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Terahertz Spectral Signatures: Measurement and Detection LDRD Project 86361 Final Report

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Terahertz Spectral Signatures: Measurement and Detection
LDRD Project 86361 Final Report

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Abstract

LDRD Project 86361 provided support to upgrade the chemical and material spectral signature measurement and detection capabilities of Sandia National Laboratories using the terahertz (THz) portion of the electromagnetic spectrum, which includes frequencies between 0.1 to 10 THz. Under this project, a THz time-domain spectrometer was completed. This instrument measures sample absorption spectra coherently, obtaining both magnitude and phase of the absorption signal, and has shown an operating signal-to-noise ratio of 10^4. Additionally, various gas cells and a reflectometer were added to an existing high-resolution THz Fourier transform spectrometer, which greatly extend the functionality of this spectrometer. Finally, preliminary efforts to design an integrated THz transceiver based on a quantum cascade laser were begun.

I. Introduction

One of the major reasons to use electromagnetic radiation at frequencies around 1 THz *(i.e.*, 10^{12} cycles per sec) is that many molecules and solids have THz spectral signatures that are far stronger and more distinctive than in either lower frequency radio-to-microwave or higher frequency infrared-to-visible. Spectral specificity arises from the fact that THz radiation generally couples to a molecule's moment-of-inertia or a solid's lattice vibrations rather than to mass alone, and hence distinguishes mass distribution and coupling forces. Advanced work on both THz spectroscopy and devices to detect distinctive THz spectral features is clearly important to building a useful THz technology base. Results of this project will be very attractive to potential follow-on customers who have specific detection and discrimination requirements. In addition, signature knowledge ties together the ongoing solid-state THz source and detector projects in many conceivable end-use system applications. For example, in high-performance sensing applications, a narrow-band, quantitatively known spectral signature defines the frequency and sensitivity performance requirements around which a tuned, matched source/detector transceiver subsystem must be designed and built.

This report covers three tasks. The first is the design, construction, and testing of a THz time-domain spectrometer that has very high dynamic range and signal-to-noise ratio and is flexible enough to use for a variety of potential customer-related needs. The second is an upgrade to the capabilities of an existing high resolution THz Fourier transform spectrometer via the addition of gas cells and a reflectometry set-up. Finally, the preliminary design of an all solid-state THz transceiver detector and the development of a quantum cascade laser local oscillator for this transceiver will be discussed.

II. THz Time-Domain Spectrometer

The goal of this task was to build a THz time-domain spectrometer (THz-TDS) versatile enough to be used to meet anticipated customer needs for signature development. These systems usually employ two types of detection: photoconductive antennas or free-space electro-optic sampling. A detailed comparison of the two techniques can be found in the literature [i], but to summarize, signal strength is higher and spectral range is lower in photoconductive antenna-based systems compared to free

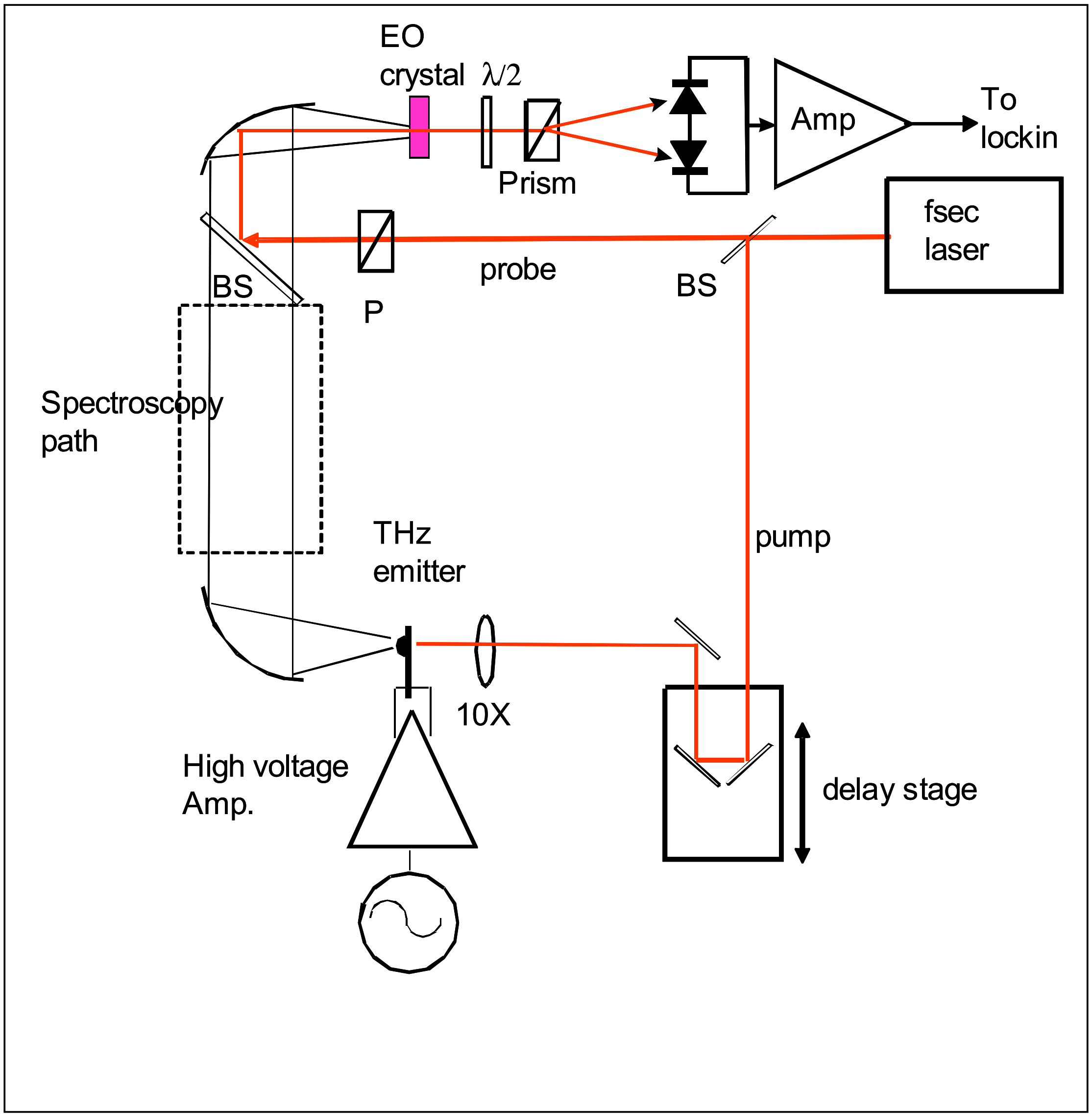


Figure 1. Block diagram of the THz-TDS system

space electro-optic sampling setups. Due to the limited availability of photoconductive antennas, we built our system based upon electro-optic sampling.

A schematic diagram of the system is shown in Fig. 1. A femtosecond laser beam (hundreds of milliwatts, ~150 fs) is split into two beams. The pump beam passes through a computer controlled delay stage and then excites a THz emitter (to be described below) through a 10X microscope objective. A short burst of THz radiation is emitted through the back of the THz emitter, and is collected using a metallic paraboloid mirror. This radiation is then combined with the weaker probe beam (after its polarization was adjusted with a polarizer P). Then both are focused onto a 1mm-thick ZnTe crystal. This electrooptic crystal performs the electrooptic detection of the THz field: when both the THz and laser probe beam overlap temporally and spatially in the crystal, the probe beam experiences a small polarization rotation that is analyzed using the subsequent waveplate, Wollaston prism and balanced photodiode pair. The photodiode balanced pair consists of an identical pair of photodiodes, and a homemade, low noise transimpedance amplifier, operating at a gain of 10^5 V/A. We performed extensive characterization of the noise performance of several amplifier designs as well as photodiode suppliers, and we can routinely achieve noise floors within a factor of 2 of the laser shot noise.

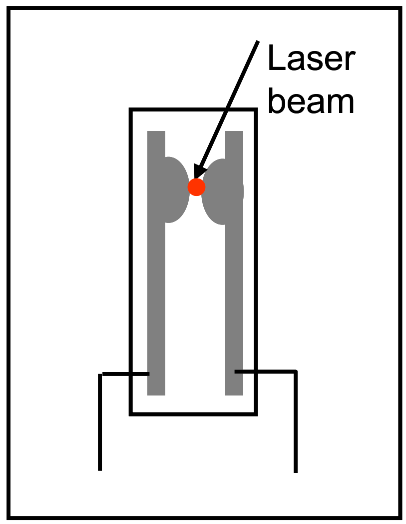


Figure 2. Schematic of THz picosecond pulse emitter. The femtosecond laser beam strikes where indicated.

The THz emitter is based on large photoconductive gaps [ii]. Two silver paint electrodes were defined on a semi-insulating GaAs substrate. A smaller gap (~200um) was defined near the edge using silver epoxy. Finally, an aplanatic Si-lens was glued to the back of the emitter in order to enhance the THz signal collection. A schematic diagram of this emitter is shown in Fig. 2. Using a high voltage amplifier and a waveform generator, we can drive this photoconductive switch with voltages of up to ± 200 V. This same waveform is used as the reference to the lockin amplifier. This has the added advantage of having no mechanical choppers in the system, and also the ability to increase the lockin frequency well above the 1/*f* noise of the laser system. We choose a modulation frequency of ~18 kHz.

An example of a typical waveform obtained using this system is shown in Fig. 3. Currently the system has no nitrogen purged box and thus water absorption significantly

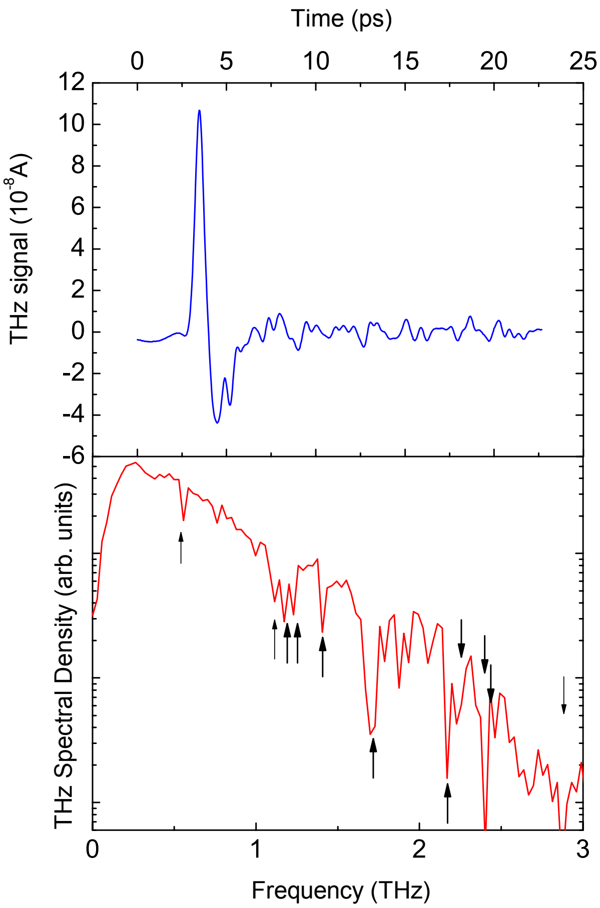


Figure 3. A typical time-domain signal (top) and its associated frequency spectrum (bottom) from the THz-TDS.

reduces the bandwidth that can be obtained. The arrows denote mostly water absorption lines, and some features that can be attributed to the emitter geometry. Another factor contributing to a reduced bandwidth is the lack of pulse shaping optics: the laser system we currently employ produces pulses that are closer to 200 fs, and we have quite a few dispersive elements along the beam path. We plan to precede the system with a pulse compressor which in principle could decrease the pulse width to ~80 fs.

One result not immediately obvious from Fig. 3 is that with no special effort, we can routinely obtain signal to noise ratios (SNR) near 10^4. The trace in Fig. 3 has SNR of 5000. Conventional free-space electrooptic sampling systems usually show SNRs of a few hundred. The driving voltage to the THz emitter was ~100 V, so there is still a lot of room for improvement. The dotted area between the two paraboloid mirrors can be used for regular spectroscopy using either a large opening gas cell, or smaller samples by focusing the THz beam with Teflon optics or paraboloid mirrors.

III. Upgrade of THz Fourier Transform Spectrometer

The capabilities of an existing Fourier transform spectrometer (FTS) specially designed to operate at THz frequencies and with very high spectral resolution (i.e., 0.002 cm^{-1} or 60 MHz instrumental linewidth) were upgraded with the addition of a few extra gas cells and a reflectometry set-up.

The gas cells consist of quartz cylindrical tubes approximately 50 mm in diameter and from 10 to 20 cm in length. Both ends of the tubes have polyethylene windows sealed vacuum tight on the quartz using Viton O-rings. Polyethylene is used because it has a relatively high (~80%) and flat radiation transmission characteristic across the THz spectrum. A gas inlet/outlet nipple with vacuum tight valve is integrated into the body of each cell. While a single gas cell can be evacuated and refilled with different chemical species, multiple gas cells are necessary to avoid cross-contamination among incompatible chemical vapors, for example acid solvent vapors.

An example absorption spectrum taken with THz FTS using one of the new gas cells is shown in Fig. 4. The vapor is carbon monoxide (CO), a molecule relevant to pollution monitoring. The CO in the gas cell is at an approximate pressure of 10 mbar, so that pressure broadening is very small and the linewidths of the measured absorption

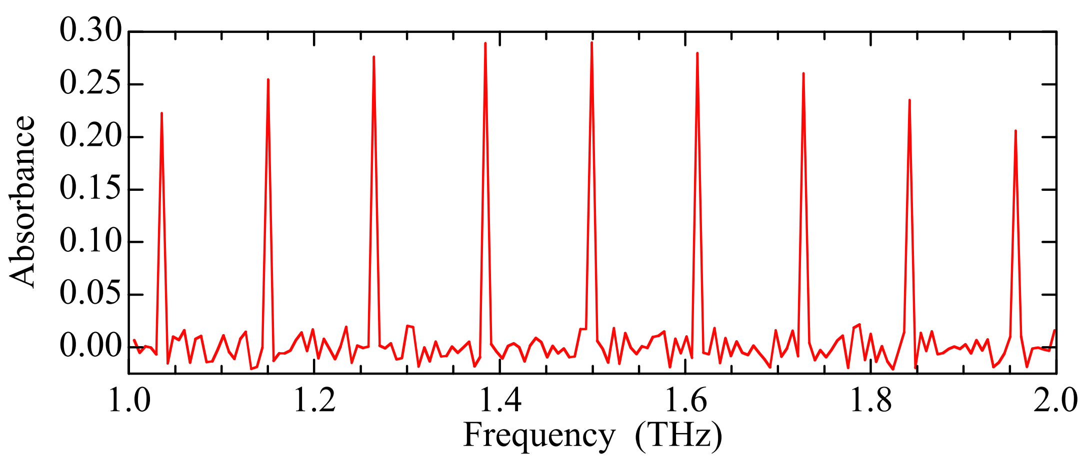


Figure 4. THz absorption spectrum of CO vapor, approximately 10 mbar pressure.

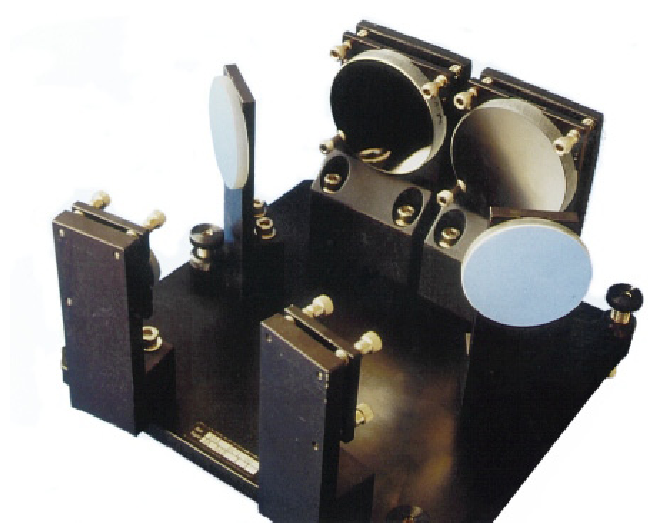


Figure 5. Photograph of THz FTS reflectometry set-up.

resonances are very narrow, requiring the very high spectral resolution of this FTS. This spectrum is normalized to the same gas cell fully evacuated.

For measuring THz signatures on solids, reflection rather than transmission is the preferred method on highly reflective and highly absorptive samples. For this purpose we developed and installed a specular reflectometry set-up into the THz FTS. This set-up focuses the spectrometer light onto a sample at a 10° angle of incidence (to the sample face normal) and collects the specular reflection. A front-surface polished gold mirror serves as a reference reflector. Fig. 5 shows a picture of the reflectometer.

An example of a reflectance spectrum taken using this reflectometer is shown in Fig. 6. The sample is a single crystal of $SrTiO_3$, and the reflection is from the polished (001)

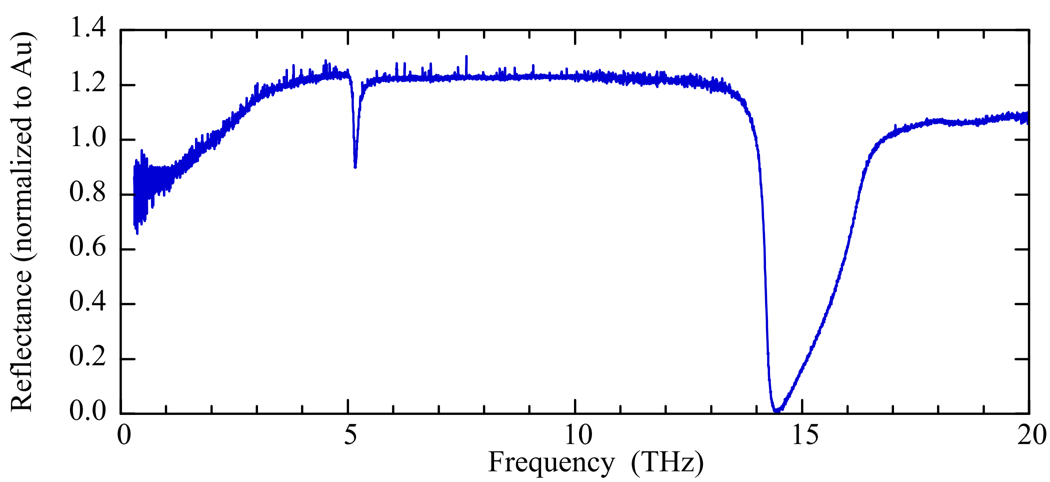


Figure 6. Reflectance spectrum (normalized to a Au mirror) of $SrTiO_3$.

crystallographic face. Clearly evident are two prominent lattice vibration absorption signatures at about 5.1 and 14.5 THz that identify this material as $SrTiO_3$. Also seen at lower frequency is a severely over-damped lattice vibration that reduces the low-frequency reflectance of the material. Around 10 THz the reflectance of this crystal exceeds the THz reflectance of the Au reference mirror. This spectrum is quantitatively consistent with sparser reflection spectra on high-quality crystals reported in the literature,[iii] but has a far higher density of points.

IV. THz Transceiver Design and Quantum Cascade Laser Local Oscillator

Preliminary work on the design of an all solid-state integrated THz transceiver capable of detecting a selected spectral signature with high sensitivity and high discrimination was carried out. An innovative idea for integrating a solid-state THz quantum cascade laser (QCL) local oscillator with a THz mixer to create a heterodyne transceiver was generated.

The ideal situation is to integrate onto a single semiconductor chip platform a THz QCL and a single or a linear array of THz mixers such as Schottky diodes. The QCL supplies the local oscillator (LO) source that pumps the mixers and also, if needed, a coherent active illumination source. Should the QCL be used as an active illumination source, the mixer can phase lock onto the LO, yielding extremely high rejection of background interference.

The innovative but risky part of the idea developed is the integration of QCL and mixer is illustrated in Fig. 7. Rather than the conventional procedure of shining the QCL light onto a mixer from the QCL's end-facet output either through free-space or an external waveguide, the mixers are actually either monolithically fabricated or flip-chip bonded into the semiconductor ridge cavity that forms the QCL. The mixers are recessed slightly into the ridge so that they are directly exposed to a portion of the internal propagating THz field inside the QCL cavity. Because this internal THz field has at least ten times more power than the light that can be output through a facet, this method of delivering LO power to the mixers has tremendous advantages in power efficiency, compactness, simplicity, and flexibility of design over more conventional methods of

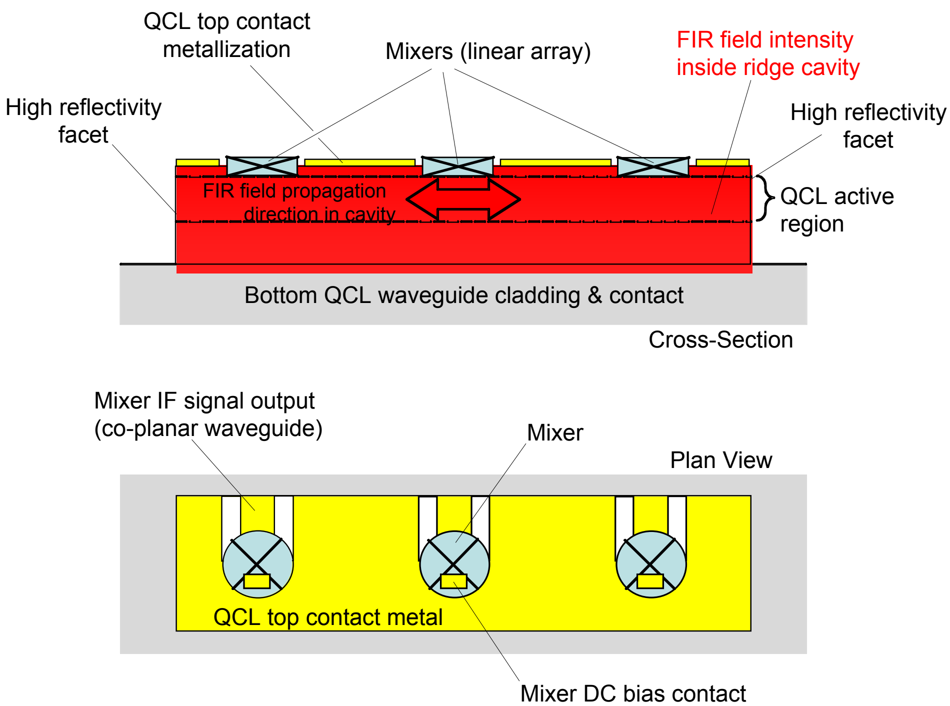
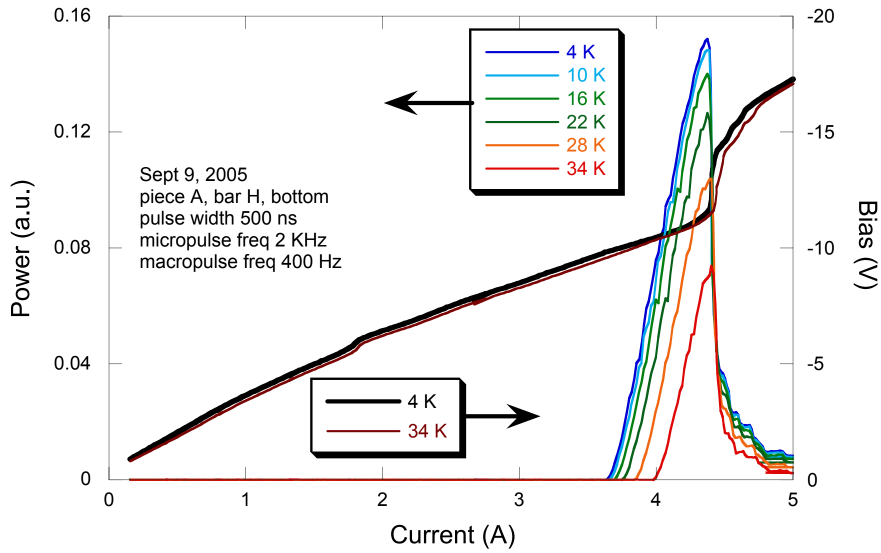


Figure 7. Illustration of the QCL/mixer integrated transceiver idea (not to scale). A mixer or array of mixers is fabricated or flip-chipped into the propagating THz field inside the QCL ridge cavity, so that the internal THz field supplies local oscillator power to the mixer(s).

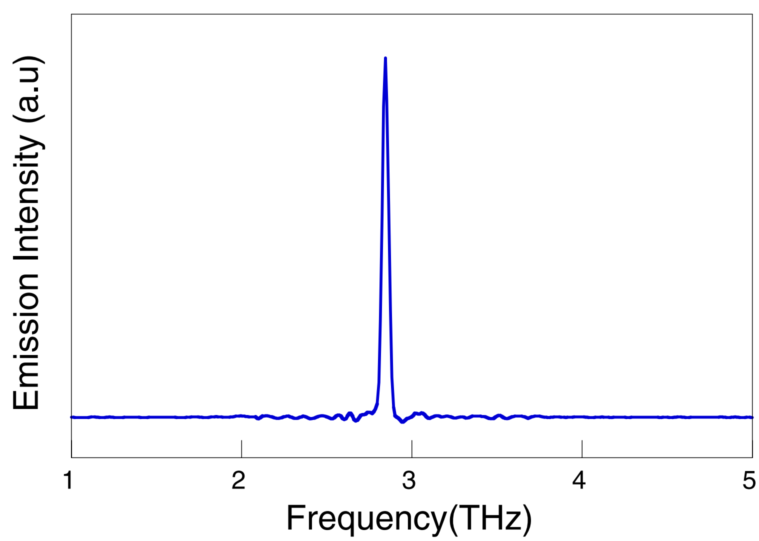
supplying LO power to a mixer. More extensive electromagnetic modeling of this configuration is being pursued to determine the viability of the idea.

It is also important to note that such a heterodyne transceiver will also enable a new THz spectroscopic tool with unprecedented ultra-high spectral resolution. The transceiver can continuously monitor a frequency band equal to the larger of the QCL tuning range or mixer IF bandwidth (typically up to 10 GHz) around a central QCL frequency. The spectral resolution is limited by the QCL linewidth, which can be as narrow as *30 kHz*. Such ultra-high resolution spectroscopy is especially important in resolving signature THz resonances of large biological molecules, which often have a series of resonances spaced by only a few MHz or less.

The first concrete step towards the development of a THz signature detector component has been made through the successful design, fabrication, and testing of a QCL. This QCL emits at 2.8 THz. Its electrical/power and spectral emission characteristics are shown in Fig. 8. More in-depth measurements of its properties are currently being pursued.



(a)



(b)

Figure 8. (a) Emitted power (uncalibrated) and electrical characteristics of a 2.8 THz QCL at various operating temperatures. Lasing occurs at between 3.5 and 4 A DC current bias. (b) Emission spectrum of the QCL in lasing mode. Linewidth of the emission is limited by the spectrometer, not by the laser itself.

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