High-resolution heterodyne spectroscopy using a tunable quantum cascade laser around 3.5 THz

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A frequency tunable terahertz heterodyne spectrometer, based on a third-order distributed feedback quantum cascade laser as a local oscillator, has been demonstrated by measuring molecular spectral lines of methanol (CH_3OH) gas at 3.5 THz. By varying the bias voltage of the laser, we achieved a tuning range of ~1 GHz of the lasing frequency, within which the molecular spectral lines were recorded. The measured spectra show excellent agreement with modeled ones. By fitting we derived the lasing frequency for each bias voltage accurately. The ultimate performance of the receiver including the resolution of noise temperature and frequency is also addressed. © *2011 American Institute of Physics*. [doi:10.1063/1.3599518]

Driven by the demands of astronomical observations and atmospheric remote sensing in the terahertz (THz) frequency range, we have recently developed a high resolution heterodyne spectrometer using a quantum cascade laser (QCL) at 2.9 THz as a local oscillator (LO) and a NbN hot electron bolometer (HEB) as a mixer.[1] However, such a spectrometer is not yet adequate for operation in a telescope because of a number of drawbacks noted during the previous experiment. First, the QCL used previously was based on a metal-metal waveguide Fabry–Perot cavity design, which has no mode control of the lasing frequency and has virtually zero tuning range by the bias voltage because of a resulting strong reduction in the output power. The tuning capability is, in general, highly desirable for application in spectroscopy as it is crucial for targeting more molecular lines, and also a means to identify unknown spectral lines when a heterodyne receiver is operated in the double sideband (DSB) mode.[1] Second, a ^{4}He flow cryostat was used to operate the QCL. For any balloon-borne and space mission, the use of a liquid-He based cryostat can impose a serious obstacle due to the relatively high DC power dissipation of the laser. Therefore, a dry, liquid cryogen-free cooler such as a pulse tube cryocooler or a Stirling cooler[2] is preferred. One of the challenges in using a dry cooler is the mechanical stability. As demonstrated in Ref. 3, the vibration in the cooler can introduce deviations in the operating point of the detector, leading to instability of the receiver.

In this letter we report on a high-resolution heterodyne molecular spectroscopic experiment applying a 3.5 THz QCL as a LO. In comparison with the previous work,[1] there are three key differences; (a) a single mode, third-order distributed feedback (DFB), tunable QCL (Refs. 4 and 5) is used as a LO; (b) a pulse tube cryocooler is applied to operate the QCL; and (c) a theoretical model for methanol molecular lines has been verified at 3.5 THz, which was not possible until now because of the lack of a heterodyne technique at such a frequency.

By using the third-order periodic structure with strong refractive index contrast gratings, not only is the single mode emission achieved in the DFB laser, but also the radiation power out-coupling from the laser to the free space is considerably improved. Moreover, the grating structure behaves like a linear phased array antenna, resulting in a low-divergent far field beam. As a result, a higher power coupling efficiency from the QCL to a HEB mixer is expected in comparison with the use of a metal–metal waveguide, Fabry–Perot cavity QCL. Our laser, based on a 10 μm thick active region, consists of 27 periods of gratings with a total length of 1070 μm. Characterized by a Fourier transform spectrometer (FTS) with a resolution of 0.6 GHz, the laser shows a single mode emission line from 3452.0 to 3450.8 GHz by increasing the bias voltage from 13.9 to 14.9 V. It provides a maximum output power of 0.8 mW when operated with 3 W dc input power at a bath temperature of ~12 K. The divergence of the far-field beam is about 12° in both vertical and horizontal directions. For the detector, we use the same spiral antenna coupled NbN HEB mixer

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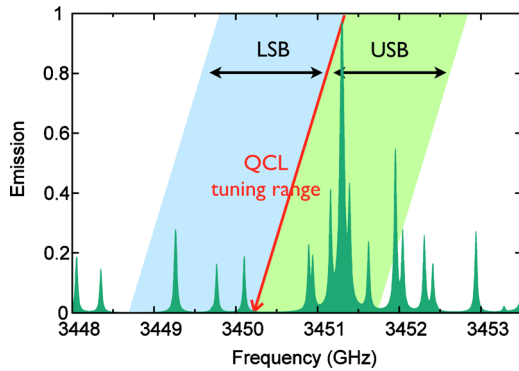
FIG. 1. (Color online) Calculated emission spectra of methanol at a pressure of 1 mbar at 300 K for a 41 cm optical path length. The red arrow indicates the tuning range of the LO frequency. The left and right shadow regions of the LSB and USB correspond to the detection band of the FFTS for the IF signal by using a 0–1.5 GHz low pass filter.
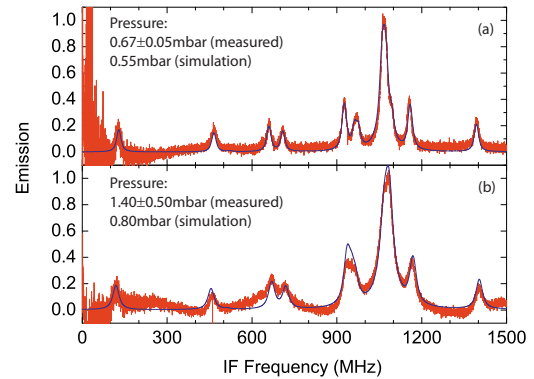

FIG. 2. (Color online) Experimental and simulated methanol spectral lines as a function of frequency at different gas pressure with a LO frequency of 3450.232 GHz. (a) The low pressure spectrum; (b) the high pressure data.

as used in Ref. 6, which has shown superior sensitivities from 1.6 to 5.3 THz.

Figure 1 shows the calculated methanol emission spectrum in the vicinity of the LO frequency. To study the methanol spectrum, we apply a measurement setup similar to the one described in Ref. 1. However, two major changes have been made. First, the QCL is mounted on the second stage of a pulse tube cryocooler, where the temperature rises up to 12 K during the laser operation. It has been found[3] that the fluctuation of the coupled power from the QCL to the HEB mixer due to the piston movement of the cooler will introduce instability in the HEB. Thus an additional damper is introduced to minimize the mechanical vibration from both the refrigerator and the flexlines so that the stability of the HEB becomes comparable with the level achieved by a liquid-He-cryostat operated QCL. Second, although the same fast FTS (FFTS) is used as the back-end spectrometer, we now only make use of the baseband (0–1.5 GHz) for sampling the intermediate frequency (IF) signals, preventing an aliasing effect.[1]

The QCL is operated without any stabilization on the phase and the amplitude. The beam from the QCL passes through a high-density polyethylene (HDPE) window of the cooler and is focused to the mixer with a HDPE lens (f=26.5 mm). The signal source is a combination of a gas cell and hot/cold blackbody loads. The blackbody load is defined as a hot load at 295 K and as a cold load at 77 K. The gas cell is a 41 cm long cylinder at room temperature and has two 2 mm thick HDPE windows. The methanol emission lines around 3.5 THz from the gas cell are combined with the QCL's radiation by a 13 μm thick Mylar beam splitter and fed into the HEB mixer. The mixer down-converts the THz spectral lines at f_S to an IF f_{IF} in the DSB mode where both the signal at upper side band (USB) $f_{USB}=f_{LO}+f_{IF}$ and lower side band (LSB) $f_{LSB}=f_{LO}-f_{IF}$ will be converted to the same IF frequency range. The IF signal is amplified first using a wide band (0.5–12 GHz) low noise amplifier at 4.2 K that is followed by a two-stage room-temperature amplifiers with a 1.5 GHz low pass filter used in between to define the detection band width.

The same method is applied to measure the spectral lines as in Ref. 7, where three IF power spectra were measured as follows: the spectrum $P_{emp,cold}(f)$, $P_{gas,cold}(f)$, and $P_{gas,hot}(f)$, taking 3 s of integration time for each single spectrum. With these three spectra, the term $2[P_{gas,cold}(f)-P_{emp,cold}(f)]/$

$[P_{gas,hot}(f)-P_{emp,cold}(f)]$ is obtained, which reflects the relative emission of the gas with respect to the 300 K blackbody radiation, and the factor of 2 reflects the DSB mode of operation of the HEB mixer.

Several methanol spectral lines around 3.5 THz were observed simultaneously within the IF frequency range. To check the consistency with the theoretical model, the spectra were taken at different pressure because of the broadening effect. We apply the model developed in our previous work,[1] which is based on the expected methanol line frequencies with intensities from the Jet Propulsion Laboratory (JPL) line catalog[8] and includes following three different broadening mechanisms: thermal Doppler broadening, pressure broadening, and opacity broadening. Comparisons were made between the measured and simulated methanol spectral lines at 3.5 THz. As shown in Fig. 2, for a low-pressure case of 0.67 mbar, an excellent agreement is obtained with respect to both the line frequencies and the relative intensities. While for the high-pressure case of 1.4 mbar, the experimental result shows deviation of the line intensities from the theoretical simulation. This deviation might be attributed to the shifting of HEB mixer's working point due to emission from the gas cell, or the so-called direct detection effect. At high gas pressure, since the emission power from the methanol molecule becomes relatively high, this effect is more prominent. The difference between the measured gas pressure and the value used for the simulation indicated in Fig. 2 is due to the inaccuracy of the gas pressure calibration. Furthermore, the pressure broadening coefficient, as defined in HITRAN database,[9] was found to be 14.8 MHz/mbar (0.5 cm^{-1}/atm), which is a factor of 2 higher than the value found at 2.9 THz, suggesting that this coefficient can vary noticeably with frequency.

To enable a frequency tunable spectroscopic measurement, not only is a relatively large frequency tuning range of the laser crucial but also the emission power should be sufficient to operate the mixer at all frequencies. As we demonstrate in this work, the third-order DFB lasers can meet both of these requirements. Figure 3 illustrates several methanol spectra recorded at different LO frequencies, tuned by the QCL bias voltage. As the QCL frequency decreases with increasing bias voltage, a strong methanol emission line located at 3451.2988 GHz shifts from 13 MHz in LSB to 1064 MHz in USB. Figure 3 shows only the spectra starting from 170 MHz in USB. Within the entire frequency tuning range of about ~1 GHz, we made comparisons between the mea-
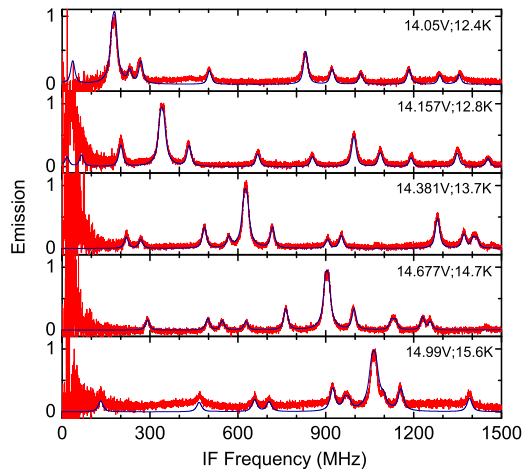
FIG. 3. (Color online) Measured methanol spectra with different LO frequency, which is tuned with the bias voltage of the QCL from 14.05 to 14.99 V. The gas pressures for all spectra are at 0.65 ± 0.05 mbar.

sured spectra and theoretical ones. As shown partly in Fig. 3, excellent agreements were achieved for the entire frequency region. By fitting with the model, we derived the lasing frequency as a function of the bias, which is shown in Fig. 4. Moreover, in contrast to a previously reported first-order DFB laser that shows single-mode operation within only a limited bias range,[10] we confirmed single-mode emission behavior of a third-order DFB QCL for the entire operation range[4] since all the measured molecular spectra could be well described.

QCL frequency tuning is dominated by the change in the refractive index with the temperature change in the active region. To distinguish electrical tuning from thermal tuning we also measured the frequency tuning by varying only the bath temperature. The results are summarized in the inset of Fig. 4. A 150 MHz tuning range is shown for a temperature range from 13.2 to 17.6 K, where a thermal tuning coefficient of −33 MHz/K was obtained. After subtraction of the thermal effect, an electrical tuning coefficient of −859 MHz/V is derived for our case. This tuning coefficient is smaller than those reported from Fabry–Perot cavity lasers.[11,12] This can be explained by the fact that the gain medium used for this laser has dual gain peaks at 3.3 and 3.8 THz. At 3.5 THz, the effects of the frequency pulling from these two peaks partially cancel out each other and thus reduce the tuning coefficient.

In the current experiment, the smallest linewidth for the methanol spectral line is measured to be 11 MHz at a pres-
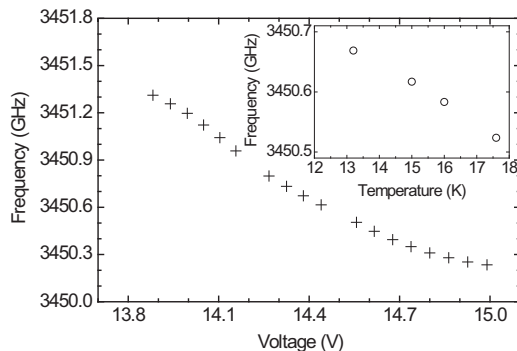
sure of 0.43 mbar. By assuming that we need a linewidth of the QCL that is at least 10% of the measured spectral linewidth we derive a linewidth of 1 MHz for the QCL. This is reasonable in comparison to the expected free-running linewidth due to jitter.[12] We note that for astronomical observations, atomic and molecular gases are at lower pressure and lower temperature, resulting in narrow spectral linewidths of ~1 MHz. Obviously, to reduce the linewidth phase or frequency locking of the QCL will be required. The rich molecular absorption lines in combination with the frequency tunability can facilitate the frequency locking.[13]

In conclusion, we succeeded in demonstrating a tunable high-resolution heterodyne spectrometer using a 3.5 THz third-order DFB QCL operated in a pulse tube cryocooler as a local oscillator. By adjustment of the bias voltage, a frequency tuning range of ~1 GHz has been achieved in heterodyne spectroscopic measurements. Within the entire frequency tuning range, the measured spectra show excellent agreement with the theoretical simulations. By fitting with the model we derived the lasing frequency versus bias voltage accurately. Even though the observed tuning range is still small, this result demonstrates the benefits to be expected from a further increase in the tuning range of THz QCLs, such as the development reported recently in Ref. 14.

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FIG. 4. QCL emission frequency as a function of bias voltage. The inset shows the measured QCL frequency at different bath temperature.