Temperature and frequency resolution of a 3.5 THz heterodyne spectrometer using a quantum cascade laser

Y. Ren, J.N. Hovenier, J.R. Gao, T.M. Klapwijk, S.C. Shi, T-Y. Kao, Q. Hu, and J. L. Reno

Abstract—Here we characterized the temperature and frequency resolution of a 3.5 THz heterodyne spectrometer based on a quantum cascade laser (QCL) as a local oscillator and a superconducting NbN hot-electron bolometer (HEB) as a mixer. High resolution spectral lines of methanol (CH₃OH) gas around 3.5 THz were obtained by performing heterodyne spectroscopic measurements. From the measured spectra, we determined the minimal temperature change that could be resolved for the heterodyne receiver in different operation manner. The spectral resolution of the receiver was also addressed.

Index Terms—terahertz, quantum cascade laser, high resolution spectroscopy, temperature resolution, frequency resolution

I. INTRODUCTION

H igh resolution heterodyne spectroscopy plays a vital role in astronomical observation and atmospheric remote sensing at terahertz (THz) frequencies, due to its high spectral resolution $(v/\Delta v>10^6)$, limited by the local oscillator (LO) and back-end spectrometer, where v is the frequency, and Δv is the frequency resolution) and excellent sensitivity (e.g., receiver noise temperature of ~1000 K at 3 THz). As the mixer, a superconducting NbN hot-electron bolometer (HEB) has been applied in the HIFI instrument on the Hershel Space Observatory covering the frequency range from 1.4 to 1.9 THz. Recently an HEB mixer has demonstrated superior sensitivity up to 5.3 THz [1]. As the LO, terahertz quantum cascade lasers (QCLs) [2] have shown eminent advantages at

This work was partly supported by China Exchange Programme executed by KNAW and CAS, and by the AMSTAR+ project of RadioNet under FP7, and NWO.

Y. Ren is with the Kavli Institute of NanoScience, Delft University of Technology, Lorentzweg 1, 2628 CJ Delft, The Netherlands, with the Purple Mountain Observatory, Chinese Academy of Sciences, Nanjing, China, and with the graduate school, Chinese Academy of Science, Beijing, China, (e-mail: y.ren@tudelft.nl)

J.N. Hovenier and T.M. Klapwijk are with the Kavli Institute of NanoScience, Delft University of Technology, Lorentzweg 1, 2628 CJ Delft, The Netherlands

J.R. Gao is with the Kavli Institute of NanoScience, Delft University of Technology, Lorentzweg 1, 2628 CJ Delft, The Netherlands, and with the SRON Netherlands Institute for Space Research, Sorbonnelaan 2, 3584 CA Utrecht, The Netherlands (j.r.gao@tudelft.nl)

S.C. Shi is with the Purple Mountain Observatory, Chinese Academy of Sciences, Nanjing, China

T-Y. Kao and Q. Hu are with Department of Electrical Engineering and Computer Science, Massachusetts Institute of Technology, Cambridge, Massachusetts 02139, USA

J.L. Reno is with Center for Integrated Nanotechnologies, Sandia National Laboratories, Albuquerque, NM 87185-0601, USA

frequencies above 2 THz, based on their high output power, wide emission frequency range and excellent power stability compared to FIR gas lasers. As the development of the heterodyne receiver moves from single pixels to arrays, higher LO power becomes crucial. Therefore, the high output power of a THz QCL appears to be extremely attractive. Recently, several progresses have been made for a THz QCL to be used as the LO, including a noise temperature measurement [3,4], excellent power stability [3] and phase-locking capability [5,6]. Furthermore, as shown in Ref. 7 and 8, heterodyne spectroscopic measurements have also been performed based on a THz QCL as a LO. High resolution molecular spectra were obtained and showed excellent agreement with the modeled ones. All these results suggest that a THz QCL is competent as a LO in a heterodyne receiver above 2 THz. For a heterodyne receiver, both temperature and frequency resolution are of crucial importance since they define the minimum temperature and frequency difference that can be resolved within the integration time [9]. A heterodyne molecular spectroscopic measurement is a direct means to characterize the ultimate performance of a heterodyne receiver in the spectroscopic mode.

In this paper we report on the temperature and frequency resolution of a 3.5 THz heterodyne spectrometer, based on a THz QCL as a LO and a superconducting NbN HEB as a mixer. We observed simultaneously several molecular spectral lines of methanol gas around 3.5 THz. From the measured spectra, the minimal temperature and frequency resolution were determined.

II. MEASUREMENT SETUP

The LO used in our experiment is designed and fabricated at the MIT group. It is a third-order distributed feedback (DFB) THz QCL [10,11], which is the same one as described in Ref. 8. Our laser consists of 27 periods of gratings with a total length of 1070 μ m. It provides a peak emission power of 0.8 mW at a bath temperature of ~12 K. Based on the third-order periodic structure with strong refractive 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. Furthermore, 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 the HEB mixer is obtained in comparison with the use of a metal-metal waveguide Fabry-Perot cavity QCL. Moreover, \sim 1 GHz frequency tuning range is achieved for the LO frequency by varying the bias voltage or the bath temperature of the laser.

The mixer is a spiral antenna coupled superconducting NbN HEB, the same as used in Ref. 1. It is glued to the backside of an elliptical, uncoated Si lens and is operated at 4.2 K.



Fig. 1. Schematic view of the heterodyne spectroscopic measurement setup.



Fig. 2. Picture of the heterodyne spectroscopic measurement setup.

The spectroscopic measurement setup is sketched in Fig.1, which is the same as applied in Ref. 8. The QCL is mounted on the second stage of a pulse tube cryocooler. In order to reduce its mechanical vibration, which causes the instability of the output beam of the OCL and in turn introduces the instability of the HEB receiver, an additional damper is used to minimize the mechanical vibration. The QCL is operated in a free-running mode, namely without any stabilization on its phase and amplitude. The radiation beam from the QCL is focused by a high-density polyethylene (HDPE) lens. The signal source is a combination of a gas cell with a hot load at 295 K and 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. Methanol (CH₃OH) gas is chosen since it has abundant absorption lines at this frequency range. 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 fs to an intermediate frequency (IF) f_{IF}. Since the HEB is working in the double

sideband (DSB) mode, 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 first amplified using a wide band (0.5-12 GHz) low noise amplifier at 4.2 K, followed by a two-stage room-temperature amplifier with a 1.5 GHz low pass filter used inbetween to define the detection bandwidth. The back-end spectrometer is a Fast Fourier Transform Spectrometer (FFTS) [12], which samples the IF signals in the baseband (0-1.5 GHz) with a spectral resolution of 183 KHz.

III. MEASUREMENT RESULTS

We start our spectroscopic experiment by characterizing the sensitivity of the entire receiver system. In this case, a band-pass filter centered at 1.4 GHz with 100 MHz bandwidth is used, and a power meter is applied instead of the FFTS. The measured $T_{DSB,Rec}$ was 3800 K for the case where the hot/cold loads are positioned behind the evacuated gas cell. This noise temperature value is higher than previous reported one [1], which can be attributed to extra losses due to air, the HEB cryostat window as well as the gas cell windows, the non-optimized IF chain, and the direct detection effect.

The same method as in Ref. 9 is applied to measure the spectral lines. With this method three IF power spectra were recorded: (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. With the three spectra, the intensity of the molecular emission lines in terms of temperature is calculated using the following expression [9]:

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

(1)

where Callen-Welton temperature [13] is used to define the effective hot and cold load temperature, and a pre-factor of 2 in Eq.(1) reflects the DSB mode operation of the HEB mixer.

By doing this, a calibrated spectrum within the IF frequency range between 0-1.5 GHz is obtained, as shown in Fig.3. Several methanol spectral lines with different intensity around 3.5 THz were simultaneously observed. For a Dicke-type radiometer, the temperature resolution is given by [9],

$$\Delta T = \sqrt{\frac{(T_A + T_R)^2}{B\tau_A} \frac{1}{d_A} + \frac{(T_{ref} + T_R)^2}{B\tau_{ref}} \frac{1}{d_{ref}}}$$
(2)

where T_A is the effective antenna temperature; T_R the receiver noise temperature; T_{ref} the reference signal temperature; B the detection bandwidth; τ_A the integration time for the spectral line signal; τ_{ref} the integration time for the reference signal; d_A and d_{ref} are the duty cycles of the observation time for the spectral line signal and reference signal, respectively. In our case, $T_A=300$ K, $T_R=3800$ K, $T_{ref}=77$ K, B=183 KHz. For each single spectrum, it takes 3 seconds of integration time. So for a single spectrum, $\tau_A=\tau_{ref}=3$ sec, and $d_A=d_{ref}=1/3$, from which we expect a ΔT of 13.2 K. As shown in Fig.3a, the standard deviation of measured noise level was 13.8 K. We also performed 18 series of the Pgas,cold(f) spectrum measurement and 21 series of the P_{emp,cold}(f) spectrum measurement. The calibrated spectrum was subsequently averaged as plotted in Fig.3b. In the time averaged case, $\tau_A{=}3$ sec*18 (18 times average), τ_{ref} =3 sec*21 (21 times average), $d_A = \frac{18}{18+21+1}$, $d_{ref} = 21/(18+21+1)$, from which we expect a ΔT of 2.5 K. Our experimental data indicate a temperature resolution of 3.5 K. Then the time averaged spectrum was smoothed to the 1.83-MHz resolution, as plotted in Fig.3c. Since B=1.83 MHz, we expect a ΔT of 0.79 K, where our experiment data is 1.44 K. Based on those data, we conclude that the temperature resolution in the single measurement follows the value expected from the radiometer equation. However, a factor of 2 difference was observed for the multiple, time averaged case. For the latter case, as the QCL was operated in the free-running mode, the 1/f noise in the amplitude of the QCL could attribute to the discrepancy between the measured results and the value from the radiometer equation. This effect may relate to the issue of the Allan variance time of the whole receiver system [14]. Since we are doing spectroscopic measurements, so the relevant Allan variance time is a spectroscopic Allan time, which should be much longer than, e.g. 3 sec. However, we do not know the upper limit of the Allan time in our case. To get such information this merits in-depth studies in the future work. The worse temperature resolution for the spectrum below 500 MHz is mainly due to higher noise contribution from the cryogenic amplifier, which is beyond its operating frequency range.



Fig. 3. High resolution methanol (CH₃OH) emission spectrum measured within the IF frequency range between 0-1.5 GHz. (a) the original spectrum, with 3 seconds integration time for 3 spectrum traces; (b) the time averaged spectrum, where 18 series of the $P_{gas,cold}(f)$ spectrum and 21 series of the $P_{emp,cold}(f)$ spectrum were averaged; (c) the time and frequency averaged spectrum, the time averaged spectrum was smoothed to the 1.83-MHz resolution.

Also the frequency resolution is crucial for a heterodyne spectrometer. In the current experiment, with the free-running QCL as a LO the narrowest linewidth for the measured methanol spectral line is 11 MHz at a pressure of 0.43 mbar. We inferred that the linewidth of the QCL should be in order

of 1 MHz. The linewidth of this laser has been studied separately and is reported in Ref. 15. In the free-running mode it was found to be around 900 KHz. Together with a 183 KHz resolution FFTS, the frequency resolution of the heterodyne spectrometer increases to 1.1 MHz. This value agrees with that inferred from our experiment. We note that for astronomical observations at THz frequencies, 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. As shown in Ref. 15, the linewidth of the QCL is reduced to 17 KHz by locking to a molecular absorption line. This locked linewidth together with the FFTS we used enable a THz heterodyne spectrometer with a frequency resolution of 200 KHz ($\nu/\Delta\nu > 10^7$). And the rich molecular absorption lines in combination with the frequency tunability facilitate the frequency locking scheme and hold the advantage for higher frequencies.

IV. CONCLUSION

In conclusion, by performing the spectroscopic experiment, we characterize the temperature and frequency resolution of a heterodyne spectrometer at 3.5 THz using a quantum cascade laser as a LO and a NbN HEB as a mixer. The receiver is capable of resolving a temperature change of 3.5 K for the effective brightness temperature of a molecular line and a frequency resolution of 1 MHz when the QCL is operated in the free-running mode. This frequency resolution can be improved to 200 KHz, limited by the back-end spectrometer, by locking the QCL to a molecular absorption line. The receiver is shown to be competent for astronomical and atmospheric applications.

REFERENCES

- W. Zhang, P. Khosropanah, J.R. Gao, E. L. Kollberg, K. S. Yngvesson, T. Bansal, R. Barends, and T. M. Klapwijk, "Quantum noise in a terahertz hot electron bolometer mixer," Applied Physics Letter 96, 111113 (2010),
- [2] B. S. Williams, "Terahertz quantum-cascade laser," Nat. Photonics 1, 517 (2007).
- [3] J.R. Gao, J.N. Hovenier, Z.Q. Yang, J.J.A. Baselmans, A. Baryshev, M. Hajenius, T.M. Klapwijk, A.J.L. Adam, T.O. Klaassen, B.S. Williams, S. Kumar, Q.Hu, and J.L. Reno, "Terahertz heterodyne receiver based on a quantum cascade laser and a superconducting bolometer," Appl. Phys. Lett. 86, 244104 (2005).
- [4] H.-W. Hübers, S. G. Pavlov, A. D. Semenov, R. Köhler, L. Mahler, A. Tredicucci, H. E. Beere, D. A. Ritchie, and E. H. Linfield, "Terahertz quantum cascade laser as local oscillator in a heterodyne receiver," Opt. Express. 13, 5890 (2005).
- [5] P. Khosropanah, A. Baryshev, W. Zhang, W. Jellema, J. N. Hovenier, J. R. Gao, T. M. Klapwijk, D. G. Paveliev, B. S. Williams, S. Kumar, Q. Hu, J. L. Reno, B. Klein, and J. L. Hesler, "Phase locking of a 2.7 THz quantum cascade laser to a microwave reference," Opt. lett. 34, 2958 (2009)
- [6] D. Rabanus, U. U. Graf, M. Philipp, O. Ricken, J. Stutzki, B. Vowinkel, M. C. Wiedner, C. Walther, M. Fischer, and J. Faist, "Phase locking of a 1.5 Terahertz quantum cascade laser and use as a local oscillator in a heterodyne HEB receiver," Opt. Express 17, 1159 (2009).
- [7] Y. Ren, J. N. Hovenier, R. Higgins, J. R. Gao, T. M. Klapwijk, S. C. Shi, A. Bell, B. Klein, B. S. Williams, S. Kumar, Q. Hu, and J. L. Reno, "Terahertz heterodyne spectrometer using a quantum cascade laser," Appl. Phys. Lett. 97, 161105 (2010).

4

- [8] Y. Ren, J. N. Hovenier, R. Higgins, J. R. Gao, T. M. Klapwijk, S. C. Shi, B. Klein, T.-Y. Kao, Q. Hu, and J. L. Reno, "High-resolution heterodyne spectroscopy using a tunable quantum cascade laser around 3.5 THz," Appl. Phys. Lett. 98, 231109 (2011).
- [9] S. Ryabchun, C.-Y. Tong, S. Paine, Y. lobanov, R. Blundell, and G. Gol'tsman, "Temperature Resolution of an HEB receiver at 810 GHz," IEEE Trans. Appl. Supercond. 19, 293 (2009).
- [10] M. I. Amanti, M. Fischer, G. Scalari, M. Beck, and J. Faist, "Low-divergence single-mode terahertz quantum cascade laser," Nat. Photonics 3(10), 586–590 (2009).
- [11] M. I. Amanti, G. Scalari, F. Castellano, M. Beck, and J. Faist, "Low divergence Terahertz photonic-wire laser," Optics Express, 18, 6390, (2010)
- [12] B. Klein, I. Krämer, S. Hochgürtel, R. Güsten, A. Bell, K. Meyer, and V. Chetik, "Fast Fourier Transform Spectrometer," Proceedings of the 20th International Symposium on Space Tera-hertz Technology (ISSTT), Charlottesville, (2009).
- [13] H. B. Callen and T. A. Welton, "Irreversibility and Generalized Noise," Phys. Rev. 83, 34 (1951)
- [14] J. W. Kooi, J. J. A. Baselmans, A. Baryshev, R. Schieder, M. Hajenius, J. R. Gao, T. M. Klapwijk, B. Voronov, and G. Gol'tsman, "Stability of heterodyne terahertz receivers", J. Appl. Phys. 100, 064904 (2006)
- [15] Y. Ren, M. Cui, J.N. Hovenier, D.J. Hayton, J.R. Gao, T.M. Klapwijk, S.C. Shi, T-Y. Kao, Q. Hu, and J. L. Reno, "Frequency locking of a 3.5 THz quantum cascade laser using a gas cell," Proceedings of the 22th International Symposium on Space Tera-hertz Technology (ISSTT), Tucson, (2011).