Frequency locking of a 3.5 THz quantum cascade laser using a gas cell

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***Abstract*—We report a frequency locking experiment of a 3.5 THz third-order distributed feedback quantum cascade laser (QCL) by using a molecular absorption line of methanol (CH_3OH) gas. With the help of the absorption line, the frequency noise of the QCL is transformed into measurable amplitude fluctuation. We first present the study of the noise of the THz QCL with the contribution from both the frequency and amplitude domain, by using a NbN superconducting hot-electron bolometer as a power detector. We then present the frequency locking measurement with a lock-in amplifier registering the derivate curve of the absorption line and a proportional-integral-derivative (PID) controller generating the feedback signal. The linewidth of the QCL in the free-running mode was found to be around 900 KHz. This linewidth is reduced to below 17 KHz (full width at half maximum) with a Gaussian-like shape when the control loop is active. Because of the frequency stabilization the noise power spectral density of the QCL shows a reduction of more than 20 dB at frequencies below 30 Hz.**

***Index Terms*—terahertz, quantum cascade laser, frequency stabilization, heterodyne, and local oscillator**

I. Introduction

In the terahertz (THz) frequency range, a high resolution heterodyne spectrometer is of crucial importance for astronomical observation and atmospheric remote sensing applications, based on its high spectral resolution and superior sensitivity. As the mixer, a superconducting NbN hot-electron bolometer (HEB) mixer has demonstrated excellent sensitivity up to 5.3 THz [1]. As the local oscillator (LO), terahertz quantum cascade lasers (QCLs) [2] have shown advantages at frequency above 2 THz, based on their single mode emission, wide frequency operating range, high output power and long term stability. Several progresses have been made for a THz QCL to be used as the local oscillator (LO) in a heterodyne receiver, including a noise temperature measurement [3,4] and a heterodyne spectroscopy measurement [5,6]. Since the spectral resolution is governed by the back-end spectrometer as well as the LO, a narrow linewidth LO is demanded for a high resolution heterodyne spectrometer, especially for application in astronomical observation, where atomic and molecular gases are at lower pressure and lower temperature, resulting in narrow spectral linewidths. For a quantum cascade laser, due to its fast non-radiative process, which strongly suppress the noise associated with spontaneous emission, the intrinsic linewidth can be as narrow as 1~10 KHz [7,8]. However, due to external jitters, which is caused by noise from the bias current source and temperature fluctuations, a practical linewidth for a free-running THz QCL is much larger, typically is 1 MHz or larger within 1 sec integration time [9,10].

Phase locking a QCL to a reference source means to control the phase of the laser radiation field precisely [11], which is to stabilize the frequency and to transfer the line profile of the reference to the laser. In the case of frequency-locking, the QCL's average frequency is fixed, but its linewidth remains equal to its intrinsic linewidth. Recently, several demonstrations have been given to lock a THz QCL to an external signal source [10, 12], where both frequency and phase of the laser are stabilized. However, in this way, additional signal source and mixer are required, which complicates the instrument and is difficult to implement especially at frequency above 3 THz because of the lack of suitable reference sources. Recently an alternative stabilization scheme for a THz QCL is demonstrated based on a molecular resonance serving as the frequency reference [13], by additionally using only a power detector and a gas cell. In this approach, a liquid-He cooled detector was applied and a locked linewidth of 300 KHz was achieved. It is worth noting that this locked linewidth is still much larger than the intrinsic linewidth of a THz QCL, which indicates that this technique was not fully understood yet.

Here we report our frequency stabilization measurement of a 3.5 THz quantum cascade laser to a molecular absorption line. We used a superconducting NbN HEB as power detector.

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The derivate curve of the absorption profile was used as the reference signal for frequency locking. We report, in comparison with the work in Ref. 13, the frequency locking experiment not only for a much higher frequency QCL, but also achieving a much narrower locked linewidth.

II. MEASUREMENT SETUP

The measurement setup is described schematically in Fig.1. A single mode, third-order distributed feedback (DFB) QCL [14,15] emitting at 3.5 THz is used. As demonstrated in Ref.6, by using the third-order periodic structure with strong refractive index contrast gratings, not only can single mode emission be achieved, but also a single spot, less divergent far-field beam is obtained. Furthermore, by electric tuning the bias voltage of the QCL, a tuning range of 1 GHz was achieved for the 3rd DFB QCL as a LO in a heterodyne spectroscopic measurement. The laser, based on a 10-µm thick active region, consists of 27 periods of gratings with a total length of 1070 µm. The QCL is mounted on the second stage of a pulse tube cryocooler without further temperature stabilization, where the temperature rises up to 12 K and stays at the same value during the laser operation. The beam from the QCL passes through a high-density polyethylene (HDPE) window of the cooler and is focused with a HDPE lens (f=26.5mm). The beam is then guided through a gas cell by a 13-µm thick Mylar beam splitter. The gas cell is a 41-cm long cylinder at room temperature and has two 2-mm thick HDPE windows. Methanol (CH_3OH) gas is chosen since it contains abundant absorption lines in the terahertz frequency range. The transmitted signal is detected by a superconducting NbN hot-electron bolometer working at liquid helium temperature as a direct power detector whereby we benefit from a low NEP (10^{-12}-10^{-13} W/Hz$^{1/2}$ [16]) and high speed of response (40 psec). The absorption spectrum is measured while the HEB is operated with a constant bias voltage, where the current level of the HEB is a measure of the transmitted THz power through the gas cell.

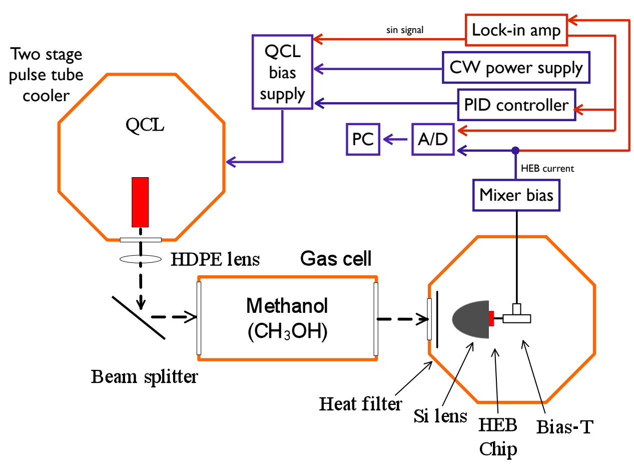


Fig. 1. Schematic view of the frequency locking experiment setup

For the bias circuit of the QCL, a summing circuit is used. It combines three input signals to independently control the bias voltage of the laser. The first input is the DC bias supply for the laser, which is used to set the DC operating point for the laser. The second input is an AC modulation signal and the third one is the control signal from the feedback loop. A sinusoidal modulation signal at around 1 KHz with relative small amplitude (< 0.1% compared with the DC bias point for the laser) was used. By feeding the HEB current to a lock-in amplifier, the derivative signal of the absorption spectrum is obtained. A proportional-integral-derivative (PID) controller is used to actively lock this derivative signal to absolute zero value, and a compensate signal is fed back to the bias circuit of the QCL. By doing this, the QCL frequency could be stabilized at the central frequency of the absorption line.

III. MEASUREMENT RESULTS

A. Characterization of the QCL's frequency noise

As shown in Fig.2, methanol absorption lines were observed by making direct absorption spectroscopic measurement [17]. By increasing the bias voltage of the QCL, the laser's emission power gets higher, as a result the HEB current decreases. Since the QCL voltage determines the emitting frequency, the frequency of the QCL decreases with the increase of the bias voltage. By sweeping a frequency range of 1 GHz, two methanol absorption lines at around 3.5 THz were observed.

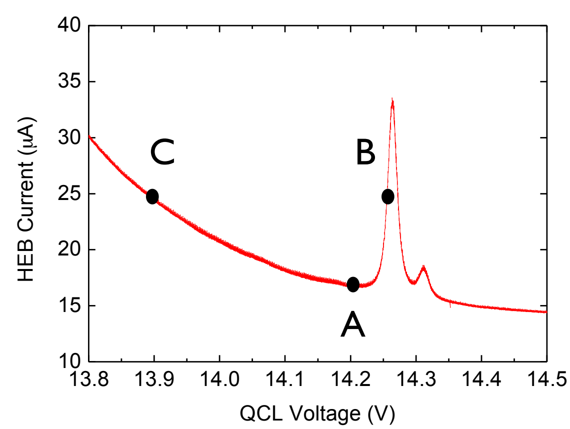


Fig. 2. HEB current measured as a function of QCL voltage that varies QCL frequency. The methanol gas is at 1.1 mbar. The points A, B and C indicate the measurement points for monitoring the HEB current variation, where point A and C contain the contribution from only the HEB noise and the laser intensity noise, but point B contains the HEB noise, laser intensity noise and also laser frequency noise contribution.

In order to study the frequency noise of the QCL, we monitor the HEB current fluctuation at different working points as shown in Fig.3. The measured current within 10 seconds is plotted and also the current noise power spectral density is calculated for each case. The intrinsic HEB current data were measured when the HEB detector was operated at a high voltage bias (3 mV) without LO power, where the HEB detector behaves like a normal resistor. From the noise power spectral density result, we observed the contribution from the laser amplitude noise as well as from the laser frequency noise for different working points. As shown, the extra noise for point A and C arises from the laser amplitude noise, which locates mainly at the frequencies lower than 100 Hz. While for point B, the extra noise represents the contribution from the QCL frequency noise, and it contributes up to 1 KHz. Please notice that the roll-off of the spectral density above 1 KHz is due to a low pass filter used for the HEB current readout circuit.

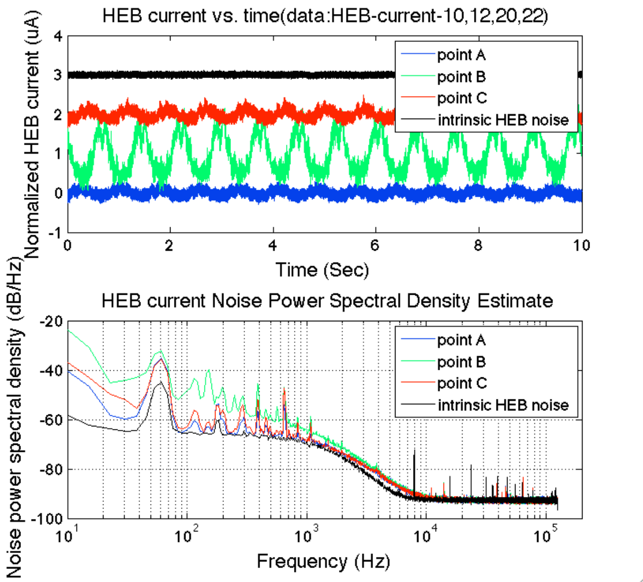


Fig. 3. HEB current measured at different working points as a function of time within 10 sec, and the absolute value is normalized. The lower plot shows the current noise power spectral density of the measured HEB current for each working point.

B. Frequency stabilization by using a methanol absorption line

Fig.4 shows the measured methanol absorption line at 3.5 THz, and a derivate signal from the lock-in amplifier. Around the absorption peak point, the linear range of the error signal indicates the feedback locking range. The error signal is used for the PID control loop, where a feedback signal is generated and fed to the bias circuit of the QCL in order to yield a stabilized error signal.

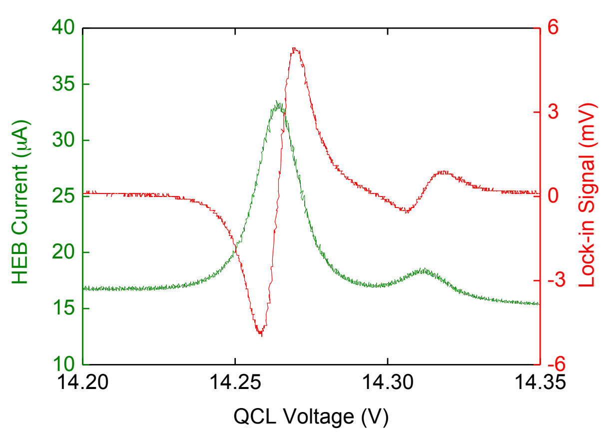
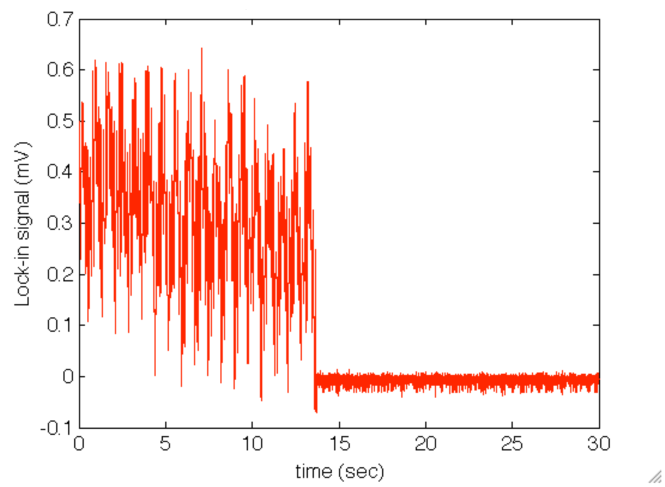
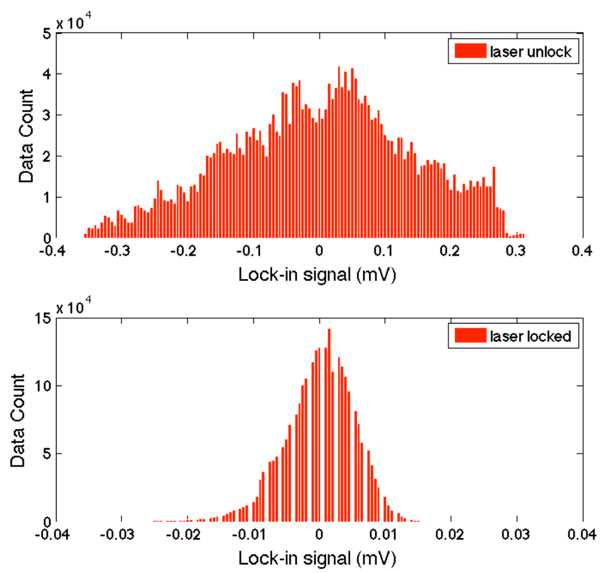


Fig. 4. Absorption profile of the methanol absorption line at 1.1mbar and the error signal was measured using the lock-in amplifier. The voltage window (linear range for the lock-in signal) used for the lock-in loop is around 8mV.

The laser frequency stabilization result is demonstrated in Fig.5. For the unlocked state, the 1 Hz variation is related to the temperature fluctuation and mechanical vibration of the pulse tube cooler. And the long term drifting is due to the non-stability of QCL emission power while operating in the cryocooler. After the PID feedback loop is in active, the error signal is well stabilized to zero value. Also, the error signal data variation is plotted in Fig.5b for both the unlocked state and locked state for the laser during a 10 sec integration time. It is shown that the error signal data variation after the frequency locking is improved by a factor of 55. Furthermore, from the noise power spectral density of the error signal as shown in Fig.6, a noise suppression of over 20dB is achieved below 30 Hz. This relatively small bandwidth is due to the 30 millisecond time constant used in the lock-in amplifier. A further improvement can be done by increasing the bandwidth of the entire feedback loop bandwidth.



a



b

Fig.5 (a) error signal in the unlocked and locked state measured as a function of time. (b) error signal value distribution in the unlocked and locked state during 10 sec integration time.

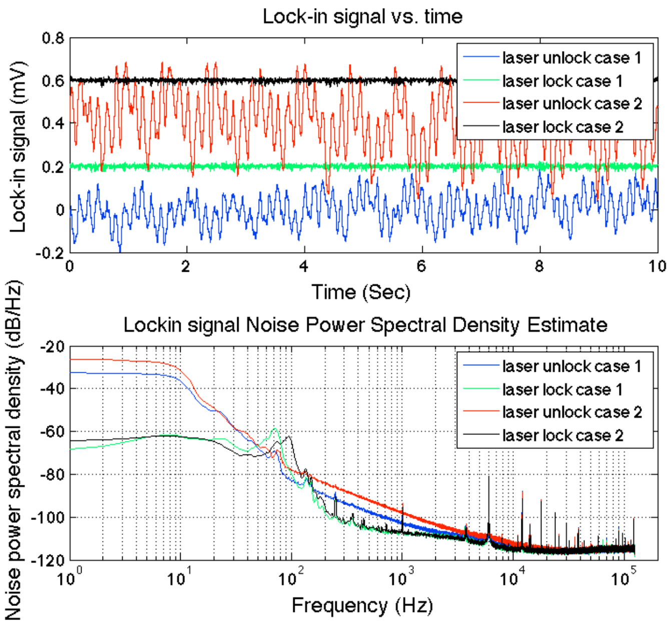


Fig.6 Noise power spectral density of the error signal for two unlocked and locked states, where different P.I.D parameters are used for the feedback loop.

C. *Laser linewidth estimation*

The laser linewidth after frequency-locking is estimated by transforming the variation of the error signal into the frequency domain. The error signal is transformed into the QCL bias voltage variation using the linear relationship between these two signals as shown in Fig.4. To do so, two different methods were used to calculate the correspondence between QCL frequency and the bias voltage. First, from the previous heterodyne spectroscopic experiment [6], the frequency tuning coefficient of this QCL at different operating points was obtained. In this way, we calculated a free running laser linewidth within the range of 443-962 KHz, and when the laser is locked, the linewidth in a range of 8-17 KHz. The uncertainty in the linewidth is caused by the uncertainty in the frequency tuning coefficient of the QCL, which is bias point dependent. Secondly, by knowing the methanol gas pressure broadening coefficient at 3.5 THz [6], a sets of methanol absorption lines at different gas pressure were measured. In this way the error signal is also transformed to the frequency domain. With this method, we obtained a free running laser linewidth of 558-798 KHz and the linewidth of 10~14 KHz for the locked state. In the 2nd case, the linewidth uncertainty is determined by the accuracy of the gas pressure calibration. It is really important to note that although we applied two different approaches, we obtained similar linewidth results.

As been found in Ref. 13, the temperature fluctuation of the cryocooler will also introduce the linewidth broadening to the QCL, since the emission frequency is related to the bath temperature of the laser. We monitored the bath temperature fluctuation with a Si diode on the cold stage of the cryocooler and a Pt-1000 thermistor on the cooper block near the laser, where less than 30 mK bath temperature variation of the QCL was measured. With a temperature tuning coefficient of -33 MHz/K obtained from Ref. 6, a free running linewidth of less than 1 MHz for the QCL is expected, which is in agreement with our measurement data. Furthermore, we notice that our frequency locked linewidth is consistant with published results in the literature, which was either 30 KHz measured by a heterodyne technique [9] in a very short period of 3 ms or 6.3 KHz obtained in a frequency locking measurement [18].

IV. Conclusion

In conclusion, we succeeded in frequency locking a 3.5 THz quantum cascade laser by using a methanol absorption line as the frequency reference. Two different methods were applied for determining the linewdith where similar linewidth results were obtained. The locked linewidth of the QCL is found to be below 17 KHz with a Gaussian-like shape. Our experiment indicates that this frequency locking scheme is robust and has advantages for QCL at higher frequencies. A future development is to replace the HEB detector in our experiment with other power detectors working at a relatively high temperature (e.g. 77 K or above) and using a small sized gas cell. So, this frequency locking scheme will be competent for practical instrument.

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