Tunable resolution terahertz dual frequency comb spectrometer

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Abstract: Terahertz dual frequency comb spectroscopy (THz-DFCS) yields high spectral resolution without compromising bandwidth. Nonetheless, the resolution of THz-DFCS is usually limited by the laser repetition rate, which is typically between 80 MHz and 1 GHz. In this paper, we demonstrate a new method to achieve sub-repetition rate resolution in THz-DFCS by adaptively modifying the effective laser repetition rate using integrated Mach-Zehnder electro-optic modulators (MZ-EOMs). Our results demonstrate that it is possible to improve the 100 MHz resolution of a terahertz frequency comb by at least 20x (down to 5 MHz) across the terahertz spectrum without compromising the average output power, and to a large extent, its bandwidth. Our approach can augment a wide range of existing THz-DFCS systems to provide a significant and easily adaptable resolution improvement.

References and links

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1. Introduction

Terahertz generation and spectroscopy, employing technologically mature semiconductor emitters/detectors with sub-picosecond carrier lifetimes, have grown dramatically with applications in a variety of fields – ranging from biochemistry to material science [1,2]. Photoconductive antennas (PCAs) based on such materials combined with femtosecond lasers have enabled the development of terahertz time domain spectroscopy (THz-TDS) [3], which offers several advantages in comparison with Fourier Transform far infrared spectroscopy (FTIR), particularly in terms of signal-to-noise ratio (SNR) and resolution. However, conventional THz-TDS still requires moving parts such as a mechanical delay stage. To overcome this issue, THz-TDS spectrometer designs now employ an asynchronous optical sampling (ASOPS) technique and have been replacing conventional systems over the past decade [4,5]. ASOPS instruments use two mode-locked lasers with a fixed repetition rate difference. An extension of ASOPS is terahertz dual frequency comb spectroscopy (THz-DFCS), wherein a train of pulses is acquired and Fourier transformed to yield a comb mode resolved spectrum [6]. Related dual comb spectroscopy in the NIR and MIR regions has shown that such systems present a unique ability to combine high resolution and broad spectral bandwidth, thus eliminating the typical tradeoff between these parameters [7–9]. However, unique to terahertz combs generated in this manner is the absence of the carrier-to-envelope offset frequency (f_{ceo}) [6,10], a fact associated with single cycle pulses and their production by optical rectification. Thus, the frequency of each terahertz comb mode is simply an integer harmonic of the original laser repetition rate (f_{rep}), meaning that spectroscopy can be performed with frequency accuracy limited only by the microwave reference clock controlling f_{rep}.

Although dual comb spectroscopy has proven advantageous relative to conventional THz-TDS, its spectral resolving power is still limited by the laser repetition rate. More recently, the so-called spectrally interleaved terahertz dual comb spectroscopy has been developed to overcome this limitation [11,12]. This technique is based on scanning the repetition rates of both lasers while maintaining a constant difference between them. After a series of acquisitions, the waveforms can be overlapped to yield a gapless comb-resolved spectrum in the frequency domain.

In this work, we demonstrate a new approach to increase the terahertz comb spectral resolution by use of electro-optic modulators external to the laser cavities. The basic idea is to drive the modulators with a train of electrical pulses derived from the lasers themselves, in order to implement a pulse-picking procedure [13,14] that effectively reduces the laser repetition rate in a manner that can be adaptively controlled. The increase in resolution associated with reducing the comb tooth spacing is particularly advantageous for high resolution gas phase spectroscopy, especially at reduced pressures. While the atmospheric pressure broadened linewidths of a few gigahertz are well sampled by a 100 MHz comb in the terahertz region, the widths decrease rapidly at lower pressures (< 0.1 kPa) to the Doppler limit of few megahertz at room temperature. Furthermore, the peak absorbance at fixed mole

fraction in air often remains nearly constant as the total pressure is reduced to the Doppler limit thereby dramatically improving the selectivity and sensitivity for multispecies detection.

2. Experimental setup

Figure 1 illustrates the concept of the pulse-picking process. After being incident on a photoconductive emitter, the unmodulated train of pulses from a femtosecond laser (top panel) yields a comb of frequencies extending from the RF to the THz region that are exact integer multiples of f_{rep}. The pulse-picking is achieved if a square wave amplitude modulation is phase-locked to the train of pulses, in which case f_{rep} is reduced. This is illustrated in the lower panels of Fig. 1 where the terahertz comb resolution is effectively doubled for a two-fold smaller f_{rep} based on pulse picking.
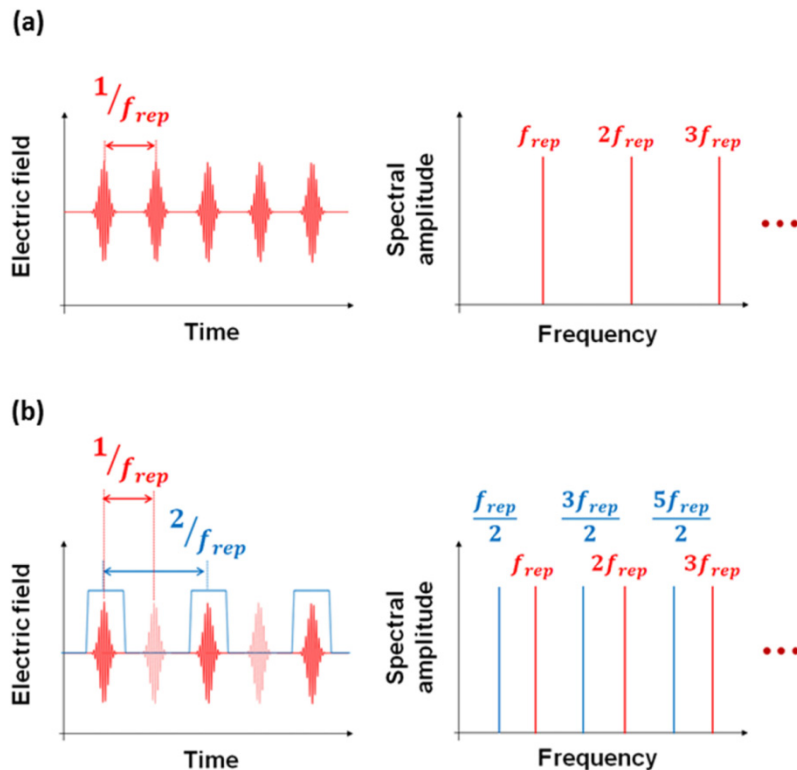

Fig. 1. Illustration of the pulse-picking concept in the time and frequency domains; (a) original pulse train (red) of a mode-locked laser (left panel) and corresponding terahertz comb spectrum (right panel); (b) pulse train where an applied phase-locked square wave amplitude modulation (blue) passes every other pulse to give the RF comb shown on the right with two-fold enhanced resolution.

Figure 2 depicts the details of our THz-DFCS spectrometer. Two Yb-doped fiber lasers based on polarization rotation nonlinear mode-locking with 100 MHz pulse repetition rate [15] are used to drive the emitter (Tx) and receiver (Rx) PCAs. A small portion of each laser beam is deflected to two fast photodiodes to provide input signals to a phase locked loop circuit that keeps the repetition rate difference (Δf_{rep}) between the oscillators under 10 Hz (such small Δf_{rep} was required because of the low 200 kHz bandwidth of our present detection system). The main outputs of both lasers are sent through polarization maintaining (PM) fibers to Mach-Zehnder intensity modulators (EOM), each driven by a DC power supply and an arbitrary function generator (AFG). Each AFG is clocked by its respective Yb oscillator to ensure a fixed phase difference between the laser and modulation signals. The output signals

of the EOMs are then fiber coupled to Yb-doped fiber amplifiers (YDFA). This amplification compensates for the insertion losses of the EOMs and free-space grating compressors [16] – which are necessary to ensure 115 fs pulses – to maintain the 20 mW average power level required to excite the PCAs (bow-tie shaped electrodes, Batop GmbH).
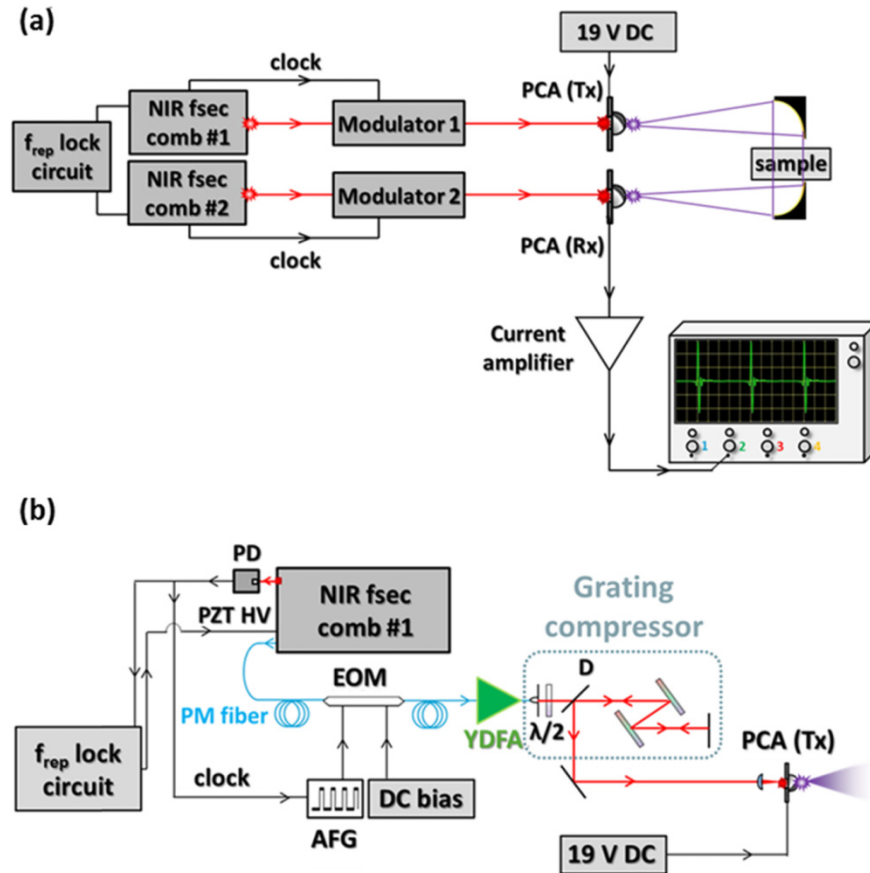

Fig. 2. (a) THz-DFCS setup overview and (b) detailed diagram shown for the generation arm only (receiver arm is nearly identical). NIR fsec comb #1 and #2: Yb-doped mode-locked fiber lasers; PD: fast photodetectors (1 GHz bandwidth); PZT HV: high voltage (100 V) piezo actuator; EOM: Mach-Zehnder electro-optic intensity modulator; AFG: arbitrary function generator; YDFA: Yb-doped fiber amplifier; D: D-shaped mirror; PM fiber: polarization maintaining fiber.

3. Results

Prior to implementing the pulse picking, the performance of our THz dual frequency comb spectrometer was evaluated at the original 100 MHz repetition rate. In Fig. 3, the time domain signals of a single scan, obtained for a 10 s time window, and an average of 256 waveforms (42 min total measurement time) are shown. As a first observation regarding the waveform amplitudes, the fact that averaging can be made over a long period of time without any phase correction, confirms that the pulse jitter stabilization requirements are not as demanding for terahertz systems as they are for Mid- or Near-IR dual-comb spectrometers. While both MIR combs generated by difference frequency generation and THz combs generated in PCAs are f_{ceo}-free, the scaling of the residual phase noise in the RF stabilization of f_{rep} is roughly 100-fold larger in the MIR region compared to the FIR/THz region. Indeed, our phase-lock loop circuit is simpler than those required for MIR or NIR dual-comb spectroscopy [17]. The pulse

repetition rate (f_{rep}) of each laser is detected directly using a photodiode and its 10th harmonic at ~1 GHz is phase-locked to a microwave oscillator referenced to a H-maser. We lock f_{rep} of one laser to that of the other by feedback only to a PZT-mounted mirror. The servo bandwidth is approximately 10 kHz. The relaxed stabilization requirements for terahertz dual combs can be understood by considering that any residual fluctuation in the error signal detected at 1 GHz will be multiplied to terahertz frequencies by factors that are 100's of times smaller than multiplication factors for MIR or NIR frequencies. A direct consequence of this readily-achieved stable phase-lock is the dramatic improvement in the SNR due to the coherent averaging of the random fluctuations. Yet, the effect of a residual jitter can nonetheless be noticed in Fig. 3(b): the first pulse that is used to trigger the waveform has considerably higher amplitude than the remaining pulses. Furthermore, amplitude fluctuations in Fig. 3(a) are clearly smaller than in Fig. 3(b) and change randomly across the whole time window. Since such differences cannot be detected without averaging, this effect can be attributed to phase variations that accumulate with time, decreasing the average amplitude of pulses detected at longer times. In fact, a slight triangular slope with time can be noticed for the pulses peak intensity in Fig. 3(b).
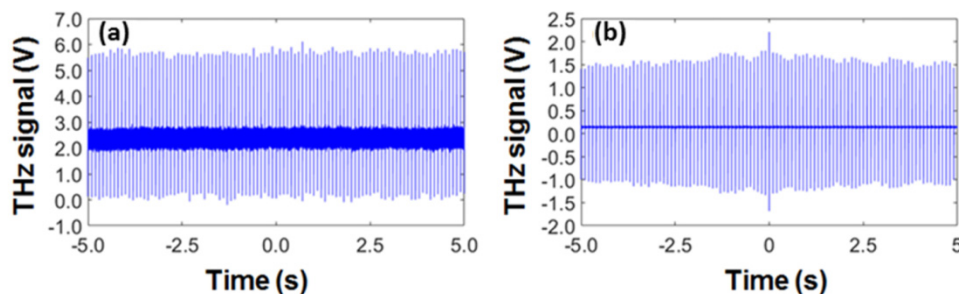

Fig. 3. Time domain signals acquired with the THz-DFCS spectrometer running at 100 MHz. (a) single waveform; (b) average of 256 waveforms.

The Fourier Transform of the averaged signal yielded a terahertz spectrum up to 800 GHz with a 100 MHz comb tooth spacing, which is presented in Fig. 4(a). This limited bandwidth is primarily due to the intrinsic limitation of the PCA emission/detection scheme. However, a comb mode resolved spectrum is readily obtained, with a peak dynamic range of approximately 23 dB (Fig. 4(b)). Figure 4(c) shows the comb lines at low frequencies and the absence of f_{ceo}. Two water lines at 557 and 752 GHz appear in the spectrum because the measurements were made without purging the path between the antennas. Figure 4(d) shows the 557 GHz water line, with a measured pressure-broadened FHWM linewidth of 6 GHz, in agreement with a simulation based on HITRAN12 for our ≈30 cm path length, atmospheric air pressure of ≈83 kPa, water vapor partial pressure of ≈0.5 kPa and room temperature (296 K). The additional structure that appears on top of the Lorentzian line shape likely arises from standing wave interference effects that can be minimized following a background ratio with the use of an evacuable absorption cell between the antennas.
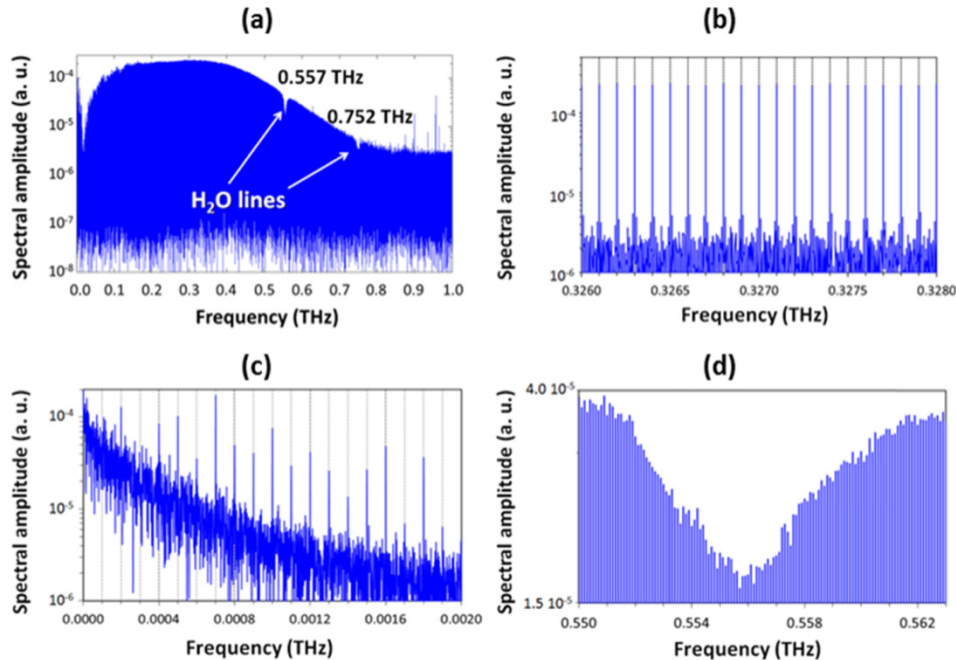
Fig. 4. Terahertz spectrum obtained with the 100 MHz dual comb system. (a) Full spectrum; (b) close up of the spectrum around 330 GHz, showing the peak dynamic range of 23 dB and the 100 MHz tooth spacing; (c) lowest frequency portion of the spectrum showing the absence of f_{ceo}. Dashed lines are offset from zero by 100 MHz intervals; (d) atmospheric H_2O line near 557 GHz.

In Fig. 5, pulse trains obtained at different modulation frequencies are compared with the original pulse train at 100 MHz. In the top three panels on the left, averages of 256 waveforms (same time window and integration time as the previous data shown in Fig. 3(b)) are shown for repetition rates of 100 MHz, 50 MHz and 25 MHz, and in the lower two panels, 64 waveforms were averaged for repetition rates of 10 MHz and 5 MHz. It can be seen that the concept of pulse-picking was successfully implemented, at least for the evaluated frequencies: the intermediate pulses were largely suppressed, and the repetition rates were effectively divided by the modulation frequency. In the frequency domain, the additional comb modes were readily observed in the expected positions in the spectra. Furthermore, the YDFAs allowed the average power level to remain nearly constant as more pulses were excluded. Since the PCAs are operated below saturation, this amplification is required to recover the original THz power, and ensure an optimal signal-to-noise ratio for each modulation frequency.

Dynamic ranges measured at the original 100 MHz comb teeth, remained near 20 dB for the higher modulation frequencies, but decreased to 13 dB and 8 dB for the 10 MHz and 5 MHz spectra, respectively. Signal-to-noise ratios followed the same trend, starting at 15 dB for the original comb, and dropping to 7 dB for the 5 MHz comb. An explanation for these findings can be drawn from the left panels in Fig. 5. It is clear that the complete suppression of the intermediate pulses was not achieved, leading to less intense comb teeth as the modulation frequency decreased. The reduced suppression was found to result from insufficient optical bandwidth of one of the EOMs which led to incomplete modulation of the full optical spectrum of one of the Yb oscillators (1020 to 1080 nm). An EOM with a well-matched optical bandwidth to the oscillator will lead to an overall flatter output comb spectrum.
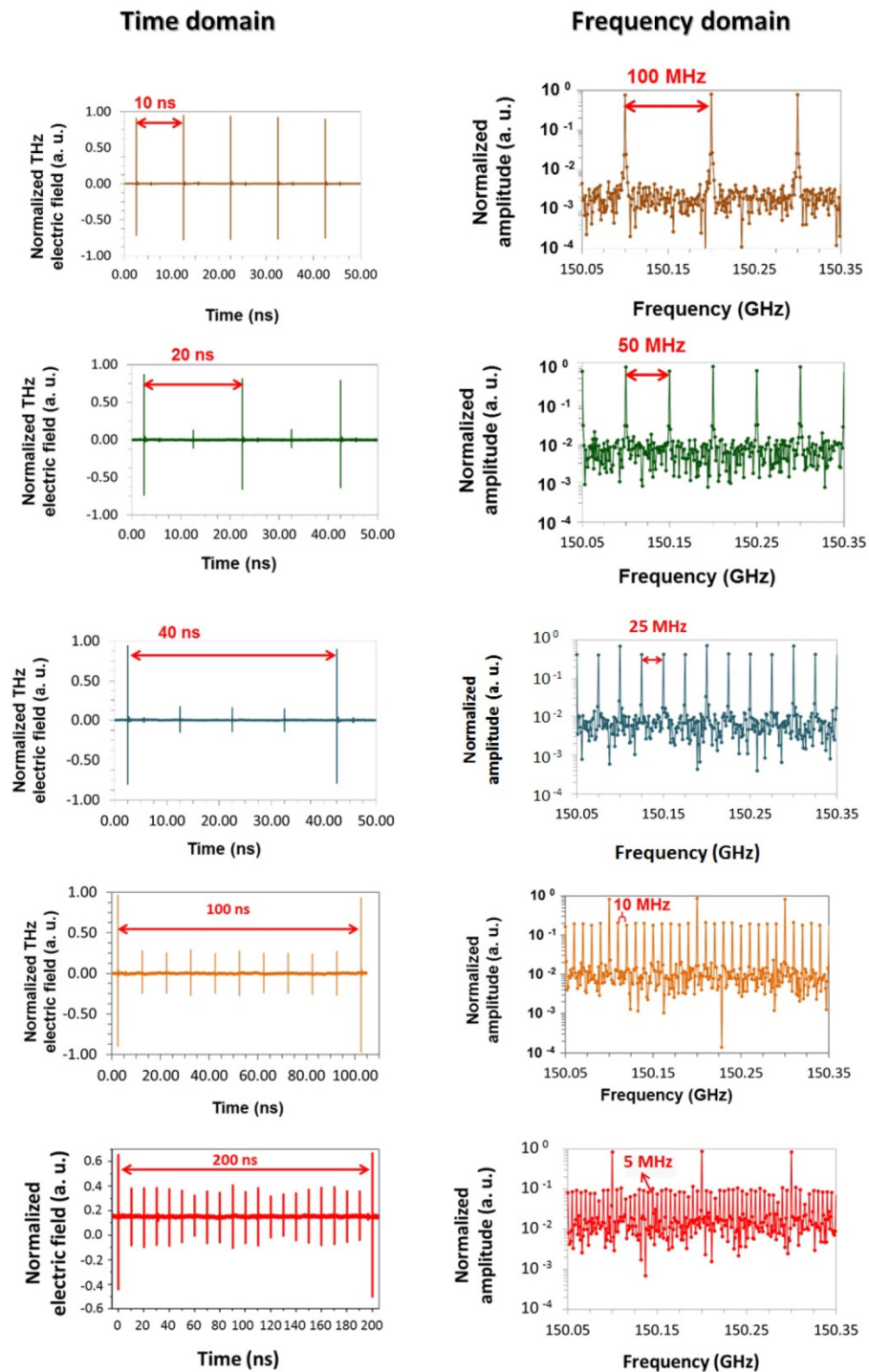
Fig. 5. Dual comb trains of pulses (left) and their spectra (right) acquired at different modulation frequencies. The upper three data sets were acquired as an average of 256 waveforms in a 10 s time window and the lower two sets were obtained for an average of 64 waveforms. From top to bottom, it is clear that the modulation introduced additional comb teeth within the 100 MHz original gaps.

4. Conclusion and perspectives

In this paper, we demonstrated a simple and effective electro-optic technique to adaptively reduce the repetition rate of a terahertz-dual-frequency comb system, thus enabling agile changes in the spectral resolution (up to 20-fold demonstrated here) for high resolution terahertz studies at low pressure where the selectivity and sensitivity are optimal for gas phase detection. The modulation frequency can be easily tuned using conventional digital synthesis, and for unsaturated THz generators (PCAs in this case), any loss in average power is readily compensated with unsaturated optical amplifiers, thus minimizing the power/resolution tradeoff resulted from the pulse-picking procedure.

The present limitations of our system that have reduced the overall terahertz bandwidth and decreased the signal-to-noise ratio observed at the highest spectral resolutions can be readily addressed by well-established approaches. Such enhancements include replacing the detection system with an electro-optic detection scheme which is well known for its broader bandwidth and sensitivity, implementing higher-power interdigitated PCAs, and improving the pulse-picking effectiveness using EOMs that suppress the full bandwidth of the optical pulses. With such improvements, this terahertz dual-frequency comb instrument will be ideally suited for precision molecular rotational comb spectroscopy, and line broadening studies over a wide range of pressure and temperatures.

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