Widely tunable quantum cascade laser-based terahertz source

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A compact, tunable, ultranarrowband terahertz source, $\Delta\nu \sim 1$ MHz, is demonstrated by upconversion of a 2.324 THz, free-running quantum cascade laser with a THz Schottky-diode-balanced mixer using a swept, synthesized microwave source to drive the nonlinearity. Continuously tunable radiation of 1 μW power is demonstrated in two frequency regions: $\nu_{\mathrm{Laser}} \pm 0$ to 50 GHz and $\nu_{\mathrm{Laser}} \pm 70$ to 115 GHz. The sideband spectra were characterized with a Fourier-transform spectrometer, and the radiation was tuned through CO, HDO, and D_2O rotational transitions.

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1. Introduction

The recent report of harmonic frequency-locking [1] and phase-locking of a 2.3 THz quantum cascade laser using Schottky-diode MMICs [2] mounted in fundamental waveguide THz-balanced mixers [3] illustrates the considerable potential of the THz QCL/balanced mixer system. Compared with earlier generation mixers, e.g., whisker-contacted Schottkys mounted in open resonator structures, the new devices possess greater nonlinearity, isolation of local oscillator (LO) and signal ports, increased power handling capability, and a flat IF response extending well beyond 100 GHz. Improvement in upconversion, i.e., sideband (SB), efficiency of the THz-balanced mixer has also been shown [4], using THz optically pumped lasers.

In this paper, the balanced mixer is employed for generation of SBs on a solid-nitrogen-cooled (SN_2) quantum cascade laser (QCL) source, and the SBs are used for high-resolution molecular spectroscopy of deuterium oxide (D_2O), HDO, and carbon monoxide (CO). Previously, SB radiation from a liquid helium-cooled 2.408 THz QCL, with a corner-cube-mounted, whisker-contacted Schottky-diode was reported [5]. Using the balanced mixer, significant improvement in SB power, spatial mode quality and bandwidth, as well as much better rejection of the unshifted laser radiation is demonstrated, with the potential for several hundred gigahertz of continuously tunable radiation.

In the present work, the steady-state frequency of the QCL is determined mainly by the equilibrium temperature reached by the device and can vary by several hundred MHz, depending on the form and location of the SN_2 mass, which determines the heat transfer. The frequency then stabilizes to

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~1 MHz and can be determined to that precision by measuring tabulated molecular transitions with the SB radiation. Greater accuracy can be obtained by adding a second balanced mixer, operating as a harmonic mixer. Then, the free-running QCL frequency can be locked [1], referenced to a microwave standard, and the SB resolution improved to the kilohertz level.

2. Continuous Wave THz Quantum Cascade Laser

A 2324 GHz laser was chosen for this work. This laser had an approximately 3 mm cavity length. The laser continues to radiate over 2 mW CW, at 60 K, from a single facet, and emits radiation in a single-longitudinal mode (SLM) up to 4.5 V DC bias. At higher bias voltages, three longitudinal modes, approximately 13 GHz apart, are observed. The laser design is based on the structure published by Vitiello *et al.* [6], where the bound-to-continuum energy level design and the semi-insulating surface plasmon waveguide structure are employed.

The QCL Dewar chamber was kept at SN_2 temperatures, which was achieved by pumping on liquid nitrogen until it solidified. After approximately one hour, a dynamic equilibrium between the sublimation and vapor pumping processes is reached inside the Dewar at an equilibrium temperature measured at the QCL mount of approximately 50 K, when the laser is not biased, and approximately 60 K when 5 W of electrical power is dissipated in the laser operating in the CW mode. The Dewar is continuously pumped with a roughing pump and the steady-state temperature is maintained until the SN_2 is nearly completely sublimed. This allows continuous operation of the 2.324 THz QCL for 20 hours with 5 W of DC power consumption [7].

3. Terahertz-Balanced Mixer

In 2008, Erickson built and tested the first fundamental waveguide THz-balanced mixer [3] using a 1.5 THz monolithic membrane-diode (MOMED) designed at UMass and fabricated at JPL [2]. 2.3 THz-balanced mixers were successfully demonstrated in 2013. The THz-balanced mixer has been shown to provide superior performance to corner-cube-mounted, whisker-contacted, Schottky diodes, both as downconverters and SB generators [3]. Figures 1(a) and 1(b) show the layout of the THz mixer MMIC. For use as a mixer-upconverter, the free-space-propagating THz signal is coupled into the RF port of the waveguide block through an integral diagonal feed horn, while the microwave signal is coupled into the IF port, and the SB radiation exits the LO port.

The mixer used in the SB generation experiment was modified by adding a separate W-band waveguide port [Fig. 1(c)] for injecting a higher frequency (70–115 GHz) microwave signal into the internal IF microstrip line, leaving the coaxial IF port for coupling in the lower frequency (2–50 GHz) signal. Figure 1(a) shows the internal details, while Fig. 1(c) is a photo of the modified mixer block.

4. Sideband

Schematic of the SB generation setups are shown in Figs. 2(a) and 2(b). The radiation from one facet of the CW 2.324 THz QCL, operated in a single longitudinal mode was focused into the mixer's RF port with a 9 inch focal length spherical mirror positioned 15° off-axis. Neither temperature nor current stabilization of the QCL was used in this experiment. A hollow dielectric Pyrex tube of 30 mm length and 1.5 mm inner diameter, positioned directly in front of the laser, significantly improved the laser radiation spatial mode quality, and provided a narrow 5° beam divergence [8].

The optimum mixer feed horn position was determined by maximizing the video output signal at low bias current (1 μA). The SB radiation was then generated by mixing the QCL and microwave signals in the balanced mixer, at 1 mA DC bias current. The microwave signal was supplied by a 0–67 GHz

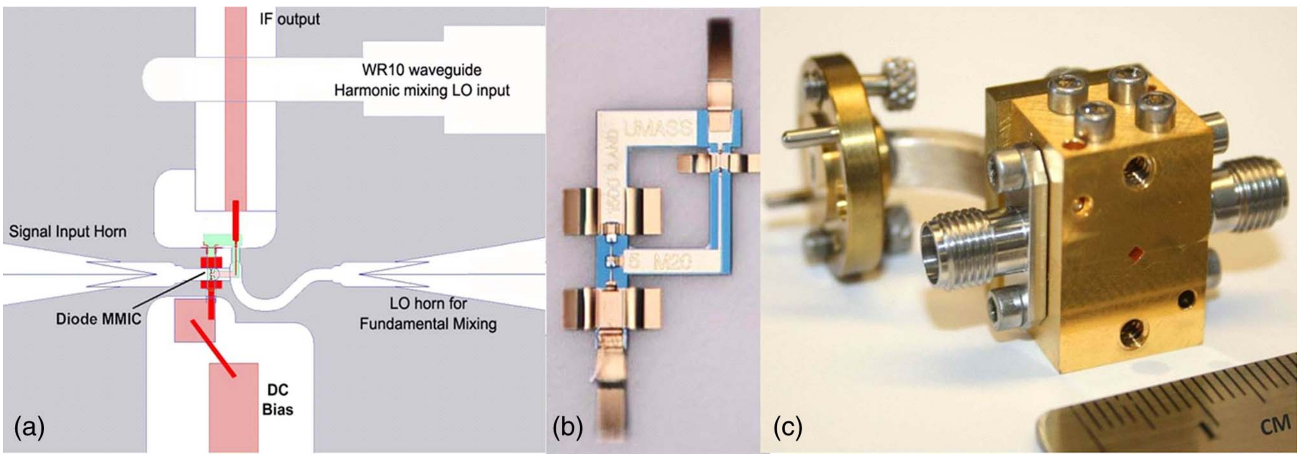


Fig. 1. (a) Schematic diagram of the balanced mixer showing the WR10 waveguide that is used as the LO input, as well as the LO feed horn that is not used for harmonic mixing. The horn on the left side is used for the QCL signal input. (b) Photograph of the device (taken from Ref. [3]). Green areas are the GaAs substrate and gold areas are metal on the MMIC chip. (c) Photograph of the mixer block with the W-band waveguide input.

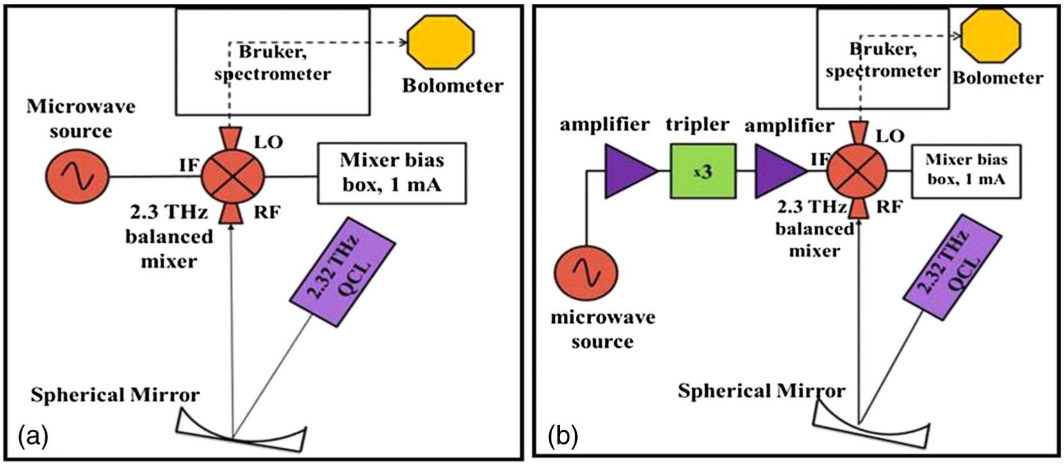


Fig. 2. Setup used for SB spectral measurements.

synthesized sweeper. Microwave power coupled to the mixer was varied with frequencies from −20 to 4 dBm, and the DC bias voltage was monitored to prevent damage to the mixer. There were two frequency branches of generated SB radiation: low (2–50 GHz) and high (70–115 GHz) frequency SBs. Low frequency signal was supplied to the IF port of the mixer through an SMA cable. An amplifier/tripler/amplifier chain was driven by the same synthesizer to produce a high frequency output. This signal was coupled into the mixer's WR 10 waveguide port to obtain high frequency SB radiation. The 20 GHz gap (50–70 GHz) was due to a combination of the high frequency loss of the RF cable used for injection into the IF port of the mixer and the bandwidth limitation of the RF amplifier in the tripler chain, rather than any problem with the IF response of the mixer.

For spectral analysis, the signal from the LO port was directed into the emission port of a FTIR spectrometer (0.125 cm^{-1} resolution), and detected with a liquid He-cooled Si bolometer, as shown in Figure 2.

Composite spectra of the low and high frequency SBs are shown in Figs. 3(a) and 3(b), respectively.

Both upper and lower SB can be seen in Fig. 3(a). Absent from the spectra in Fig. 3(b) is the lower SB that was strongly absorbed along the 0.5 m atmospheric path length from the SD to the spectrometer by the strong water line at 2.264 THz. Another water line at 2.345 THz explains the absence of the upper SB radiation around 24 GHz [Fig. 3(a)]. A weak blackbody (BB) spectrum can be seen as a baseline in the spectra. In the case of rapid scan modulation, BB radiation from the room temperature, the evacuated spectrometer box was directly modulated, together with the SB radiation, by the moving mirror, and is present in the CW spectra. The unshifted QCL radiation is also present in the spectra, with intensity roughly equal to the SB signal. The QCL signal was observed to double when the microwave signal was coupled to the mixer, in comparison with the case when the microwave source was turned off. Small dips to the left of the emission peaks are believed to be artificial spectral features, created by a fast Fourier-transformation.

To measure the SB power, the output beam was sent directly onto the bolometer with a measured responsivity of 2.08×10^5 V/W. Double sideband (DSB) power (both SBs) of 1 μW was generated with 200 μW of drive power from the laser operating in a single longitudinal mode. This implies a power upconversion efficiency of 5×10^{-3}, which represents a 10-fold improvement over the reported corner-cube data on QCL-based THz SB generation [5]. The potential upconversion efficiency is even greater, since coupling

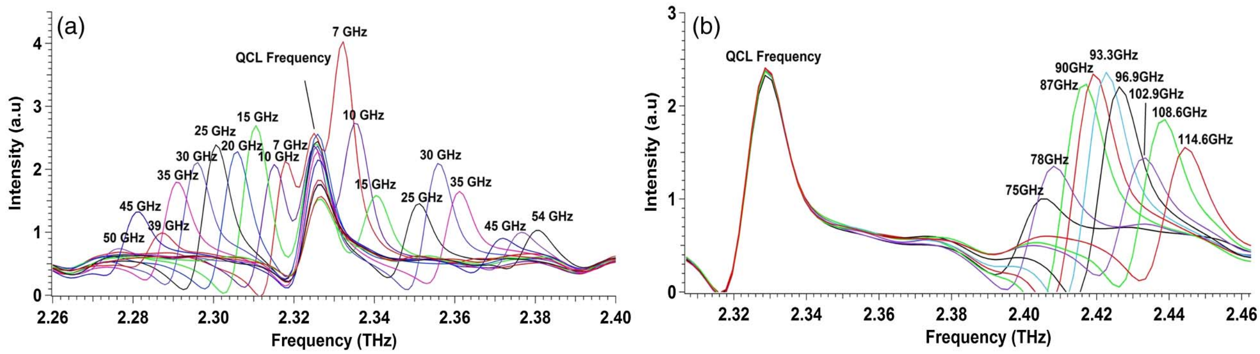


Fig. 3. Spectra of the low (a) and high (b) frequency SBs produced by applying different microwave frequencies in CW regime with the 2.32 THz QCL operating in a single longitudinal mode.

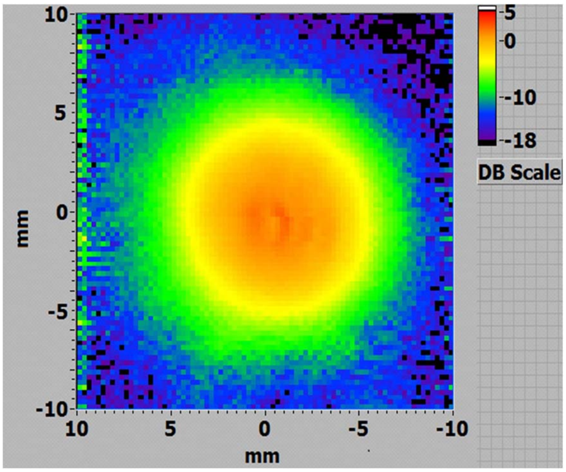


Fig. 4. 2D mode scan of the SBs and unshifted laser.

of the QCL radiation into the mixer waveguide and onto the device is constrained by the mode quality of the QCL beam. An efficiency of 3×10^{-2} has been reported at 1.56 THz using a TEM_{00}-mode optically pumped laser line and a 1.5 THz-balanced mixer [4].

Another advantage of the balanced mixer is the considerable isolation between the LO and RF ports, which correspond to the QCL input and SB output signals, respectively. As a result, the unshifted laser signal at the output port was only 0.5 μW, implying an approximate unity ratio of single-sideband (SSB) versus unshifted laser power. In contrast, the unshifted laser power reradiated by a corner-cube mixer antenna is 10–20 dB greater than the SB signal. There, to isolate the SB radiation from the much stronger laser signal, frequency-selective components (diplexers or etalons) are required. The isolation provided by the balanced mixer will simplify and, in some cases, eliminate the necessity for such components.

The fundamental waveguide and diagonal horns of the balanced mixer design generate SBs with excellent spatial mode quality, as shown in Fig. 4. This will enable the SBs to be employed in many applications, where a spatially coherent beam is required. Terahertz SB radiation from a corner-cube reflects its poor antenna pattern and requires spatial filtering; therefore, power degradation produces a profile that begins to approximate the pattern in Fig. 4.

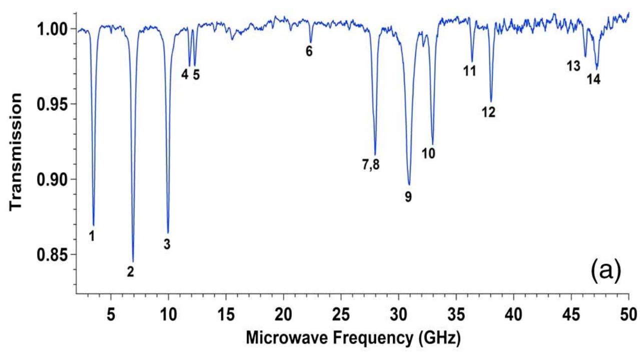
5. Gas Spectroscopy on D_2O, HDO, and CO

SB spectral resolution is limited by temperature and current fluctuations in the QCL. With respect to temperature, the SN_2 cryogen system has achieved a QCL frequency drift of ±650 kHz over an extended time period (1 h 40 min) and considerably less drift over durations required for typical spectral measurements [7]. To determine the resolution and demonstrate the large useful bandwidth of the QCL-driven SB source, rotational transitions in CO, D_2O, and HDO were measured, both in the low pressure, Doppler-broadened regime, and at higher pressures.

For these experiments, the spectrometer was replaced with a 55 cm long gas cell with wedged TPX input and output windows. SB radiation was directed through the cell and then focused onto the bolometer with an off-axis parabola. The detected signal was measured with a lock-in amplifier and saved to a computer through a data acquisition card. The maximum synthesizer sweep time of 100 s was used. Sampling at the preset number of 1600 points, within a frequency range of 48 GHz (2–50 GHz), gave an incremental step of 30 MHz.

Figure 5(a) displays the D_2O and HDO (the sample is a mixture of two gases) spectra measured at a gas pressure of 0.5 Torr, as the microwave source was swept from 2 to 50 GHz. The plots are ratios of scans taken with and without gas samples in the cell.

Fourteen lines of D_2O and HDO were identified [9] as transitions at the first-order upper sideband (USB) and lower sideband (LSB), and listed in Table 1 with their tabulated intensities and frequencies ("intensity" is the base 10 logarithm of the integrated intensity). The relatively large frequency step (30 MHz) used in the 2–50 GHz synthesizer sweep is the limiting factor in determining the precision of the transition frequencies. Lines 7 and 8 are overlapped and cannot be resolved at this resolution. Figure 5(b) shows the spectrum of CO. The CO rotational transitions are more sparsely distributed and only one transition, which overlaps with the LSB, is observed. To determine which SB generates the transition, a well-known technique is used, where an intentional frequency change of the QCL leads to a corresponding shift of the microwave frequency at resonance. The direction of the shift reveals whether



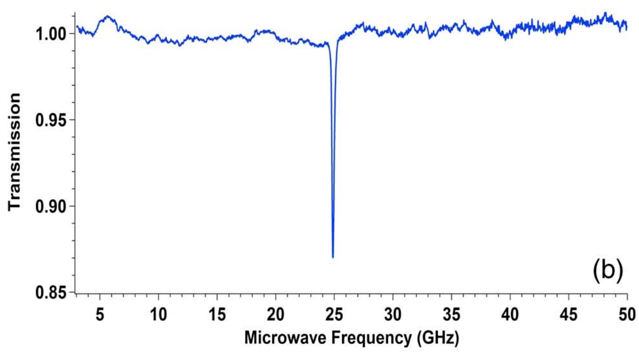


Fig. 5. Spectra of (a) D_2O/HDO at 500 mTorr and (b) CO at 10.1 Torr produced by sweeping the microwave frequency from 2 to 50 GHz.

Table 1. List of Measured Rotational Transitions of D_2O and HDO

Line No.	Sideband	Molecule	Tabulated Freq. (MHz) [9]	Tabulated Intensity [9]	Microwave Freq. (GHz)
1	Upper	HDO	2327428.7661	−0.5485	3.47
2	Upper	D_2O	2330852.8459	−0.8852	6.92
3	Upper	D_2O	2333888.4524	−0.8406	9.95
4	Upper	HDO	2335787.6952	−1.6298	11.81
5	Upper	HDO	2336219.6051	−1.6296	12.29
6	Lower	D_2O	2301705.7221	−2.9305	22.34
7	Upper	HDO	2351731.3737	−0.4094	27.95
8	Lower	HDO	2296248.2228	−0.8915	27.95
9	Lower	D_2O	2293369.4818	−0.1585	30.89
10	Lower	HDO	2291309.2981	−0.1126	32.96
11	Upper	HDO	2360339.9528	−1.1497	36.38
12	Lower	HDO	2286212.6389	−0.5202	38.03
13	Lower	HDO	2278017.0944	−0.5630	46.19
14	Lower	D_2O	2277014.1173	−0.6227	47.21

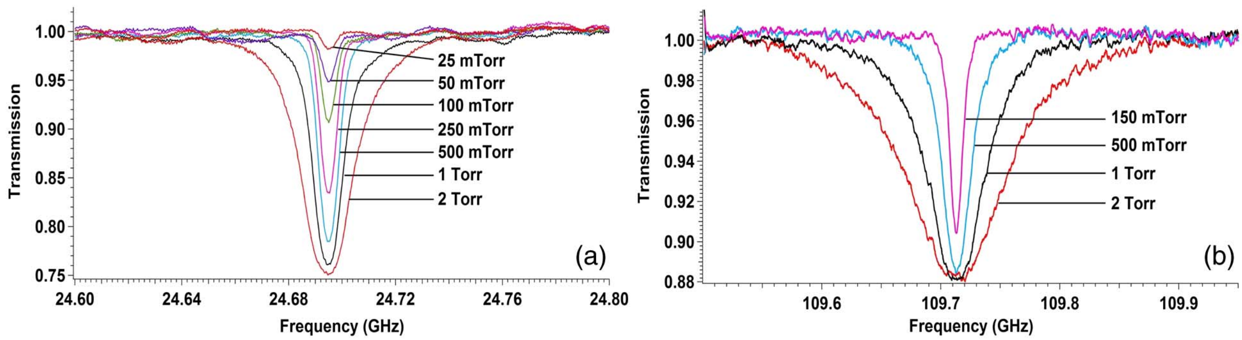


Fig. 6. (a) CO spectra lower SB sweep and (b) D_2O spectra upper SB sweep.

the transition was generated by the LSB or USB. In conducting the spectroscopy measurements, no attempt was made to filter out the unshifted laser radiation or to separate the USB and LSB. Therefore, the transmitted intensities at resonance do not directly relate to the maximum absorption coefficient $\alpha_{\max}$.

To examine a transition at higher resolution, a series of SB sweeps over the $J = 20 \leftarrow 19$ CO rotational line were taken at different pressures. The synthesizer was swept over 200 MHz and sampled at 1600 points to yield an incremental step of approximately 125 kHz. Figure 6(a) displays plots of the relative transmitted intensities as the CO pressure was varied from 2 to 25 mTorr. The theoretical Doppler linewidth for this transition is 5.4 MHz, which is in very good agreement with the measured FWHM of 5.3 ± 0.2 MHz at pressures below 100 mTorr. Deconvolution allowed us to separate an instrumental effect (i.e., spectral line broadening due to QCL linewidth) from the measured spectral line profile and to estimate the system's spectral resolution to better than 1 MHz. CO rotational transitions have been measured and calculated to a precision of 10 kHz [10]. In particular, the $J = 20 \leftarrow 19$ transition is centered at 2,299,570 MHz; thus, the QCL frequency was at 2,324,264 MHz during the course of these measurements.

High-resolution sweeps of the D_2O transition centered at 2,433,780 MHz, at different pressures, are shown in Fig. 6(b) to illustrate the useful tuning range of the system, which extends beyond ± 100 GHz. For these measurements, which were conducted approximately two weeks after the CO results were obtained, the QCL frequency calculates as 2,324,067 MHz. The ~200 MHz shift in operating frequency is attributed mainly to different steady-state laser temperatures and, to a lesser extent, different DC bias conditions. This illustrates the necessity, as pointed out earlier, for locking the laser frequency to a stable reference to insure a reproducible, highly accurate and precise QCL-driven SB source.

6. Conclusion

Continuous tuning of a CW 2.3 THz source with 1 MHz spectral resolution and excellent spatial mode quality has been demonstrated using a QCL-driven Schottky-diode-balanced mixer as an upconverter. The ± 100 GHz tuning range can be further expanded with higher frequency millimeter wave amplifier/multiplier sources. SB power levels, currently about 1 μW, will increase with higher-power QCLs and laser designs with better spatial mode quality. Spectral resolution can be further improved by locking the QCL to a stable reference source.

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