

Terahertz heterodyne spectrometer using a quantum cascade laser

Y. Ren^{1,2,3}, J.N. Hovenier¹, R. Higgins⁴, J.R. Gao^{1,5}, T.M. Klapwijk¹, S.C. Shi², A. Bell⁶, B. Klein⁶, B. S. Williams⁷, S. Kumar⁷, Q. Hu⁷, and J. L. Reno⁸

¹Kavli Institute of NanoScience, Delft University of Technology, Lorentzweg 1, 2628 CJ Delft, The Netherlands

²Purple Mountain Observatory (PMO), Chinese Academy of Sciences, 2 West Beijing Road, Nanjing, JiangSu 210008, China.

³Graduate School, Chinese Academy of Sciences, 19A Yu Quan Road, Beijing 100049, China

⁴Department of Experimental Physics, National University of Ireland, Maynooth, Ireland

⁵SRON Netherlands Institute for Space Research, Sorbonnelaan 2, 3584 CA Utrecht, The Netherlands

⁶Max-Planck-Institut für Radioastronomie (MPIfR), Auf dem Hügel 69, 53121, Bonn, Germany

⁷Department of Electrical Engineering and Computer Science, Massachusetts Institute of Technology, Cambridge, Massachusetts 02139, USA.

⁸Sandia National Laboratories, Albuquerque, NM 87185-0601, U.S.A.

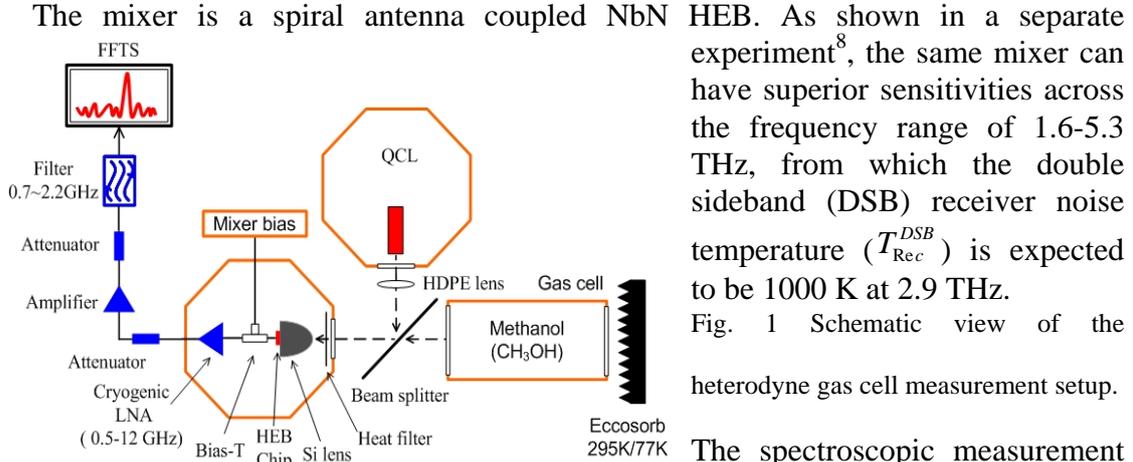
A terahertz (THz) heterodyne spectrometer is demonstrated based on a quantum cascade laser (QCL) as a local oscillator (LO) and an NbN hot electron bolometer (HEB) as a mixer, and it is used to measure high-resolution molecular spectral lines of methanol (CH₃OH) between 2.913-2.918 THz. The spectral 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.

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 and excellent sensitivity. Heterodyne receivers based on superconducting 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 lasers¹ (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 blackbody radiation (hot/cold loads) as the calibration source^{2,3}. A number of milestones for use a THz QCL as an LO have been demonstrated, such as phase-locking capability⁴, narrow intrinsic linewidth⁵, and excellent power stability². 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 spectroscopy⁶. 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 design⁷. 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 shown in a separate experiment⁸, 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_{Rec}^{DSB}) is expected to be 1000 K at 2.9 THz.

Fig. 1 Schematic view of the heterodyne gas cell measurement setup.

The THz radiation beam from the QCL first passes through the high-density polyethylene (HDPE) cryostat window and is then focused with a HDPE 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. 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. 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 spectroscopic measurement setup is sketched in Figure 1.

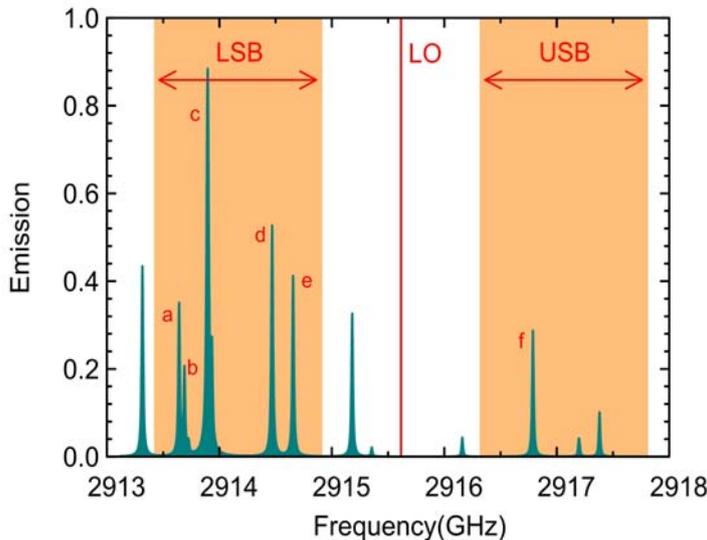


Fig. 2 (Colour online) Calculated emission spectra of methanol (CH_3OH) at a pressure of 1 mbar and at 300 K, for a 0.5 m optical path length. The red line indicates the LO at 2915.6 GHz. The shadow regions of the lower side band (LSB) and the upper side band (USB) correspond to the detection band of the FFTS for the IF signal by using a 0.7-2.2 GHz band pass filter. Several methanol lines within the shadow regions, labelled with “a” to “f”, are expected to be seen.

The back-end spectrometer is a Fast Fourier

Transform Spectrometer (FFTS)¹⁰. 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, labelled with “a” to “f” in Figure 2, distributed in the two corresponding bands (LSB and USB) at the THz frequencies.

To measure the spectral lines of the gas, three IF power spectra were measured¹²: 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 seconds. With the three spectra, the brightness of the emission lines in terms of temperature is calculated using the following expression¹²

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

where T_{hot} and T_{cold} are the effective hot and cold load temperatures defined by the Callen-Welton formula¹³, and the factor of 2 in Eq. 1 is derived from the DSB mode operation of the HEB mixer.

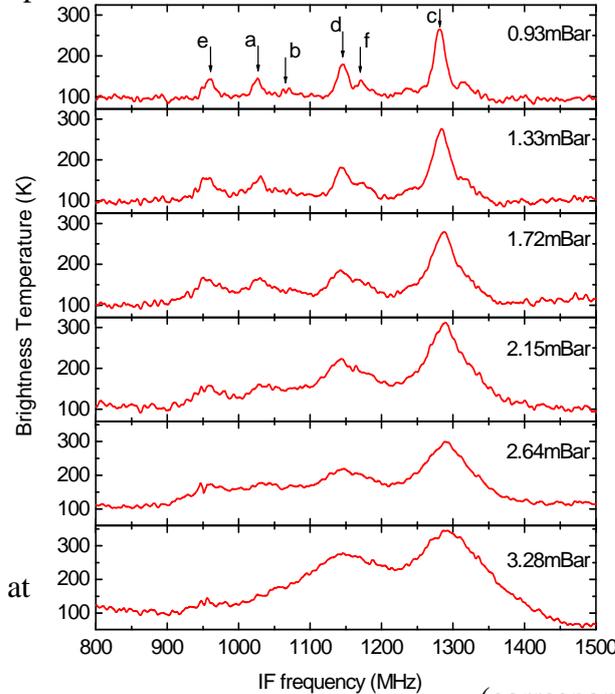
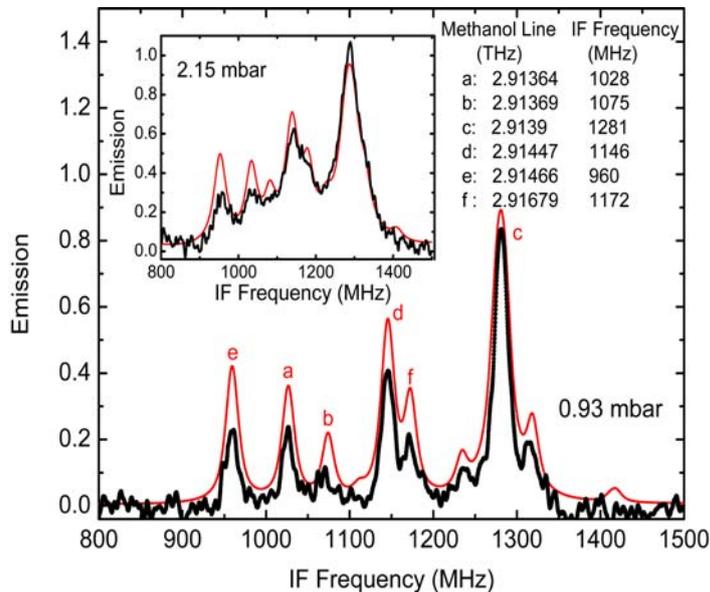


Fig. 3 (Colour online) Measured methanol (CH_3OH) emission spectra within the IF range between 0.8 and 1.5 GHz at different gas cell pressure which varies from 0.93 mbar to 3.28 mbar. The QCL's frequency is 2915.6 GHz.

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 1281 MHz is observed, which is assumed to be the “c” line at 2913.896 GHz in the LSB of Figure 2 (corresponding to P-transition, from the upper state ($J=22, K=9, v=0$) to the lower state ($J=23, K=10, v=0$))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 catalog¹⁴ and also the effects of line broadening, the parameters of the gas cell, and the FFTS spectrometer were included. 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.

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. 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.

Fig. 4 (Colour online) Measured methanol emission spectra (black curve) and the simulated spectra (red curve) at a gas pressure of 0.93 mbar.

References

- [1] B. S. Williams, *Nature photonics*, 1, 517 (2007).
- [2] 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, *Appl. Phys. Lett.* 86, 244104 (2005).
- [3] 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, *Optics Express*, 13, 5890(2005).
- [4] 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, *Opt. Lett.* 34, 2958 (2009)
- [5] A. Barkan, F. K. Tittel, D. M. Mittleman, R. Dengler, P. H. Siegel, G. Scalari, L. Ajili, J. Faist, H. E. Beere, E. H. Linfield, A. G. Davies, and D. A. Ritchie, *Opt. Lett.* 29, 575 (2004).
- [6] H.-W. Hübers, S.G. Pavlov, H. Richter, A.D. Semenov, L. Mahler, A. Tredicucci, H.E. Beere, and D.A. Ritchie, *Appl. Phys. Lett.* 89, 061115 (2006).
- [7] B. S. Williams, S. Kumar, Q. Hu, and J. L. Reno *Opt. Exp.* 13, 3331(2005).
- [8] W. Zhang, P. Khosropanah, J.R. Gao, E. L. Kollberg, K. S. Yngvesson, T. Bansal, R. Barends, and T. M. Klapwijk, *Appl. Phys. Lett.* 96, 111113 (2010).
- [9] R. D. Higgins, D. Teyssier, J. C. Pearson, C. Risacher and N. A. Trappe, 21st International Symposium on Space Terahertz Technology (ISSTT 2010), Oxford.
- [10] B. Klein, I. Krämer, S. Hochgürtel, R. Güsten, A. Bell, K. Meyer, and V. Chetnik, 20th International Symposium on Space Terahertz Technology (ISSTT 2009), Charlottesville.
- [11] J. Kauppinen, J. Partanen, *Fourier Transforms in Spectroscopy*, Wiley-VCH Verlag GmbH, Berlin (2001).
- [12] S. Ryabchun, C.-Y. Tong, S. Paine, Y. Iobanov, R. Blundell, and G. Gol'tsman, *IEEE Trans. App. Supercond.* 19, 293 (2009). Note the type error, missing a factor of two in front of the second term in equation 4.
- [13] H. B. Callen, and T. A. Welton, *Phys. Rev.* 83, 34 (1951).
- [14] H. M. Pickett, R. L. Poynter, E. A. Cohen, M. L. Delitsky, J. C. Pearson, and H. S. P. Muller, *J. Quant. Spectrosc. & Rad. Transfer*, 60, 883 (1998)