Cryogen-Free Solution for Terahertz Absorption Spectroscopy

ABSTRACT

Researchers from Germany have developed a high-speed pyroelectric receiver using a $LiTaO_3$ pyroelectric detector as a cryogen-free solution for terahertz (THz) absorption spectroscopy measurements. This receiver can record spectra at frequencies up to 281 Hz without introducing artifacts into the observed spectral absorption profile. By operating at room temperature, the compact system provides a practical and efficient alternative to cryogenically cooled detectors, making it more accessible for various applications. The high sensitivity and stability of the receiver in capturing THz absorption spectra show its potential as a reliable platform for THz spectroscopy, offering robust performance without the need for complex cooling infrastructure.

CRYOGENIC COOLING

Cryogenic cooling has long been the standard in high-sensitivity detection systems, particularly in applications requiring the measurement of low-energy signals, such as terahertz (THz) spectroscopy. By cooling detectors to extremely low temperatures, often using liquid helium or nitrogen, thermal noise can be minimized, leading to improved signal-to-noise ratios and enhanced detection sensitivity. These cryogenically cooled systems have been widely used in fields like chemistry, physics, medical imaging, and material analysis, where accurate detection of weak electromagnetic signals is crucial.

However, the reliance on cryogenic cooling presents significant challenges. Cryogenic systems are complex, expensive, and require continuous maintenance, including replenishing cooling agents. The use of liquid helium, in particular, has become more problematic due to increasing scarcity and cost. Additionally, these systems are bulky and limit the portability of devices, making them less practical for widespread use, especially in industrial or field-based applications. As a result, there is a growing demand for cryogen-free detectors that can operate at room temperature while still providing the high sensitivity required for precise measurements.

Cryogen-free detectors offer a more practical, compact, and cost-effective solution, eliminating the need for cumbersome cooling systems. Developing such detectors, including those based on pyroelectric materials, is critical for high-resolution THz spectroscopy in astronomy, atmospheric research, and plasma diagnostics.[1] These detectors can enable more accessible and flexible setups, allowing for wider adoption across different sectors and applications without sacrificing performance.

PROBLEMS AND GOALS

Although cryogenic detectors are highly effective for THz spectroscopy, their drawbacks highlight the need for more practical, cost-efficient solutions. The limited bandwidth of cryogenically cooled devices, such as bolometers, imposes significant constraints on measuring THz absorption spectra, as it hampers their ability to effectively capture fast or broadband signals. Similarly, early pyroelectric detector designs are hindered by their low speed and relatively poor sensitivity, making them less suitable for high-precision applications. While cryogenically cooled detectors have traditionally provided high sensitivity to reduce thermal noise, these approaches involve complex and costly setups, requiring careful maintenance and specialized environments.

Researchers have explored cryogen-free detector alternatives that can operate at room temperature while maintaining the sensitivity required for precise THz measurements. Many of these developments still face challenges related to sensitivity, speed, or noise performance, making them less suitable for high-resolution applications in THz spectroscopy. While cryogen-free designs are explored further, rigorous testing is needed to ensure these devices are comparable to the accuracy and sensitivity of standard cryogenically cooled detectors. A highly sensitive detection with high bandwidth, low cost, compactness, and the ability to record spectra at high frequencies without deformations is essential in the photonics world for THz spectroscopy applications. This study addresses these challenges by developing a cryogen-free pyroelectric receiver capable of high-speed detection without sacrificing sensitivity.

METHOD

Researchers from Germany have developed a high-speed $LiTaO_3$ pyroelectric receiver used as a cryogen-free detector for THz absorption spectroscopy measurements. The experimental setup consists of the $LiTaO_3$ pyroelectric detector and amplifier electronics, as shown in **Figure 1**. The pyroelectric receiver is designed to operate at room temperature, eliminating the need for complex cryogenic cooling setups. The goal of this design is to increase speed and detectivity for undistorted measurements of infrared signals down to 0.5 THz.[1]

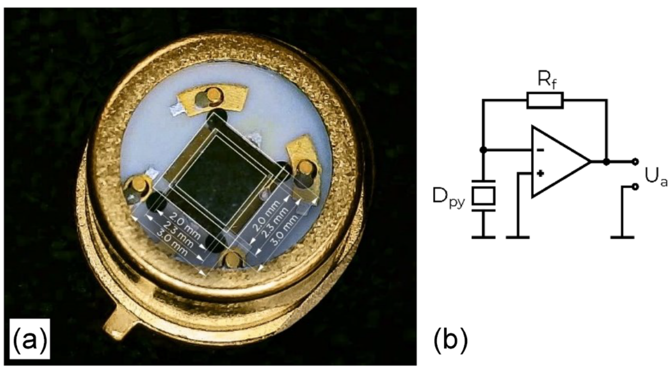

Figure 1. (a) A photograph (30x magnification) of the $LiTaO_3$ chip mounted on a header. A thin membrane (2.3 x 2.3 mm², 6 µm thickness) is etched into a frame (3.0 x 3.0 mm², 20 µm thickness) and then coated (2.0 x 2.0 mm²). Vibration dampening is achieved by 4 flexible posts. All wire bonds have been made on the frame. The element is sealed by a TO-can assembly with an HDPE window (0.8 mm thickness). (b) A simplified schematic circuit of the current mode configuration.[1]

The entire system for this experiment is enclosed in a vacuum-sealed chamber to prevent thermal fluctuations from the environment, ensuring that the detected signal originates solely from the THz source. No cooling is required, and all measurements were performed at room temperature. The detector comprises a LiTaO3 pyroelectric element, which uses a proprietary current mode configuration for radiation detection. As the pyroelectric detector absorbs radiation, the increased temperature leads to a charge separation at the detector interfaces which generates a current that is amplified and converted to a voltage by the amplifier electronics. The current mode allows for a much higher gain at low noise levels while still offering an electronic bandwidth of 8 kHz. Traditional voltage mode detectors typically only offer 10 - 100 Hz bandwidth which limits the measurement capabilities.

Not only does the newly developed receiver have a higher bandwidth than traditional pyroelectric detectors in voltage mode, but it also achieves high internal amplification and therefore high responsivity of up to 70 kV/W at low noise density (25 µV/√Hz). This results in a high detectivity of up to 4 x 10^8 cm √Hz/W. With this design, signals can be measured without distortion and with a linearity of 1% or better over four orders of magnitude in power.[1]

The THz radiation is generated by a tunable, continuous-wave THz quantum cascade laser (QCL) with an output of ~4 mW. The output of the QCL was tuned by linearly ramping the input current using Wavelength Electronics' QCL1000 OEM driver. Continuous ramping with a sawtooth waveform enabled fast measurements with high spectral resolution. The spectral resolution was given by the laser linewidth which was ~6 MHz. Because the detector has a larger bandwidth than traditional cryogen-cooled detectors, the laser tuning frequency can be increased without causing deformation to the absorption features. The tuning frequency of the QCL was varied from 11 Hz to 4 kHz for the experiment.

To minimize atmospheric water vapor absorption, the majority of the laser beam path was purged with nitrogen gas, as shown in **Figure 2**. A parabolic mirror can be removed so that the laser beam is incident on the bolometer instead of the pyroelectric detector. This allows the bolometer to be used to compare the results to validate the pyroelectric receiver. This figure also shows the size difference and compact design of the pyroelectric receiver.

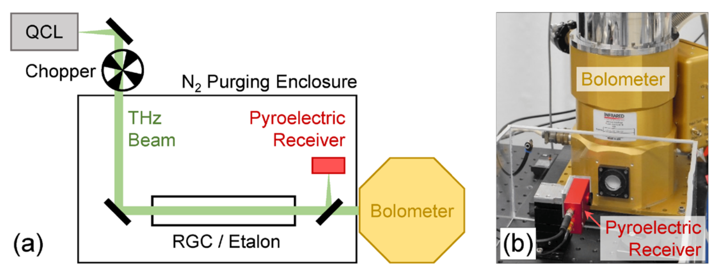

Figure 2. (a) A schematic overview (not to scale) of the THz beam path from QCL to detector (green) with either the pyroelectric receiver (red) or the bolometer (yellow) as detector, and (b) a photograph showing both detectors.[1]

To achieve high-speed performance, the researchers optimized the thermal properties of the $LiTaO_3$ element and the electronic readout system. This allowed the receiver to record THz spectra at rates up to 281 Hz without introducing artifacts into the observed spectral absorption profile. This rate could be increased with upgrades to the design, particularly the chopper. The system's response time and frequency range were carefully calibrated to ensure the accurate detection of rapid changes in the absorption characteristics of the tested materials.

After the detector was characterized and calibrated with a laser diode, THz absorption spectroscopy measurements were performed to test and validate the capabilities of the cryogen-free detector. All results were performed with a bolometer as a separate THz detector to compare results with known values. The THz absorption spectroscopy was performed on a reference gas cell (RGC) filled with ammonia (NH_3). The RGC has a length of 15 cm and was filled with pure NH_3 to a precisely known pressure of 1.61 ± 0.01 mbar.[1] Measurements were made with laser tuning frequencies of 11 Hz and 201 Hz with a tuning range of 480 mA to 600 mA.

RESULTS

The raw results of the experiments are presented in **Figure 3**. Two absorption features can be observed which are identified as the R(7,5)-transition of $^{15}NH_3$ at approximately 580 mA and the aR(7,2)-transition of $^{14}NH_3$ at approximately 530 mA. The more prominent absorption feature at ~580 mA can be better seen in **Figure 3**b with a more narrow tuning range. Both absorption features were recorded without significant deformations even for the high tuning frequency of 201 Hz. In comparison, the same absorption features measured with a bolometer were asymmetrically deformed for a tuning frequency of only 10 Hz.[1] Deformation limits with the pyroelectric receiver were only seen when tuning frequencies were above the maximum speed of the chopper. Changing to a faster chopper would enable high-frequency analysis.

The major difference between the 11 Hz and 201 Hz frequencies is the baseline or the line corresponding to zero intensity detected. A flat horizontal line is approximated for the 201 Hz frequency while a sloping line is approximated for the 11 Hz frequency. If the baselines were subtracted from the curves, the shapes would, theoretically, be the same. Because correctly determining the baseline is critical for accurately calculating the ammonia density and pressure in the cell, the pyroelectric receiver is best suited for absorption measurements with laser tuning frequencies of 201 Hz or higher.

From the raw spectrum in **Figure 3**, the absorbance can be calculated using the measured intensity concerning the baseline and the intensity that would have been measured without absorption. The resulting absorption spectrum is shown in **Figure 4** where it has no pronounced W shape or distortion when compared to measurements with a bolometer.[1]

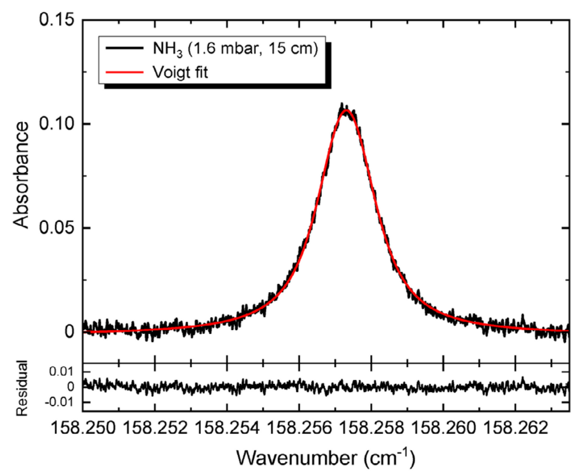

Figure 4. Spectral absorption profile of the R(7,5)-transition of $^{15}NH_3$ at 158.257314 cm^{-1}, measured in an RGC with NH_3 (pressure of 1.6 mbar, length of 15 cm) using a laser tuning frequency of 201 Hz and a tuning range of 564 mA to 590 mA. The measured data are averaged over 2000 single measurements. A Voigt fit was fitted to the profile; the corresponding residual is given in the bottom part of the figure.[1]

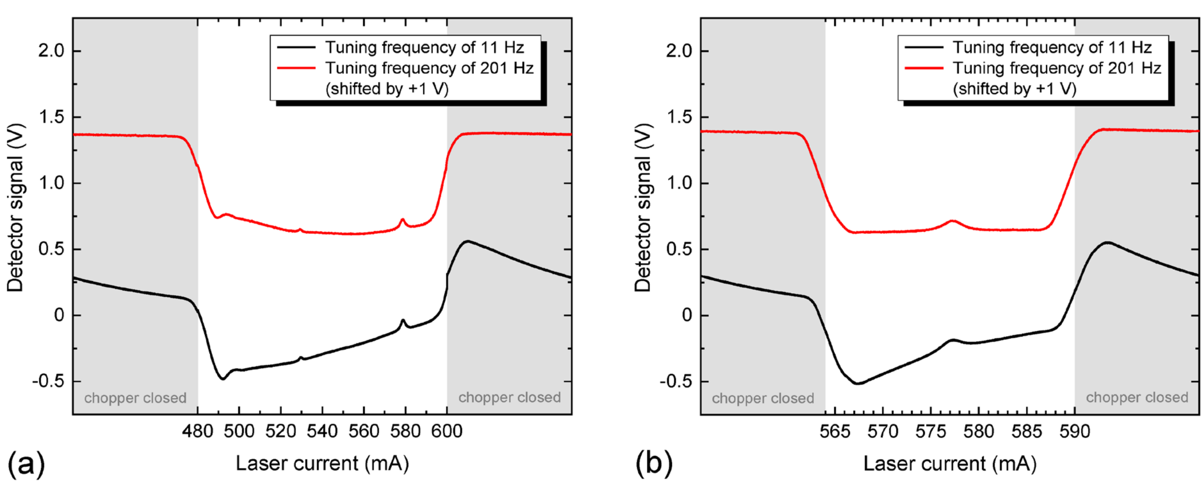

Figure 3. Measurements on an RGC with NH_3 (pressure of 1.6 mbar, length of 15 cm) for different laser tuning frequencies using (a) a tuning range of 480 mA to 600 mA and (b) a tuning range of 564 mA to 590 mA. The measured data are averaged over 2000 single measurements. The data for 201 Hz are vertically shifted by +1 V for a better comparison.[1]

According to the Lambert-Beer law, the area under the absorption profile is directly proportional to the density of ammonia measured which can be related to the pressure inside the cell. Using this, the calculated pressure inside the gas cell resulted in 1.65 ± 0.10 mbar which is in excellent agreement with both the analysis with a broader tuning range and with the precisely known pressure of 1.61 ± 0.01 mbar.[1]

Unlike cryogen-free spectrometers, the pyroelectric detector enables fast data acquisition and high sensitivity with its high bandwidth for avoiding deformations of the absorption features.[1] To further demonstrate this design as a reliable detector for absorption spectroscopy measurements in the THz range, the noise level of the pyroelectric receiver was compared to that of the bolometer. **Figure 5** shows the standard deviation of the noise of the absorption spectra as a function of total measurement time.[1]

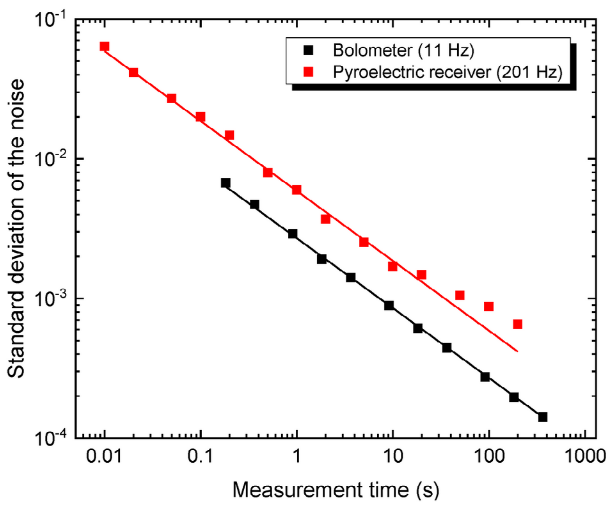

Figure 5. Standard deviation of the noise of absorption spectra recorded with the bolometer (at a tuning frequency of 11 Hz) and the pyroelectric receiver (at a tuning frequency of 201 Hz) as a function of the total measurement time.[1]

The results for both the bolometer and pyroelectric receiver lie on a line with a slope of -0.5, as is expected for shot-noise limited detection. This implies that white noise is the dominating noise contribution for measurement times below 10 seconds.[1] For single measurements with the pyroelectric receiver, the SNR is worse by approximately a factor of 10 compared to the SNR of single measurements performed by the bolometer. However, the SNRs of absorption spectra recorded with the same measurement time differ by only a factor of 2. Purging can improve the SNR by a factor of 3 to 4 depending on experiment humidity levels. Based on the noise measured, the resulting detection limits are 4×10^{14} cm^{-3} for the pyroelectric receiver, which is only a factor of 2 higher than the bolometer.

This research demonstrates the successful implementation of a high-speed, cryogen-free pyroelectric receiver for terahertz absorption spectroscopy, offering a compact and efficient alternative to traditional helium-cooled detectors. The experimental setup enabled high-resolution spectral measurements with minimal noise and artifact-free absorption profiles, achieving data acquisition rates up to 281 Hz and frequencies up to 1 kHz are possible with a design upgrade. The results were in excellent agreement with known values obtained through other established methods, validating the reliability and accuracy of this innovative approach. By eliminating the need for cryogenic cooling, this design simplifies experimental setups and represents a significant step forward in advancing practical terahertz spectroscopy systems in a broad range of applications.

WAVELENGTH'S ROLE

To optimize the performance and testing of the high-speed $LiTaO_3$ pyroelectric receiver, researchers integrated the innovative capabilities of Wavelength Electronics' QCL1000 OEM driver. This essential component is designed to provide precise control over QCL drive currents, which is critical for ensuring optimal sensitivity and accuracy in THz absorption spectroscopy measurements.

Given the significant impact of temperature and noise on the pyroelectric receiver's performance, ultra-low noise current drivers were required. Wavelength Electronics' QCL1000 OEM driver operates with a low noise floor, maintaining an output current RMS noise of less than 0.7 μA at operating frequencies up to 100 kHz. This level of noise reduction is vital for accurately capturing the rapid spectral features without introducing artifacts, thereby ensuring a clean spectral absorption profile.

The driver was specifically utilized to drive the THz quantum cascade laser that emits the THz radiation necessary for the absorption spectroscopy measurements. It offers a maximum output of 1 A with a compliance voltage of 16 V, allowing researchers to finely tune the applied current to the QCL and subsequently to the pyroelectric receiver. Continuous ramping of the laser current was required for fast measurements with high spectral resolution. Because the spectral resolution of the system is given by the laser linewidth, the laser drive must maintain a narrow linewidth from the QCL. The QCL1000 OEM driver achieved this with a narrow linewidth of 2×10^{-4} cm^{-1}, approximately 6 MHz.

With a typical stability of around 10 ppm, the QCL1000 OEM driver ensures optimal operating conditions for both the THz QCL and the pyroelectric receiver, significantly enhancing measurement capabilities. This integration has proven instrumental in achieving high-speed data acquisition rates of up to 281 Hz without compromising the integrity of the recorded spectra. By utilizing the advanced features of the QCL1000 OEM driver to drive the THz QCL, the study of THz absorption phenomena has become more robust, paving the way for innovative applications in spectroscopy and materials analysis.

PRODUCTS USED
QCL1000 OEM

KEYWORDS
Terahertz, spectroscopy, pyroelectric receiver, detector, quantum cascade laser, QCL, THz, QCL1000 OEM, room-temperature, helium-cooled, high-resolution, laser driver, miniaturized detectors, ammonia

REFERENCES

1. Wubs, J.R., Macherius, U., Lu, X., et al. Performance of a High-Speed Pyroelectric Receiver as Cryogen-Free Detector for Terahertz Absorption Spectroscopy Measurements. App Sci 14, 3967 (2024). **https://doi.org/10.3390/app14103967**

USEFUL LINKS

- QCL1000 OEM **Product Page**

PERMISSIONS

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