mechanical cryo-cooler. In order to minimize vibrations the laser holder is mechanically isolated from the 4 K stage by copper wires. The cooler has a heat extraction capacity of 1 W at 4 K. Since the input power of the laser is 5-10 W the smallest achievable temperature at the position of the QCL is ~20 K during laser operation. The laser threshold is about 80 A/cm^2 at 20 K, the maximum output power is about 6 mW, and the laser works up to 58 K in continuous wave. Due to the limited capacity of the cryo-cooler the operation temperature changes with current.

III. FREQUENCY CHARACTERIZATION OF THE QCL

The linewidth and frequency tunability of the QCL were measured by mixing the radiation from the DFB laser with the radiation from an optically pumped gas laser operating on the methanol emission line at 2.5227816 THz (pump line of the CO_2 laser: 9P36 [8]). The radiation from both lasers was superimposed by a wire grid and focused onto a GaAs Schottky

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diode with a quasi-optical 4λ corner cube antenna [9]. The difference signal was amplified and analyzed with a spectrum analyzer up to a maximum frequency of 18 GHz which was set by the available amplifiers. By these means the linewidth as well as the absolute frequency and the frequency tunability of the QCL as a function of current and temperature were measured. The experimental set-up is shown in Fig. 1.

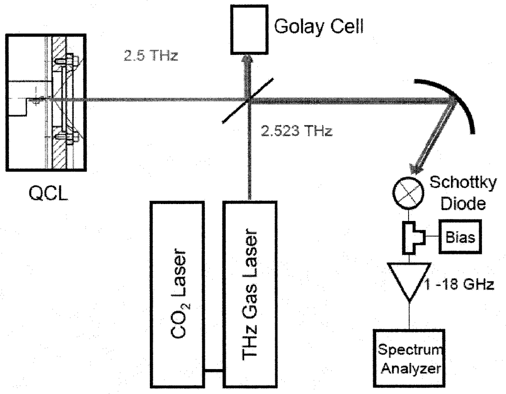

Fig. 1: Experimental set-up of the mixing experiments.

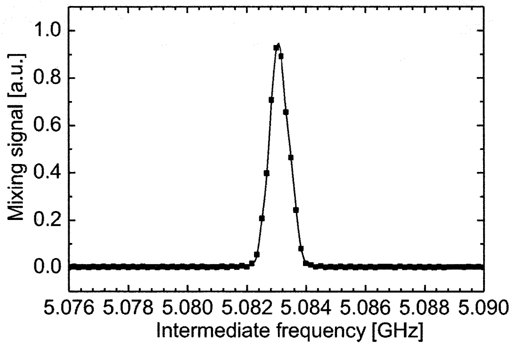

Fig. 2: Mixing signal of the QCL and the 2.5 THz gas laser.

The linewidth measured with an integration time of 5 s is shown in Fig. 2. The full width at half maximum (FWHM) is ~1 MHz. With an integration time of 0.3 s it reduces to 300 kHz. This is sufficient for many applications in high resolution gas phase spectroscopy. For example the Doppler limited linewidth of $^{12}CH_3{}^{16}OH$ is 5.5 MHz at 2.5 THz and 300 K. The tuning rate is almost linear with current (+8.0 MHz/mA, Fig. 3). The temperature related frequency tuning varies between - 20 MHz/K and - 100 MHz/K depending on current and temperature (Fig. 4). For both lasers the tunability is mainly caused by changes of the refractive index with temperature and current.

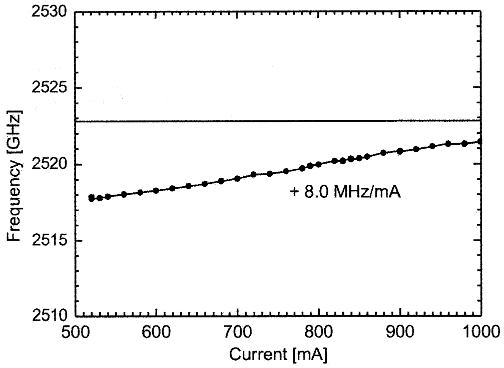

Fig. 3: Frequency of the QCL as a function of current (solid line: gas laser frequency).

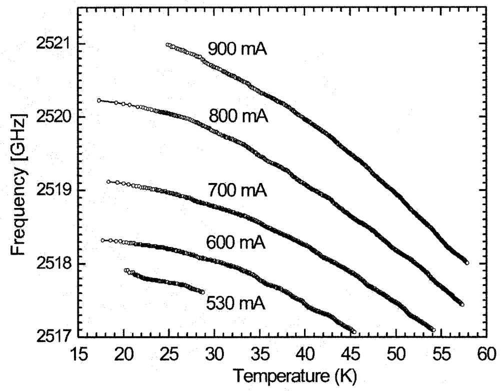

Fig. 4: Frequency of the QCL as a function of temperature for fixed current.

The set-up of the spectrometer is sketched in Fig. 5. For frequency tuning the temperature of the QCL is set and the current is swept in steps corresponding to frequency steps of 0.2 MHz to 2 MHz. The radiation from the QCL is reflected by a wire grid into a 0.5 m long absorption cell. The cell was equipped with two polyethylene windows and the pressure was measured with a capacitance manometer. For each measurement the cell was filled with methanol gas at a certain pressure and sealed off. The transmitted radiation is collimated by an off-axis parabolic mirror and detected with a Ge:Ga photoconductive detector. The transmitted radiation was mechanically chopped in front of the detector and detected with a lock-in amplifier. A small part of the radiation from the QCL is transmitted through the wire grid. This is superimposed with the radiation from the gas laser operating on the 2.5 THz line. At the output of the gas laser a grid with wires oriented perpendicularly to the wires of the first grid was used to define the polarization in a way that it is reflected by the first grid. The radiation from both lasers is focused onto a GaAs Schottky diode. The signal at the difference frequency is amplified and its frequency is measured with a spectrum analyzer. This is especially important because the frequency tuning of the QCL is not linear with current. In total a frequency range from 2.517 THz to 2.521 THz is available.

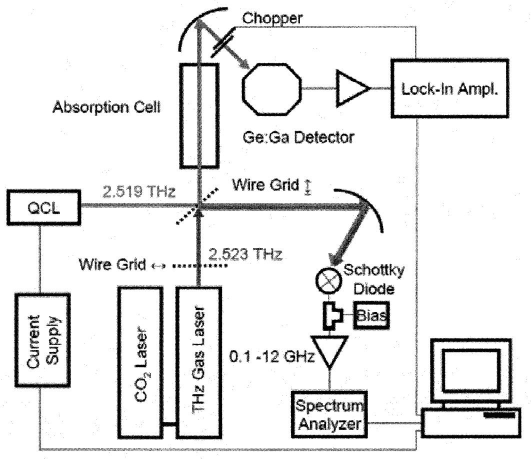

Fig. 5: THz spectrometer for high resolution gas phase spectroscopy.

V. RESULTS

Fig. 6 shows the absorption line measured with the QCL along with a Voigt profile fitted to the measured profile. As can be seen the agreement is very good. The center frequency of the absorption line is 2.519112(1) THz. This agrees well with published data measured with a Fourier transform spectrometer (2.519107(2) THz [10]). The pressure broadening 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). A least squares fit to the FWHM data yields a pressure broadening coefficient of 229(2) kHz/Pa (Fig. 7). This is similar to pressure broadening coefficients of other methanol lines (265.6(2) kHz/Pa at 76 GHz [11] and 290 kHz/Pa at 2.524 THz [12]).

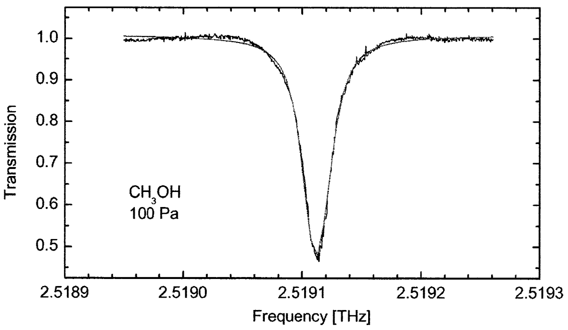

Fig. 6: CH$_3$OH absorption spectrum measured at 100 Pa. The solid line is a fit of a Voigt profile to the absorption line.

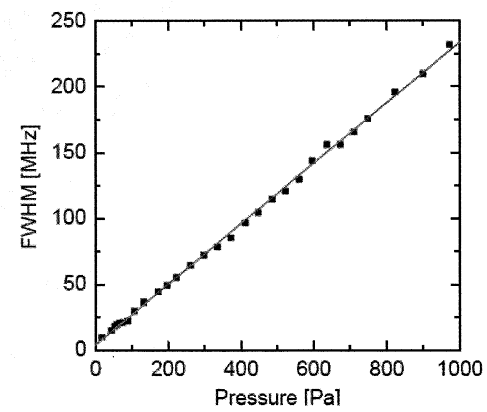

Fig. 7: FWHM of the CH$_3$OH rotational transition as a function of pressure.

VI. SUMMARY AND CONCLUSIONS

In summary, the linewidth of a DFB QCL operating at 2.5 THz as well as its tuning rates as a function of current and temperature have been determined. Based on these results, a THz spectrometer for high-resolution gas phase spectroscopy with a QCL as the radiation source has been realized. Frequency calibration of the spectra was achieved by instantaneously measuring the frequency difference between the QCL and a THz gas laser. We have measured the transition frequency and pressure broadening of a methanol rotational transition at 2.5 THz. The comparison of our data with other spectroscopic data shows good agreement. The results show that QCLs are very promising radiation sources for high resolution absorption spectroscopy. In a heterodyne receiver the frequency resolution is to a large extent determined by the linewidth and frequency stability of the LO. Our results show that the QCL performance is sufficient also for high resolution heterodyne spectroscopy.

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