

# Heterodyne gas cell measurements at 2.9 THz using a quantum cascade laser as local oscillator

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## ABSTRACT

High-resolution heterodyne spectrometers operating at above 2 THz are crucial for detecting, e.g., the HD line at 2.7 THz and oxygen OI line at 4.7 THz in astronomy. The potential receiver technology is a combination of a hot electron bolometer (HEB) mixer and a THz quantum cascade laser (QCL) local oscillator (LO). Here we report the first high-resolution heterodyne spectroscopy measurement of a gas cell using such a HEB-QCL receiver. The receiver employs a 2.9 THz free-running QCL as local oscillator and a NbN HEB as a mixer. By using methanol (CH<sub>3</sub>OH) gas as a signal source, we successfully recorded the methanol emission line at 2.92195 THz. Spectral lines at IF frequency at different pressures were measured using a FFTS and well fitted with a Lorentzian profile. Our gas cell measurement is a crucial demonstration of the QCL as LO for practical heterodyne instruments. Together with our other experimental demonstrations, such as using a QCL at 70 K to operate a HEB mixer and the phase locking of a QCL such a receiver is in principle ready for a next step, which is to build a real instrument for any balloon-, air-, and space-borne observatory.

**Keywords:** THz quantum cascade laser, superconducting HEB mixer, heterodyne spectroscopy

## 1. INTRODUCTION

High-resolution spectroscopy, as a technology to study the rotational lines from ions, atoms and molecules, plays a vital role in atmospheric and astronomical research at terahertz frequency in space. Heterodyne spectroscopy based on superconducting heterodyne receiver provides the most promising performance for its nearly quantum limited sensitivity and ultra high spectral resolution ( $\nu/\Delta\nu > 10^6$ , where  $\nu$  is the frequency). A superconducting HEB mixer has shown a superior sensitivity up to 5.3 THz [1]. Availability of suitable local oscillator sources (LOs) above 2 THz becomes the only limiting factor for the future applications. Thanks to the recently developed terahertz QCLs [2], we have a promising candidate. THz QCLs, compared with other sources such as solid state THz sources based on multipliers and optically pumped gas lasers, they show advantages with regard to the output power, frequency coverage, compactness, and power consumption. Until now, THz QCL has demonstrated performances such as covering almost the whole THz frequency range [3], peak emission power over 100 mW [4] and maximum operating temperature 190 K in pulse mode [5]. It has been proved to be a suitable source for various applications such as sensing, imaging [3], gas phase spectroscopy [6], and LO for a heterodyne receiver [7,8]. Besides, THz QCL has exhibited excellent power stability [7], phase-lock capability [9], and narrow intrinsic linewidth [10], which meet essentially the requirements as a LO. Many experiments have been reported to demonstrate the suitability of a QCL as LO, however, no truly heterodyne spectroscopic measurement using a QCL as LO has been reported until now. In this paper we present the first high resolution spectroscopic measurement at 2.9 THz using a gas cell containing methanol and using a double sideband heterodyne receiver based on a THz QCL as LO and a NbN HEB as mixer.

## 2. THz QCL

The QCL used in this experiment is a metal-metal waveguide QCL, developed by Qing Hu's group at MIT. It is based on resonant phonon scattering as described in reference [11]. The active region contains 176 GaAs/Al<sub>0.15</sub>Ga<sub>0.85</sub>As quantum well modules. The 40 $\mu$ m wide ridge waveguide was cleaved at both ends to form a 1.18mm long Fabry-Perot cavity, and it is fabricated using a copper-to-copper thermocompression wafer bonding technique. The QCL is indium soldered on a copper mount and is mounted on the cold stage of a helium-flow cryostat. As shown in Figure 1, the QCL emits a single-mode emission line at 2.918THz in continuous wave mode. The spectrum was measured using a Fourier-transform Spectrometer (FTS) [12]. Latter has a spectral resolution of 0.7GHz, which is much larger than the intrinsic linewidth of the QCL, typically in the range of 6-30KHz. In the current measurement, we used a free-running QCL (namely without any stabilization on the phase/frequency and the amplitude), Based on the previous experience [9], due to the jitter we expect a linewidth of less than 1MHz in practice. The QCL provides a maximum output power of 1mW in the CW mode and at a temperature of 30K. Although the laser can be operated up to about 100K, in our experiment we operated the laser at the lowest temperature ( $\sim$ 30K).

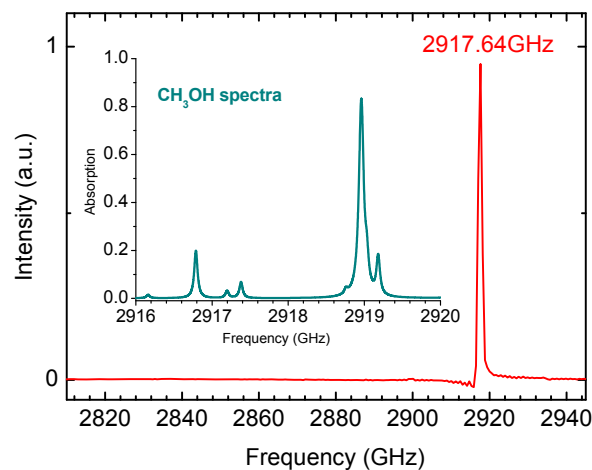


Fig. 1 Measured emission spectrum of a THz quantum cascade laser biased at 16.44V at a bath temperature around 30K in CW mode. It is measured with a FTS with a resolution of 0.7GHz. The inset shows simulated methanol (CH<sub>3</sub>OH) spectrum [15] for a 0.5m optical path length and gas cell pressure of 5.9mBar between 2916 and 2920GHz.

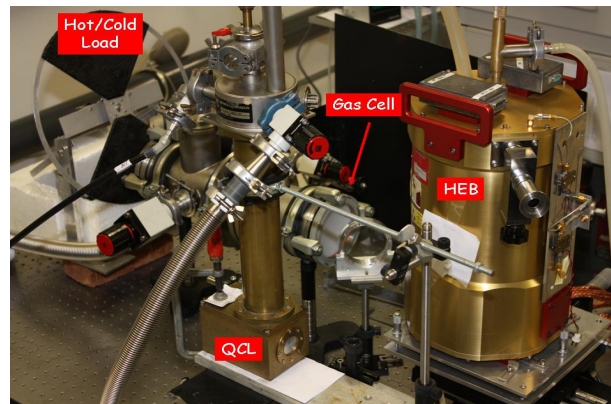
## 3. HEB MIXER

We use a NbN HEB mixer, which consists of a 2 $\mu$ m wide, 0.2 $\mu$ m long, and 5.5nm thick NbN bridge [13]. The bridge is connected with a tight winding spiral antenna, which is made of a 170nm thick Au layer, and has an inner diameter of 6.6 $\mu$ m. This special design expands the cut off frequency up to 6THz [14]. The HEB has a low-temperature normal-state resistance ( $R_N$ ) of 83 $\Omega$ , a critical temperature of 9.3K, and a critical current of 210 $\mu$ A at 4.2K. In a separate experiment [1], the mixer has shown superior sensitivities across the frequency range of 1.6-5.3THz, from which the receiver noise temperature ( $T_{N,rec}$ ) around 2.9THz is expected to be 1000K [1].

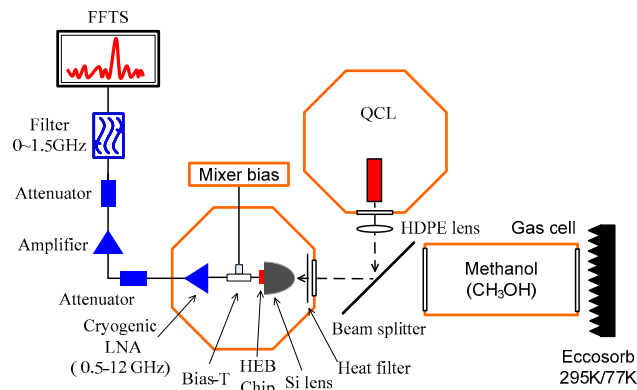
## 4. MEASUREMENT SETUP

The spectroscopic measurement setup is shown in Figure 2. We use two separate L-He cryostats for QCL and HEB mixer, respectively. For coupling the THz radiation from free space to the antenna of the HEB mixer, we used an elliptical silicon lens without anti-reflection coating. The lens is placed in a metal mixer unit, which is mounted on the 4.2K cold plate of the cryostat. The THz radiation beam from the QCL firstly passes through the high-density polyethylene (HDPE) window of the QCL's cryostat and then is focused with a HDPE ( $f=26.5$ mm) lens. As signal

source, it is a combination of a gas cell and hot/cold blackbody loads. The gas cell is a 50cm long cylinder with an inner diameter of 10cm (at room temperature). It has two HDPE windows with a thickness of 1.5mm at two ends of the gas cell. The blackbody load is defined as a hot load at 295K and as a cold load at 77K. The gas we used is methanol ( $\text{CH}_3\text{OH}$ ), from which a few absorption lines are expected in the vicinity of the LO frequency [15]. The expected spectral lines with different strength are plotted in the inset of figure 1. By applying three valves for methanol gas (two) and vacuum pump (one), the gas pressure inside the cell can be controlled relatively accurately. The gas pressure in the cell is measured by a gas-independent gauge [16]. The methanol emission lines and the QCL signal are combined by a  $3\mu\text{m}$  thick Mylar beam splitter and are fed further into the HEB mixer. The intermediate frequency (IF) signal is amplified first using a wide band low noise amplifier (0.5-12GHz) at 4.2K, and then followed by a room-temperature amplifier with 13dB attenuator in between. As back-end spectrometer, we used a Fast Fourier Transform Spectrometer (FFTS) [17], which is developed by MPIfR in Bonn. This FFTS provides 8192 channels for the frequency range of 0 to 1.5GHz, and thus gives a spectral resolution of 183KHz. In order to match the optimal input signal intensity required by the FFTS a 6dB attenuator is employed, and to avoid aliasing effect a 1.5GHz low pass filter is added before the FFTS.



(a)



(b)

Fig. 2 Picture (a) and Schematic view (b) of the heterodyne gas cell measurement setup.

## 5. RESULTS

Before we perform spectroscopic measurement, we calibrated the sensitivity of the whole system at the optimal operating point (bias and LO power) for the HEB mixer. We measured first the receiver noise temperature for the case where the hot/cold loads are positioned just after the beam splitter, and found it to be 2500K. When the hot/cold loads are positioned after the gas cell, which is evacuated, the receiver (system) noise temperature increases to 3800K. The difference between these two can be attributed to the loss due to two windows ( $2 \times 0.9\text{dB}$ ) of the gas cell and the

additional loss due to the air in the optical pass. The value of 2500K is higher than other measurement [1], that can be attributed to the losses due to the window of HEB cryostat and the air, to non-optimised IF chain due to the use of the wide band low noise IF amplifier, and to the direct detection effect. To measure the spectral lines of methanol, we used the same method as described in reference. 18. We record three IF noise power traces using the FFTS, which are: 1) the spectrum  $S_{emp77}(f)$  when the cold load is behind the evacuated gas cell; 2)  $S_{gas77}(f)$  when the cold load is behind the filled gas cell; 3)  $S_{gas300}(f)$  when the hot load is behind the filled gas cell. All three traces are recorded when the HEB is operated at its optimal operating point. Each trace is measured by the FFTS with an integration time of 5 second. The brightness of the methanol emission lines in terms of temperature is calculated according to the following expression:

$$T_{gas}(f) = T_{cold} + 2 \cdot (T_{hot} - T_{cold}) \frac{S_{gas77}(f) - S_{emp77}(f)}{S_{gas300}(f) - S_{emp77}(f)}$$

(1)

where all the temperatures are the effective temperatures defined by the Callen-Welton form [19] because of the THz frequency, and a factor of two is derived from the double sideband operation of the HEB mixer.

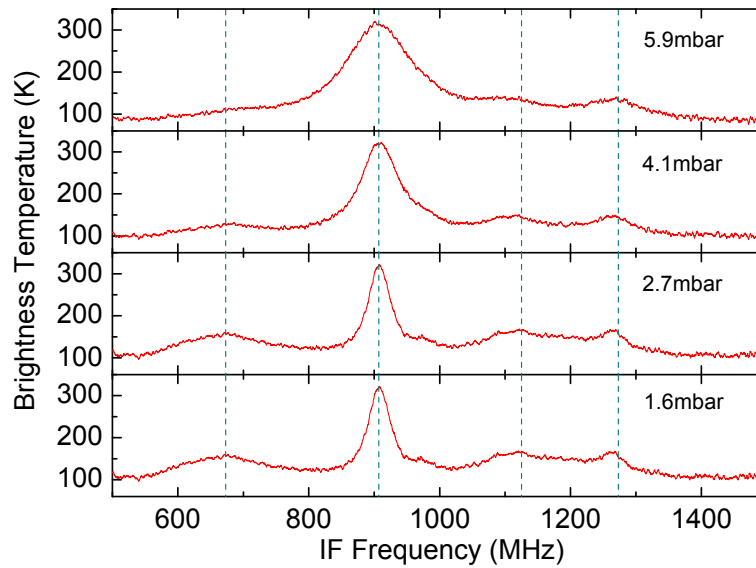


Fig. 3 Measured line spectra of methanol ( $\text{CH}_3\text{OH}$ ) gas within the IF range between 0.5 and 1.5GHz. The QCL's frequency is 2918.053GHz and the gas cell pressure varies from 1.6mBar to 5.9mBar. The dash line indicates positions of the theoretical simulated methanol lines.

By varying the gas pressure we are able to measure the methanol spectral lines at different pressures. Figure 3 shows such a set of spectra within the IF frequency range between 0.5 to 1.5GHz and with increased pressures varying from 1.6mBar to 5.9mBar. A relatively strong methanol emission line is found at an IF frequency of 907MHz, which is assumed to be the methanol line at 2918.96GHz in the upper sideband (USB) with respect to the LO frequency, as shown in the inset of figure 1. Since our receiver measures double sideband, we should see lines from both the upper side band and lower side band (LSB). Three relatively weak lines, as shown in the inset of figure 1, were also successfully detected. Those are at 673MHz (lower sideband, LSB), 1125MHz (USB) and 1273MHz (LSB), respectively. Having done such a comparison, we can also derive the QCL's frequency that is 2918.053GHz, which agrees well with the frequency measured by FTS.

Since the frequency of the QCL can be tuned slightly by varying its bias voltage, we can make use of this to have an additional check for the correspondence between the expected spectral of methanol and the measured ones. As shown in figure 4, the measured spectra are indeed varied by changing the bias of the QCL. A detailed analysis shows that all those spectra correspond well to the predicted emission lines in the inset of figure 1. We find a tuning coefficient for this specific QCL to be about 550MHz/V. It is known that this is a compromise between the "red shift" caused by the thermal tuning and the "blue shift" due to the Stark effect by the bias [9].

We have fitted the measured strong line at 907 MHz using a Lorentzian line profile. The result is shown in figure 5, the reasonable fitting quality suggests that measured line profile is Lorentzian. Theoretically, besides the pressure broadening, there are two main factors to determine the intrinsic methanol line profile. One is natural broadening and the other is Doppler broadening. The natural broadening process and pressure broadening both will give a Lorentzian line profile. For the second case, the Doppler broadening effect will impose a full width at half maximum (FWHM) of 200 KHz and should give a Gaussian line profile. Obviously this does not apply to our experiment. So in our case we should expect a Lorentzian line profile and this is what we saw experimentally.

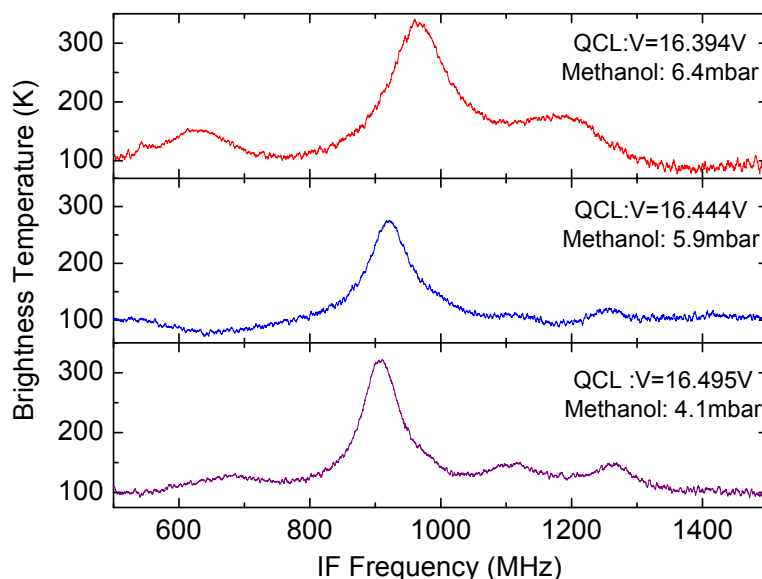


Fig. 4 Line spectra of methanol ( $\text{CH}_3\text{OH}$ ) gas within the IF range between 0.5 and 1.5GHz measured with different the bias voltage to the QCL. The different bias varies the QCL frequency a bit. Although the pressure is slightly different, the key point is the change of line positions.

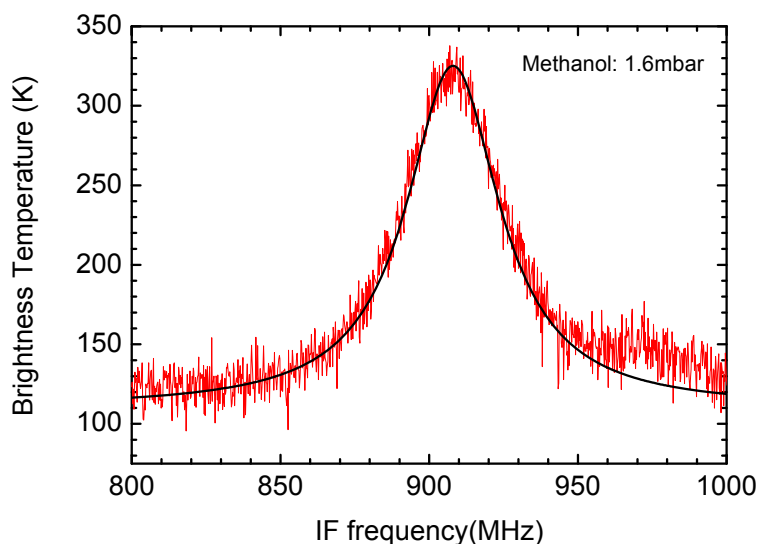


Fig. 5 Measured line spectra of methanol ( $\text{CH}_3\text{OH}$ ) gas at 2918.96GHz, where the gas cell pressure is 1.6mBar. Also shown is a Lorentzian profile fitting.

Using the strong line measured at pressures from 1 to 6.5mBar, we can determine the pressure broadening coefficient by plotting FWHM as a function of the methanol pressure. We find it to be  $15 \pm 2$  MHz/mBar. This value is considerably lower than what expected by the model [15]. One hypothesis is that we might have leakage of the air into the gas cell. Because of the leakage, we are able to obtain the exact methanol spectra, but not correct pressure broadening coefficient. By the time we are writing this paper, we have discovered the leakage in one of the valves in our setup, which controls methanol into the gas cell. With the improved setup we have taken new methanol spectra successfully. However, a detailed analysis of the data is needed and is also planned.

## 6. SUMMARY

In conclusion we have for the first time succeeded in demonstrating high-resolution heterodyne spectroscopic measurement using a double side band heterodyne receiver based on a THz quantum cascade laser as local oscillator and a NbN HEB as mixer. The QCL is operated in a free running mode and emits LO signal at 2918.053GHz. Within an IF bandwidth of 0.5 - 1.5GHz we observed 4 methanol lines. One of them is a relatively strong line at 2918.96GHz. Our gas cell measurement is a crucial demonstration of the QCL as LO for practical heterodyne instruments. Together with other recent progresses, such as the wide tuning range of THz QCL ( $\sim 100$ GHz) [20], phase locking [9], and pumping a HEB mixer when a QCL is operated at 60-70K [21], THz QCL is in principle ready for building a real instrument at the super-THz frequencies.

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