Demonstration of a room temperature 2.48–2.75 THz coherent spectroscopy source

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We report the first demonstration of a continuous wave coherent source covering 2.48-2.75 THz, with greater than 10% instantaneous tuning bandwidth and having 1–14 μ W of output power at room temperature. This source is based on a 91.8-101.8 GHz synthesizer followed by a power amplifier and three cascaded frequency triplers. It demonstrates for the first time that purely electronic solidstate sources can generate a useful amount of power in a region of the electromagnetic spectrum where lasers (solid state or gas) were previously the only available coherent sources. The bandwidth, agility, and operability of this THz source have enabled wideband, high resolution spectroscopic measurements of water, methanol, and carbon monoxide with a resolution and signal-to-noise ratio unmatched by any other existing system, providing new insight in the physics of these molecules. Furthermore, the power and optical beam quality are high enough to observe the Lamb-dip effect in water. The source frequency has an absolute accuracy better than 1 part in 10¹² and the spectrometer achieves sub-Doppler frequency resolution better than 1 part in 108. The harmonic purity is better than 25 dB. This source can serve as a coherent signal for absorption spectroscopy, a local oscillator for a variety of heterodyne systems and can be used as a method for precision control of more powerful but much less frequency agile quantum mechanical terahertz sources. © 2011 American Institute of Physics. [doi:10.1063/1.3617420]

I. INTRODUCTION

The region between 1 and 10 THz corresponds to the peak of thermal emission from dust and molecules throughout the Universe.¹ It also contains the Boltzmann peak of emission from the rotational bands of light molecules at temperatures of planets including the Earth.² In the 1-10 THz region, the spontaneous emission linewidth of molecules is less than 1 MHz, so that the observed line shape is entirely due to Doppler and pressure broadening effects.³ This spectral region contains many phonon modes of solid state materials, potentially allowing fingerprinting of compounds.⁴ Unfortunately, generation and control of radiation in the 1-10 THz region is particularly challenging for electronic systems where parasitic capacitance limits circuit elements much smaller than a wavelength and for quantum electronics where thermal excitation limits the population difference in closely spaced energy levels. In spite of the technical difficulties, the terahertz regime is particularly well suited to perform remote sensing since all polar molecules feature a rotational band in this frequency region. As such, there has been an explosion of approaches to cover this "THz

The present system was constructed to be used as a local oscillator for receivers operating in the 2.5–2.7 THz range. The primary interest in this band is the J=1-0 line of HD, the simplest closed shell heteronuclear diatomic molecule. In astrophysics, the HD abundance is important as a tracer of the cosmologically significant D/H ratio. The HD also serves as a tracer of the total column of molecular gas and is expected to figure prominently in the formation of the

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gap" region of the electromagnetic spectrum. However, these approaches have often been severely limited in frequency tuning range, instantaneous bandwidth, sensitivity, and often require cryogenic operation. Many types of system implementations require precise knowledge of frequency, a high degree of coherence, and the ability to modulate, tune, and select frequencies rapidly. The THz gap can be addressed by electro-optical down conversion, electronic up conversion, or by direct quantum mechanical frequency generation. To date, there is no preferred approach, since no single method has achieved all the desired characteristics. We present here a relatively simple completely solid state room temperature electronic source covering 2.48-2.75 THz (> 10% instantaneous and tuning bandwidth) with an absolute frequency accuracy better than one part in 10^{12} . This system is suitable for both high and low resolution applications within this band and demonstrates the potential for conventional electronics to ultimately fill the THz gap.

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first generation of stars.⁷ In planetary science, HD provides critical insight into the evolution of giant planets and the atmospheric evolution of rocky planets and moons.⁸ A local oscillator system must provide enough tuning range to cope with significant Doppler shifts, has a precisely known frequency for determination of Doppler shifts, and a linewidth significantly narrower than the signal being received so that the received signal is preserved.

A large number of systems have been built in an attempt to address various applications in the terahertz frequency range. Broadband systems include terahertz time domain⁹ and Fourier transform spectrometers with thermal 10 and synchrotron sources. 11 More recently, the beat of two laser frequency combs has been used as a means to perform Fourier transform spectroscopy. 12 In all of these approaches, it is very difficult to obtain Doppler limited spectral resolution and impossible to obtain the sensitivity available to a coherent source. Coherent sources have been produced by direct photomixing of two¹³ or three lasers, ¹⁴ mixing two lasers and a microwave signal in tunable far infrared (TuFIR)¹⁵ and mixing a single laser and a microwave source in laser sideband. 16 All these approaches are difficult to tune, have low or very low output power, relatively small instantaneous bandwidth, and require great care in obtaining precise frequency calibration and coherence. 14-18 Recently, terahertz quantum cascade lasers (QCL) have shown enormous potential as a terahertz sources; 19 however, a number of significant practical problems remain to be solved, such as large cryogenic heat lift, beam quality,²⁰ and a means for precise frequency calibration and control.^{21,22}

Upconversion of electronically generated sources either for signal control or signal generation remains the preferred method of radiation production below 1 THz and practically the only one below 0.3 THz. In our case, an electronically generated source is a device where oscillation is achieved through interaction with a resonant circuit, gain is achieved with classical transistor-like devices, and harmonics are generated (multiplied) with nonlinear solid state devices. Signal control implies that the frequency accuracy and phase of a reference is transferred to the output either through a phaselock loop or the 20Log(N) direct multiplication phase noise relationship. Electronic upconversion has been the method of choice in laboratory molecular spectroscopy, ^{23,24} remote sensing,²⁵ network analysis,²⁶ and communications.²⁷. We present here a spectroscopic demonstration of a 2.48-2.75 THz system that could have been equally well used for network analysis, covering both low and high resolution applications and discuss the excellent prospects of this approach to fill the terahertz gap.

II. SYSTEM DEMONSTRATION

The source being discussed here is constructed from three cascaded frequency triplers, which together multiply the frequency of the input (at ~ 100 GHz) by a factor of 27 (see Fig. 1). The first tripler utilizes a waveguide hybrid coupler at its input with two two-way splitters and four multiplier devices in parallel followed by two two-way hybrid couplers at the output, to withstand large input power levels. Each

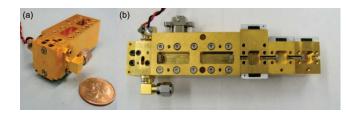


FIG. 1. (Color online) (a) Photo of the three cascaded frequency multipliers showing the 0.69×0.69 mm integrated diagonal feed horn in the 2.7 THz output stage. (b) Photo of the multipliers with two GaAs amplifiers followed by a larger GaN amplifier. A significantly larger power combined GaAs amplifier was used for the measurements described.

multiplier device is composed of six anodes in a balanced configuration on a Schottky planar membrane technology.²⁸ This multiplier generates \sim 35 \pm 3 mW of power when driven with 400 mW input power between 92 and 102 GHz.²⁹ The second tripler utilizes a two-way power splitter and two devices in parallel followed by a two-way combiner to generate \sim 1.3 mW of power in the 828–918 GHz region when driven by the preceding tripler.³⁰ Each multiplier device utilizes four varactor anodes in a balanced configuration on Schottky planar membrane technology. The final tripler, shown in Fig. 2, is a balanced two anode planar Schottky multiplier which achieves $2-14 \mu W$ output over the 2.49-2.72 THz range when driven by the preceding two triplers. RF power can be generated with a power combined GaAs amplifier³¹ or a GaN amplifier³² providing the required 400+ mW of RF power in the 92-102 GHz band. Figure 1 shows a picture of the multiplier chain with a GaN amplifier attached. Figure 2 shows the final stage tripler mounted in half of the split waveguide block. The diode is the smallest yet constructed with the planar technology utilized. The dimensions of the contact are $0.2 \mu m$ by $0.3 \mu m$. Electron beam lithography can fabricate significantly smaller features in one dimension, 33 but the necessity for small size in two dimensions is more challenging. However, further reduction in feature size is likely to be easily achievable with present fabrication technology.

Demonstrations of source performance utilized the spectrometer system are described in Drouin *et al.*²⁴ The input signal was generated by an Anritsu 69253A or HP 8341B synthesizer (each with phase noise of < 100 dBc/Hz at 10 kHz

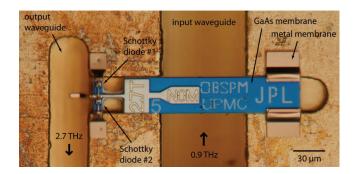


FIG. 2. (Color online) Split block mounting of the 2.7 THz balanced tripler device. Beam leads are used to hold the 3 μ m thick device in place and provide the ground contact for the diodes in the balanced antiparallel configuration. The integral waveguide probes can be seen projecting into the small output waveguide at the left and much larger input waveguide in the center.

offsets) locked to a frequency standard with an accuracy of better than 1 part in 10¹² followed by a Millitech AMC-10 active x6 multiplier with output in the 75–110 GHz range. This was the input signal that was amplified by the W-band power amplifier to drive the cascade of three frequency triplers. Bias on the terahertz frequency multipliers and W-band amplifiers was fixed. The GaAs amplifier driven chain had conversion efficiency (dc to RF) of 1.8×10^{-6} while a similar GaN amplifier driven chain has an efficiency of 3.5×10^{-6} . Signal control is achieved at the synthesizer with amplitude modulation being used for power spectra and tone burst or frequency modulation for second derivative spectra. Two detectors were used; a room temperature 900 GHz pumped Schottky diode used as a sub-harmonic mixer³⁴ and a 4 K Infrared Labs (LN6-C) composite silicon bolometer. All the signal processing is performed below 20 GHz. The terahertz electronics have 270 GHz of instantaneous bandwidth. As such, any arbitrary waveform including modulation, chirps, or spread spectrum could be generated at frequencies below 20 GHz and upconverted to the output band. The frequency accuracy, spectral resolution, and signal purity are entirely determined by the input signal and the usual 20log(N) phase noise growth, where N is 162 in this configuration. Like saturated amplifiers, multipliers efficiently produce intermodulation products so that multiple frequency signals will mix according to the previous relationship. The subsequent subsections describe a number of experiments, which demonstrate (1) the spectral characteristics, (2) the precision, control, accuracy, and sensitivity, (3) high sensitivity THz spectroscopy without cryogenics, and (4) the broad spectral coverage obtained by the THz source.

A. Spectral characteristics

A major consideration in the construction of any system, which performs electronic up conversion, is the level of spurious response. This can include harmonically related spurs generated by imbalance in the frequency multipliers, nonharmonically related spurs from parametric oscillations, and pick up of other signals. The first test injected the output of the multiplier chain at several different frequencies into a Bruker IFS-125 Fourier transform spectrometer. In all cases, two signals were observed, the desired fundamental, and a response of less than 10^{-3} at twice the desired frequency, which was likely aliased. Total signal-to-noise was ~40 dB placing upper limits on spurious content. The second test was to look closer to the carrier than possible with the Fourier transform spectrometer. In this case, a 900 GHz local oscillator was used to drive a 900 GHz fundamental mixer³⁴ used in a subharmonic mode, which down converted the 2.7 THz source so that it could be observed with a spectrum analyzer. The spectrum analyzer and both synthesizers (900 GHz LO and 2.7 THz chain) were locked to a common 10 MHz reference. Observed spectral purity was slightly better than specified for the synthesizer multiplied by 20Log(N) where N is the total harmonic number of 162. Some 60 Hz modulation was observed but is most likely a pickup in one of the lock-loops of the test equipment. The last test was to measure the out-

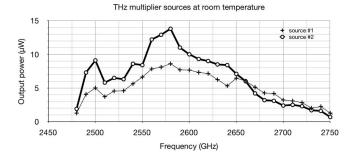


FIG. 3. Calibrated output power spectrum of two THz multiplier sources at room temperature in a pure nitrogen atmosphere. Sources #1 and #2 are two different multipliers of the same design, demonstrating that the performance is repeatable.

put power in a calibrated way. For this purpose, an Erickson PM-4 power meter was used with a nitrogen purge. The path length of the terahertz propagation was kept as short as possible to minimize the impact of any residual water vapor. The results for two completely different multiplier chains are given in Fig. 3.

B. Control, accuracy, and sensitivity

The present multiplier was designed to serve as a local oscillator in high resolution heterodyne spectrometers where the resolution can be as high as $\lambda/\Delta\lambda=10^7$. High resolution absorption spectroscopy where the Doppler width is ~ 1 part in 10^6 and the transition frequencies are constants of nature provide a convenient method of verifying the control and accuracy of a local oscillator source. The sensitivity can also be judged accurately by observing isotopologues in natural abundance. The first test of our system was to observe carbon monoxide and its isotopologues.

Carbon monoxide is one of the most abundant molecules in the Universe and has been very carefully studied in the laboratory so its spectrum has been very well modeled^{35,36} making it useful for evaluating absolute sensitivity and Doppler limited measurement accuracy. A sample of CO was held at 100 mTorr pressure in a 2.3 m absorption cell with high density polyethylene windows and small frequency windows near the absorption features were recorded. Frequencies were determined by fitting the Gaussian lineshape but not any standing waves or power variations. The measurements shown in Table I describe the precision and accuracy of this spectrometer in the context of a similar, albeit much more complex, TuFIR spectrometer. 15 The precision, with which the center of a transition can be determined, is ultimately determined by the signal-to-noise and the ability to cope with systematic errors, such as standing waves. A rule of thumb is that line center can be determined to the sum of half the linewidth divided by the signal-to-noise plus the uncertainty in the absolute frequency. For the strong CO transitions, the line was symmetric suggesting that standing waves and power variation were at 1% or less making the signal-to-noise ratio where the effective noise is the standing wave greater than 100. The Doppler half width at half maximum at 300 K is 1.17 parts in 10^{-6} (~2.7 MHz HWHM) so the accuracy is ~25 kHz.

TABLE I. Measured and calculated CO frequencies.

Transition $J' \leftarrow J$	Observed ^a MHz	Observed ^b MHz	Calculated ^c MHz
22←21	2 528 172.037(25)	2 528 172.060(22)	2 528 172.065(12)
23←22	2 642 330.337(25)		2 642 330.347(14)
24←23	2756387.490(75)	2756387.584(34)	2756 387.586(16)

aMeasured, this work.

Both the J=21 and 22 transitions were observed under nearly ideal conditions, but the J=23 transition had lower signal-to-noise due to being at the extreme upper end of the source bandwidth. The primary difference is that the TuFIR experiments modulated the path length to average standing waves.³⁵ The results on CO were completely consistent with expectations.

The sensitivity of our system was tested by detecting carbon monoxide incorporating the trace isotopes ¹³C, ¹⁸O, and ¹⁷O in natural abundance, 1%, 0.2%, and 0.037%, respectively. Table II provides the measured frequencies and uncertainties of ¹³C¹⁶O, ¹²C¹⁸O, ¹²C¹⁷O, ¹³C¹⁸O, and ¹³C¹⁷O in natural abundance. Both singly substituted isotopologues were readily measured (0.5 s integration time per point) at frequencies between 2500 and 2700 GHz. Using the signalto-noise ratio of ¹²C¹⁸O in natural abundance the absorption sensitivity $\alpha_{\rm min} = 2.5 \times 10^{-6} \text{ cm}^{-1} \text{ s}^{-1/2}$, where $\alpha_{\rm min}$ is the minimum absorption resulting in a change of power that can be detected in a 1 s integration. This sensitivity is on a par with that of similar spectrometers based on powerful, tunable, monochromatic sources³⁷ and in line with the expected value given by the source power, detector noise, and estimated optical transmission. It does suggest that the signalto-noise could be improved by two orders of magnitude with a background limited single mode detector. By comparison, the photomixing sensitivity¹⁴ is $\alpha_{min} = 1 \times 10^{-4} \text{ cm}^{-1} \text{ s}^{-1/2}$ and FTIR sensitivity is at best $\alpha_{\min} = 1 \times 10^{-3} \text{ cm}^{-1} \text{ s}^{-1/2}$. Table II gives a list of the new measurements of CO isotopologues compared to the extrapolated calculations.

The agreement between calculations and measurements is good.

The primary scientific motivation for the present system development is astronomical measurement of the J = 1-0transition of HD to provide insight into the D/H ratio and the total molecular gas column. In order to confirm the performance of the source at the HD frequency, the spectrum of HD was observed in the laboratory. HD features a small 8.83 \times 10⁻⁴ Debye dipole moment arising from the small difference between the center of charge and mass.³⁸ The initial measurement of a commercially prepared HD sample was performed at room temperature and agreed well with the previously reported 2674986.66(15) MHz frequency.³⁹ During this experiment it was realized that there was a substantial pressure shift and that the large Doppler width would limit the measurement accuracy. As a result, the spectrum was measured at 18 K in a 15 cm path as a function of pressure. Figure 4 shows a tone burst modulated (2nd derivative), increasing pressure sequence of parahydrogen broadened HD lines. The strongest signal is 0.56 Torr and the weakest is 7 Torr. The bottom panel shows a blow up of the higher pressure spectra where the noise becomes visible. The linewidth is a complex function of Doppler broadening, collisional (Dicke) narrowing⁴⁰ and pressure broadening. The shift in the line center is apparent as well. The line shift and width were measured as a function of pressure for HD, normal H₂, para H₂, and Helium at 18 K. The modulation depth was fixed so that a convolution method could be used to retrieve the line shape.⁴¹ When using this method, the higher pressure spectra become significantly undermodulated resulting in the apparent loss of intensity. The derived rest frequency at zero pressure is 2 674 986.094(25) MHz or 566 kHz lower than the previous measurement with a factor 6 less uncertainty.⁴²

Spectroscopy of a number of other molecular species has also been performed. The hyperfine structure of OH, 42 several transitions of N_2H^+ , the vibrational difference spectrum of acetylene, 43 and the pure rotation spectra of methane, water, 42 ammonia, 42 and dimethyl ether have also been measured with this source and will be reported elsewhere. It is noted that acetylene and methane have no permanent dipole moment so observing their spectra is also a good demonstration of sensitivity.

TABLE II. Observed and calculated frequencies of CO isotopologues measured in natural abundance.

Transition $J' \leftarrow J$	Species	Observed ^a MHz	Calculated ^b MHz	Maximum J Previous
23←22	¹³ C ¹⁶ O	2 526 491.466(50)	2 526 491.457(16)	29
23←22	$^{12}C^{18}O$	2516912.372(50)	2516912.337(96)	17
23←22	$^{12}C^{17}O$	2 575 801.319(150)	2 575 801.192(291)	7
24←23	$^{13}C^{16}O$	2 635 583.461(25)	2635583.440(18)	29
24←23	$^{12}C^{18}O$	2 625 593.624(50)	2 625 593.575(130)	17
24←23	$^{13}C^{18}O$	2 504 724.604(150)	2504724.365(103)	8
24←23	$^{13}C^{17}O$	2 566 168.975(150)	2566168.757(937)	8
25←24	$^{13}C^{18}O$	2 608 347.720(200)	2608347.305(117)	8
25←24	$^{13}C^{17}O$	2 672 314.453(300)	2 672 314.923(1070)	8

^aMeasured, this work.

^bMeasured, Ref. 35.

^cCalculated, Ref. 36.

^dNote: Uncertainty in () in kHz.

^bCalculated, Ref. 36.

^cNote: Uncertainty given in () in kHz.

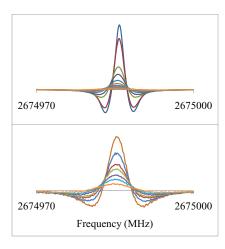


FIG. 4. (Color online) Top panel is the complete sequence of tone burst modulated line measurements made on HD broadened by parahydrogen at 18 K. The modulation and HD pressure were fixed while the parahydrogen pressure was varied resulting in total pressures between 0.56 Torr (strongest line) and 7 Torr (weakest line). The bottom panel is a blow up of the higher pressure measurements from the top panel allowing the noise level to be observed. The apparent loss of line strength is due to the broader lines becoming less efficiently modulated in the convolution method used to retrieve the line parameters (Ref. 41). The zero pressure rest frequency was derived from the complete series of similar measurements including the self-broadening, normal hydrogen broadening, helium broadening, and the data shown here (Ref. 42).

C. Cryogen free THz spectroscopy

Spectroscopy at THz frequency generally requires cryogenic detectors for sufficient sensitivity against the thermal background. However, cryogenics can be problematic in many remote sensing applications where being able to perform sensitive spectroscopy may still be necessary. Three methods of cryogen free spectroscopy have been used in the past. The first uses a room temperature detector, usually a Schottky diode in direct detection, the second uses a mixer as a means of down conversion, and the third utilizes acoustic detection energy deposited in the absorbing medium. We utilized the second approach with a 900 GHz fundamental mixer³⁴ operated in a sub-harmonic mode. The mixer performance was at least one order of magnitude worse than in fundamental mode, but it was trivial to detect the source power and modulation. Figure 5 is a spectrum of water in natural abundance using this system. This experiment along with the previous Doppler limited experiments only proved that the resolution was better than 1 part in 10⁶ and demonstrated that the beam quality from the source was satisfactory.

Saturated absorption spectroscopy provides a stringent method of testing the coherence, power, and beam quality of a source. Observation of a Lamb-dip requires a double pass configuration with the beams precisely overlapping and sufficient power to saturate the molecules. Several recent studies have shown that conventional Lamb-dip measurements were possible up to threshold of the THz gap. $^{44-46}$ An experiment was performed using the $4_{1,4}$ – $3_{0,3}$ transition of ortho water in a 5 cm long double pass cell with the same heterodyne detector used to look at the linewidth details. Optimal sensitivity was achieved with a small (100 kHz depth) frequency modulation of the transmitted signal. The sub-Doppler

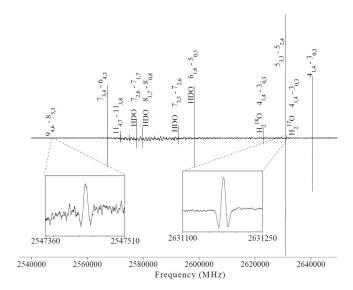


FIG. 5. Room temperature heterodyne spectrum of water in natural abundance in a 10 cm long absorption path at 2 mTorr of pressure showing ¹⁶O, ¹⁷O, ¹⁸O, and HDO transitions. The 100 GHz of frequency space was sampled in 200 kHz steps resulting in approximately half million data points. The 5_{3,3}–5_{2,4} transition is optically thick. The signal-to-noise ratio approaches 10⁵ demonstrating that high sensitivity broadband spectroscopy without any cryogenics is possible above 2 THz.

spectrum shown in Fig. 5 with a 75 kHz HWHM, 40x smaller than the Doppler width, but broadened by the residual 60 Hz. The total integration time was 17 h. Figure 6 shows the Doppler line profile at the same resolution as the Lamb-dip, a blow up of the observed Lamb-dip, and the hyperfine structure of the transition. The measured frequency of the blended F+1 to F components and their crossover dips of this transition is 2640473.813(3) MHz, corresponding to an accuracy of 1 part in 10¹⁰. For comparison, the measured TuFIR frequency is 2640473.836 MHz,⁴⁷ and our Doppler limited frequency is 2640473.830(30) MHz. The limited signal-tonoise ratio and slightly asymmetric line shape limit the linecenter determination of the Lamb-dip. Nevertheless, the accuracy is about ten times better than the Doppler measurement. The present measurement is the highest frequency Lamb-dip ever observed with conventional electronics by a factor of 3. In spite of much more available output power, sub-Doppler spectroscopy remains to be demonstrated with THz quantum cascade lasers due to difficulties with beam quality and frequency control.

D. Spectral coverage

The other aspect of control is to demonstrate the complete useful tuning bandwidth of the source. The spectrum of the simplest stable alcohol, methanol, is an asymmetric top with a large-amplitude internal rotation of the CH $_3$ group relative to the OH group. As such methanol features numerous rotational and torsional transitions throughout the entire terahertz spectral range spanning many orders of magnitude in intensity. A sample pressure of ~ 10 mTorr of methanol was maintained to assure a Doppler limited spectrum through a slow flow and the entire spectrum was recorded. The background power, top trace in Fig. 7, including cell

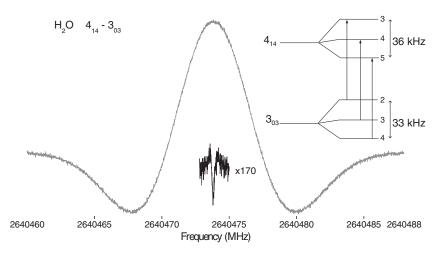


FIG. 6. The Doppler limited absorption profile of the $4_{1,4}$ – $3_{0,3}$ ortho water line recorded at the same 10 kHz per point as the observed Lamb dip shown in the inset. The other inset shows the hyperfine structure of the transition. The half-width and half-height of the dip is \sim 75 kHz and is broadened by some residual 60 Hz in the frequency source. Room temperature heterodyne detection was utilized.

transmission, residual atmospheric water, and detector response, was recorded by stepping the source with amplitude modulation in 20 MHz intervals. The molecular absorption spectrum, bottom trace in Fig. 6, was recorded with a 200 kHz step size with tone burst modulation and a 30 ms lock-in time constant. The signal-to-noise ratio (in 30 ms) exceeds 10 000 in the regions with strong absorption and highest power. The entire spectrum contains 1.35×10^6 data points.

Confirmation of assignments in complex spectra generally requires observation of multiple members a series of transitions. The wide bandwidth techniques, such as FTIR and Terahertz time domain, generally are only sensitive enough to detect the strongest transitions while the available coherent techniques TuFIR and QCL based absorption are limited in their tuning range. Figure 8 show an expanded part of the spectrum in Fig. 7 featuring a Q-branch ($\Delta J = 0$) involving E-symmetry transition between the K = -8 of the second excited torsional state and K = -9 of the first excited torsional state. Overlaid is the Fourier transform spectrum.⁴⁸ The J

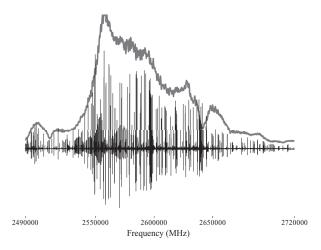


FIG. 7. The top trace is the amplitude modulated power spectrum in arbitrary units through the 2.3 m absorption cell. The bottom trace is the second derivative tone burst modulated spectrum of methanol vapor. The strongest lines have optical depths of $\sim\!\!10$. The usable source bandwidth extends 10 GHz below and 30 GHz above the frequency range shown.

= 28 member of this branch was used in line locking a QCL.⁴⁹ The frequency precision of a line stabilized laser is at best the knowledge of the line frequency but it can be substantially worse due to pressure shifts and the quality of control loop in stabilizing the laser on the peak. Figure 8 shows the $v_t = 2$ -1 (J_K) 20₂–21₃ E-state transition at higher pressure measured with a QCL locked to a free running methanol gas laser to be at 2.519112(1) THz.⁵⁰ The other trace shows our measurement at 2.519109986(50) THz, in excellent agreement with the calculated frequency of 2.519110022(152) THz.⁵¹ Also shown are two other weak transitions one of which can be assigned quantum numbers through extension of the spectroscopic analysis which is ongoing in our laboratory. The lower frequency transition is now assigned to the (J_K^{Parity}) $11_7^{+/-}$ to $12_4^{+/-}$ v_t = 1 A-state line at 2.518914556(100) THz and the weaker, still unassigned feature not detected with the QCL on the high frequency side of the main line, denoted with a U in Fig. 9, is measured to have a frequency of 2.519148974(150) THz. The frequency stability of a free running gas laser is $\sim \pm 1$ MHz unless it is actively stabilized to a known standard. The asymmetry in the QCL line is due to a strong slope in the laser power due to the current ramp used in tuning. Power slopes lead directly to a shift in the measured line center unless accounted for in the line fitting. A complete analysis of the methanol spectrum is in preparation and will be published along with a detailed comparison of laser sideband, TuFIR, QCL, and FTIR techniques.⁵²

III. DISCUSSION

The present demonstration shows that electronics can generate significant signals at room temperature in the 2.48–2.75 THz frequency range. A number of other fix-tuned frequency multipliers with greater than 10% bandwidth between 1 and 2 THz have previously been designed and fabricated.^{53–55} The conversion efficiencies of nonlinear frequency multipliers are limited by the Manley-Rowe relationship.⁵⁶ The output power is, however, currently limited by thermal management at the input frequency rather

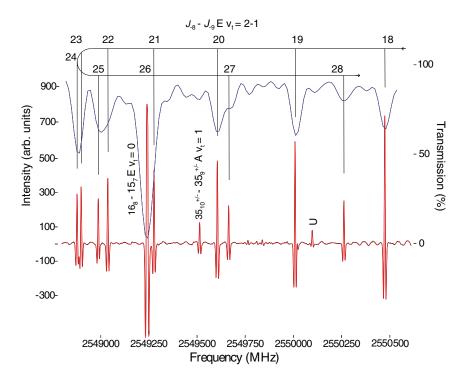


FIG. 8. (Color online) Enlargement of a portion of Fig. 6 showing the turning point of the E-state $K = -8 v_t = 2$ to $K = -9 v_t = 1 Q (\Delta J = 0)$ of methanol near 2.549 THz (bottom trace second derivative) with scale on the left compared with an instrument limited Fourier Transform spectrum in (top trace in absorption) with scale on the right recorded in a 2.7 meter path (Ref. 48). The noise in the bottom trace is about 5 counts peak-to-peak, but the spectrum is oversampled so that it can be smoothed to 1 count without loss of signal. The standing waves are 10–50 counts peak-to-peak. The J = 28 Q-branch line at 2 550 262 MHz was used to stabilize a QCL (Ref. 49).

than parasitic capacitance at the output frequency. Recent demonstration of in-phase power combining of nonlinear multiplier diodes²⁸ and improved heat sinking using CVD diamond⁵⁷ have enabled the construction of frequency multiplier circuits that can effectively handle larger amounts of input power. A confluence of electronic processing technology is pushing the capability of amplifiers into the 1 THz regime³³ and improving power added efficiency at lower

frequencies, 32 enabling higher power sources and potentially shifting the power handling to higher frequencies. The power handling of multipliers drops linearly as a function of frequency since the thermal conduction length varies as f^{-1} while the cross section for conduction drops as f^{-2} . However, the available power declines much faster than linearly making amplification at higher frequencies very desirable. The other new factor is the quality of commercially

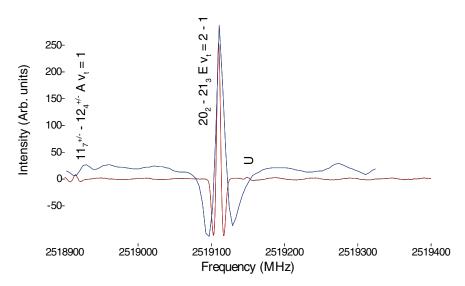


FIG. 9. (Color online) Doppler limited spectrum of methanol recorded near 2.5191 GHz (narrower line) compared to a quantum cascade laser recording (broader line) of the same frequency window scaled to approximately the same peak height. The quantum cascade laser was locked to a free running methanol laser and recorded at higher pressure to measure the pressure broadening (Ref. 50). The asymmetry in the QCL line shape is due to the strong power slope of the QCL. The strong central feature, the $v_t = 2-1\ 202-213$ E state line, is measured to have a frequency of 2.519109985(50) THz. The standing wave between the source and detector is apparent, but is small even in comparison to $v_t = 1$, $11_7^{+/-}$ to $12_4^{+/-}$ A state line at 2.518914556(100) THz on the left-hand edge of the spectrum. The line denoted with a U is measured to have a frequency of 2.519148974(150) THz.

available electromagnetic simulators. These programs not only predict the frequency response of multipliers properly but are beginning to be able to make high fidelity predictions of the conversion efficiency as well. Hence, it is rapidly becoming possible to design systems with the required bandwidth and performance. As a result, there is reason to believe that similar performance to that demonstrated here can soon be extended to \sim 5 THz and that greater than $100~\mu\mathrm{W}$ is possible at frequencies as high as 3 THz.

The present source demonstrates all the characteristics required to make a good local oscillator. It has 270 GHz of tuning bandwidth and its frequency is precisely known through transfer of a 10 MHz frequency standard in the phase locking of the 20 GHz synthesizer. The phase is under control with the usual 20Log(N) penalty for upconversion, as demonstrated by the heterodyne tests. Finally, the linewidth is comparable to our ability to measure it implying that its effect will be negligible in any molecular observation. The beam is as expected for the integral diagonal feed incorporated into the output multiplier. Other than the laboratory spectroscopy demonstrated, the immediate application is in astrophysical spectroscopy where the source and others like it will be used to pump superconducting mixers in the 2-5 THz band. This band contains many key tracers of the interstellar medium and has barely been exploited to date due to the lack of systems having spectral resolution sufficient to study clouds of gas with velocity dispersion of a few km/s ($\lambda/\Delta\lambda > 10^6$). The major limitation has been the lack of spectrally pure, single mode, amplitude and frequency-stable local oscillators, a problem which in the present work has been overcome. The relatively wide tuning range of the system described here not only allows tracking Doppler shifts resulting from differential motions within the Milky Way, but observation of nearby galaxies. These galaxies can be readily studied using the Stratospheric Observatory for Infrared Astronomy (SOFIA), which will fly above almost all terrestrial water vapor and by balloon-borne platforms, which allow longer flights at higher altitudes, extending the access to the spectrum. Broadly tunable, high spectral-resolution heterodyne systems operating on a variety of platforms will give astronomers access to important new probes of interstellar processes including cosmological nucleogenesis, the dynamics of active galactic nuclei, to present-day star formation.

The present technology can also be readily adapted to other kinds of terahertz systems. A compelling application would be as a frequency extender for a vector network analyzer. A full four port measurement could be made with two transmit modules and four receive modules. The latter would require only the first two stages of frequency multiplication (x9) followed by a sub-harmonic (3rd harmonic) mixer. This is possible since the power required to optimally pump a mixer or to achieve efficient multiplication is remarkably similar. The expected dynamic range would be similar to ~100 dB observed in our measurements. The four port measurement and the large dynamic range would be an enormous improvement over existing time domain systems, which generally measure only one port of the four at a time with less than 50 dB dynamic range. A second application of the system could be a source for time domain spectroscopy. In this

case, a chirped pulse would be injected to the source and a second into a single sub-harmonic mixer LO such as the 900 GHz system used in our tests with a suitable delay. The advantage to this method of terahertz generation is that the pulse can be shaped as desired electronically. In this application, the entire bandwidth of the source could be used. In such applications, the power in any particular frequency is low, but the multiple microwatts available represent a large premium over the integrated background power in the band.

The absolute frequency calibration, referenced to a master oscillator, makes this source ideal for controlling the phase and frequency of quantum mechanical sources in the THz gap. As demonstrated by the QCL methanol measurements, all far infrared laser systems require a means to measure and at least stabilize the frequency if they are to be used in any sort of high precision Doppler limited application such as measurement of wind speeds in the atmospheres of planets. As such a technology such as the one described here is going to be necessary for production of the reference signal for locking or stabilizing any far infrared laser. The most interesting future application would be to seed a terahertz laser based amplifier, which would be able to produce significantly more power than the currently available.

IV. CONCLUSIONS

The capability of conventional solid state electronics continues to extend into the THz gap as demonstrated by the frequency source covering 270 GHz of bandwidth between 2.48 and 2.75 THz described here. This source features unprecedented simplicity in the control of the signal by allowing conventional microwave signal generation to be upconverted. As such, it is possible to utilize the source in continuous wave, modulated or pulsed mode. When modulated the effective bandwidth is the entire available 270 GHz. This means that the source has the simplicity of an RF amplifier allowing for enormous flexibility in the signals generated. The output power varies between -27dBm and -18dBm over the 2.5-2.7 THz band with no adjustment of bias or tuning mechanisms. The system can be adapted for network analysis, time domain studies, or coherent transmitters used in laboratory spectroscopy or as local oscillators in remote sensing. It also represents a simple solution for providing reference signals for control of terahertz quantum mechanical systems. Currently available electromagnetic simulation capabilities allow for accurate design of frequency response and accurate prediction of power levels. This capability coupled with rapidly advancing electronics processing technology suggests that electronics can now be designed to fill the THz gap. As demonstrated by the present work, the source has already opened an enormous region of the THz gap to high precision spectroscopy for laboratory, astrophysical, planetary, and Earth atmospheric applications that were previously nearly impossible.

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