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A terahertz (THz) heterodyne spectrometer is demonstrated based on a quantum cascade laser (QCL) as a local oscillator (LO) and an NbN hot electron bolometer as a mixer, and it is used to measure high-resolution molecular spectral lines of methanol ($CH_3OH$) between 2.913–2.918 THz. The spectral lines are taken from a gas cell containing methanol gas and using a single-mode QCL at 2.9156 THz as an LO, which is operated in the free running mode. By increasing the pressure of the gas, line broadening and saturation are observed. The measured spectra showed good agreement with a theoretical model. © *2010 American Institute of Physics*. [doi:10.1063/1.3502479]

A high-resolution heterodyne spectrometer is of crucial importance for astronomical observation and atmospheric remote sensing in the terahertz (THz) frequency range. It consists of essentially a mixing detector, a local oscillator (LO), a low noise amplifier, and a GHz-band back-end spectrometer, providing both uniquely high spectral resolution ($\nu/\Delta\nu > 10^6$, limited by the LO and back-end spectrometer, where $\nu$ is the frequency and $\Delta\nu$ is the frequency resolution) and excellent sensitivity (e.g., receiver noise temperature of ~1000 K at 3 THz). Heterodyne receivers based on super-conducting mixers and electronically tunable solid-state multiplier-chain LO sources have been realized up to 2 THz for ground based, balloon-borne, and space telescope instruments but the development of receivers at higher frequencies will be determined by the availability of suitable solid-state LO sources. Recently developed THz quantum cascade lasers[1] (QCLs) are the candidates for the LO at frequencies above 2 THz. Heterodyne receivers using a THz QCL as an LO and an NbN hot electron bolometer (HEB) as a mixer have demonstrated high sensitivity using broadband black-body radiation (hot/cold loads) as the calibration source.[2,3] A number of milestones for using a THz QCL as an LO have been demonstrated, such as phase-locking capability,[4] narrow intrinsic linewidth,[5] and excellent power stability.[2] A remaining key step is a direct measurement of spectral lines by a heterodyne spectrometer using a THz QCL as an LO. A spectroscopic measurement using a THz QCL as an active tuning source has been reported for gas phase spectroscopy.[6] However, the detection itself was not based on the heterodyne principle.

In this paper we report high-resolution spectroscopic measurements using a heterodyne spectrometer with a 2.9 THz QCL as an LO and an NbN HEB as a mixer. We observed simultaneously several molecular emission lines of methanol gas around 2.9 THz. By varying the pressure of the gas, we are able to follow the line broadening and also make a comparison between measured and theoretical spectra.

The LO used in our experiment is a metal-metal Fabry–Perot ridge waveguide THz QCL, based on the resonant phonon depopulation design,[7] with cavity dimensions of 1.45 mm $\times$ 25 $\mu$m. The QCL is mounted on the cold stage of a helium-flow cryostat, and emits a single-mode emission line at 2.9156 THz in continuous wave (CW) mode as measured by a Fourier-transform Spectrometer. While the intrinsic linewidth of a QCL is expected to be in the range of 6–30 KHz,[5] in the current measurement, the QCL is operated in free-running mode without any stabilization on the phase and the amplitude. Based on the previous experience,[4] we expect a free-running QCL linewidth of less than 1 MHz, which is sufficiently narrow for this laboratory spectroscopic measurement.

The mixer is a spiral antenna coupled NbN HEB, which is glued to the backside of an elliptical Si lens and is operated at 4.2 K. As shown in a separate experiment,[8] the same mixer can have superior sensitivities across the frequency range of 1.6–5.3 THz, from which the double sideband (DSB) receiver noise temperature ($T_{DSB,Rec}$) is expected to be 1000 K at 2.9 THz.

The spectroscopic measurement setup is sketched in Fig. 1. The THz radiation beam from the QCL first passes through the high-density polyethylene (HDPE) cryostat window and is then focused with a HDPE (f=26.5 mm) lens. The signal source is a combination of a gas cell and hot/cold (295 K/77 K) blackbody loads. The gas cell is a 50 cm long cylinder with an inner diameter of 10 cm at room temperature and has two 2 mm thick HDPE windows. The gas pressure inside the cell is measured using a gas-independent
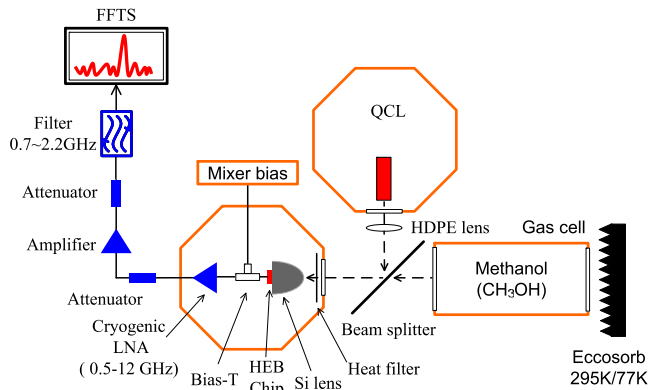
FIG. 1. (Color online) Schematic view of the heterodyne gas cell measurement setup.

gauge, where the pressure can be controlled with an accuracy of at least 0.1 mbar. Figure 2 shows a calculated methanol emission spectrum in the vicinity of the LO frequency ($f_{LO}$),[9] where the intensity is relative to the 300 K blackbody radiation. In the experiment, the signal source and the QCL's radiation are combined by a 3 $\mu$m thick Mylar beam splitter and fed further into the HEB mixer. The mixer down-converts a spectral line at $f_s$ to an intermediate frequency $f_{IF}$, where $f_{IF} = |f_{LO} - f_s|$. Since the HEB mixer is operated in DSB mode, both the signal at above $f_s = f_{LO} + f_{IF}$ (upper side band, USB) and below $f_s = f_{LO} - f_{IF}$ (lower side band, LSB) 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, and then followed by two stages of room-temperature amplifiers.

The back-end spectrometer is a Fast Fourier Transform Spectrometer (FFTS),[10] which samples the IF signals in the baseband (0–1.5 GHz) or in the second Nyquist band (1.5–3.0 GHz) with a spectral resolution of 183 KHz. In our experiment, a 0.7–2.2 GHz band pass filter is applied at the input of the FFTS, which defines the actual band at the IF frequency. Consequently, we expect at least six emission lines, labeled with "a" to "f" in Fig. 2, distributed in the two corresponding bands (LSB and USB) at the THz frequencies.
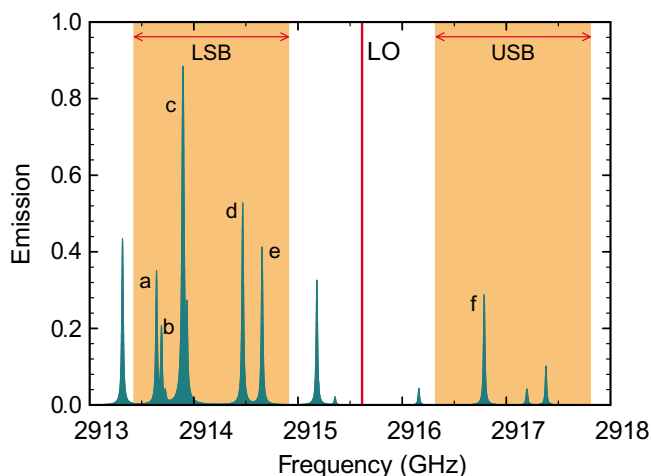


FIG. 2. (Color online) Calculated emission spectra of methanol ($CH_3OH$) at a pressure of 1 mbar and at 300 K, for a 0.5 m optical path length. The black line indicates the LO at 2915.6 GHz. The shadow regions of the LSB and the USB correspond to the detection band of the FFTS for the IF signal by using a 0.7–2.2 GHz band pass filter. Several methanol lines within the shadow regions, labeled with "a" to "f," are expected to be seen.

Because of a combination of the bandpass filter with the FFTS, there will be a so-called aliasing effect[11] which means that the methanol lines at $f_{LO} \pm f_{IF}$ ($f_{IF} > 1.5$ GHz) in the second Nyquist band will overlap with the methanol lines at $f_{LO} \pm f_{IF}$ ($f_{IF} < 1.5$ GHz) in the baseband at the same IF frequency.

We start our spectroscopic experiment by characterizing the sensitivity of the whole receiver system. The measured $T_{DSB,Rec}$ was 2500 K for the case where the hot/cold loads are positioned just behind the beam splitter. This value is higher than the expected one[8] (1000 K) which can be attributed to the extra losses due to air and the HEB cryostat window, the nonoptimized IF chain, and the direct detection effect. To measure the spectral lines of the gas, three IF power spectra were measured:[12] (1) the spectrum $P_{emp,cold}(f)$ when the cold load is behind the evacuated gas cell; (2) the spectrum $P_{gas,cold}(f)$ when the cold load is behind the filled gas cell; (3) the spectrum $P_{gas,hot}(f)$ when the hot load is behind the filled gas cell. Each trace is recorded using the FFTS with an integration time of 5 s. With the three spectra, the brightness of the emission lines in terms of temperature is calculated using the following expression:[12]

$$T_{gas}(f) = T_{cold} + 2(T_{hot} - T_{cold})\frac{P_{gas,cold}(f) - P_{emp,cold}(f)}{P_{gas,hot}(f) - P_{emp,cold}(f)}, \quad (1)$$

where $T_{hot}$ and $T_{cold}$ are the effective hot and cold load temperatures defined by the Callen–Welton formula,[13] and the factor of 2 in Eq. (1) is derived from the DSB mode operation of the HEB mixer. The term $2[P_{gas,cold}(f) - P_{emp,cold}(f)]/[P_{gas,hot}(f) - P_{emp,cold}(f)]$, reflects the relative emission of the gas with respect to the 300 K blackbody radiation. Due to the aliasing effect an additional factor of two was introduced into the relative emission term in Eq. (1), which accounts for the additional folding of the data for our particular experiment.

By varying the gas pressure we are able to map out the methanol spectral lines at different pressures. Figure 3 shows such spectra within the IF frequency range between 0.8 and 1.5 GHz and with increased pressures from 0.93 to 3.28 mbar, where several lines with different intensity were simultaneously observed within the IF band. A relatively strong emission line at 1281 MHz is observed, which is assumed to be the "c" line at 2913.896 GHz in the LSB of Fig. 2 [corresponding to R-branch transition from the upper state (J=23, K=10, and v=0) to the lower state (J=22, K=9, and v=0), E symmetry].[14] Additionally, several relatively weak lines from both LSB and USB are observed. With increasing gas pressure, the spectral linewidths become broader and the line intensities increase, until at a high pressure of 3.28 mbar the spectral lines are observed to saturate.

To simulate the spectra, a theoretical model was generated based on the expected line frequencies and intensities for methanol from the JPL line catalog[14] and also the effects of line broadening, the parameters of the gas cell, and the FFTS spectrometer were included. The profile of each line shown in Fig. 3 is a convolution of several different broadening mechanisms: thermal Doppler broadening, pressure broadening, and opacity broadening. The Doppler broadening can be described by a Gaussian profile where the linewidth is a function of gas temperature, line frequency, and molecular mass of the gas. The pressure broadening effect is characterized by a Lorentzian profile that is determined by
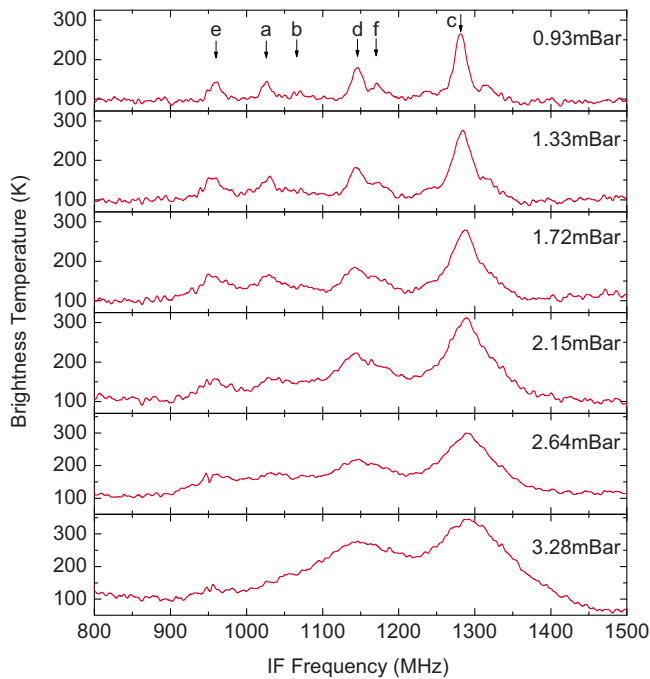
FIG. 3. (Color online) Measured methanol (CH$_3$OH) emission spectra within the IF range between 0.8 and 1.5 GHz at different gas cell pressure which varies from 0.93 to 3.28 mbar. The QCL's frequency is 2915.6 GHz.

the gas pressure and a pressure broadening parameter. The opacity broadening (Beer–Lambert law) is an exponential function of the path length of the gas cell and describes the transmission of radiation through the gas column. Figure 4 shows the calculated and measured methanol emission spectra at two different pressures. The calculated spectra show a reasonable agreement with measurements with regard to both the line frequencies and intensities. Since no pressure broadening data were available in the literature for methanol at the frequencies of our experiment, this parameter was left as a free parameter in a fitting routine. The optimum value for the pressure broadening, as defined in the HITRAN database,[15] was found to be 7.4 MHz/mbar (0.25 cm$^{-1}$/atm) for our case.
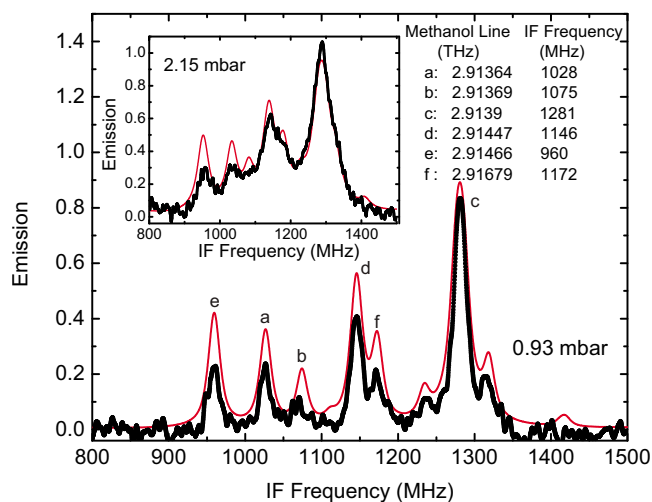


FIG. 4. (Color online) Measured methanol emission spectra (black curve) and the simulated spectra (red curve) at a gas pressure of 0.93 mbar. The spectral lines are labeled according to what is given in Fig. 2. The correspondence between IF frequency and THz frequency for each line is also listed. The inset shows the measured (black curve) and the simulated spectrum (gray curve) at a higher pressure of 2.15 mbar.

The following effects can influence the measured amplitudes of the spectral lines and may explain the differences between data and model: (a) the side band ratio of the receiver is assumed to be unity but in practice it can deviate from unity; (b) while we assume the receiver gain at the different IF frequencies between the baseband and second Nyquist band to be equal, this is not necessary true. Furthermore, the use of a broadband IF amplifier in our case can cause possible standing waves. These effects, together with the uncertainty of the measurement governed by stability of the receiver, can cause the apparent relative emission to be greater than unity (inset of Fig. 4) so that the brightness of the "c" line exceeds 300 K at higher pressure (Fig. 3).

In conclusion we have succeeded in demonstrating high-resolution spectroscopic measurement using a heterodyne receiver based on a 2.9 THz quantum cascade laser as a local oscillator and a NbN HEB as a mixer. We measured the molecular spectra of methanol gas with good agreement to simulated model spectra. Our gas cell measurement is a crucial demonstration of the QCL as an LO for practical instruments. These high-resolution spectra of the different molecular lines at such a high frequency indicate that the heterodyne spectrometer based on a QCL and a HEB mixer has unique advantages for its high resolution and excellent sensitivity, and can be applied at any THz frequency.

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