Decade-Spanning High-Precision Terahertz Frequency Comb

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Decade-spanning, high-precision Terahertz frequency comb

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The generation and detection of a decade-spanning TeraHertz (THz) frequency comb is reported using two Ti:Sapphire femtosecond laser oscillators and ASynchronous OPtical Sampling THz Time-Domain Spectroscopy (ASOPS-THz-TDS). The comb extends from 0.15-2.4 THz, with a tooth spacing of 80 MHz, a linewidth of 3.7 kHz, and a fractional precision of 1.8×10^{-9}. With time-domain detection of the comb, we measure three transitions of water vapor at 10 mTorr between 1-2 THz with an average Doppler-limited fractional accuracy of 6.1×10^{-8}. Significant improvements in bandwidth, resolution, and sensitivity are possible with existing technologies.

The large bandwidth and high-resolution of optical frequency combs have made them an essential part of contemporary physics [1]. In metrology, visible/near-IR frequency combs have played an integral role in optical clocks and the most accurate measurements of fundamental constants [2], while in chemical physics they have been used for high-resolution molecular spectroscopy [3]. In addition, frequency combs have provided astrophysicists with a precise calibration source for radial velocity searches for Earth-sized exoplanets [4] and in measurements of the universe's expansion history [5].

Uniquely informative transitions are present in the TeraHertz (0.1-10 THz), or far-infrared, region of the electromagnetic spectrum, including molecular rotations and low-energy, large-amplitude vibrations. Various continuous wave technologies have been developed at THz frequencies, including those that potentially can be tuned over a decade of bandwidth rapidly [6], but for poorly characterized systems the search space is exceedingly large. Broadband, high-precision measurements of complex molecular spectra with THz combs will benefit fields such as molecular astrophysics [7] and atmospheric chemistry and physics [8], as precise line-centers and intensity measurements over wide spectral regions are critical for interpreting observational data. Further, THz studies of hydrogen-bonded clusters provide exacting tests of intermolecular forces [9], but particularly suffer from the need to detect narrow transitions over wide spectral windows.

Thus, extending the precision and bandwidth achievable with frequency combs into the THz region is an active area of research. The two main approaches used to date involve direct synthesis via quantum cascade lasers (QCLs) [10, 11] and downconversion of femtosecond pulse trains from optical oscillators [12]. QCLs generally yield higher-precision, lower-bandwidth measurements, while downconversion techniques generally yield lower-precision, higher-bandwidth spectra. Alternatively, hybrid approaches with a narrowband QCL coupled with a downconverted optical oscillator [13], or electronic heterodyne detection coupled with a downconverted optical oscillator [14], have also enabled high-precision, low-bandwidth measurements. It should be emphasized, that in all of these techniques, there is a trade-off between the detected bandwidth and precision.

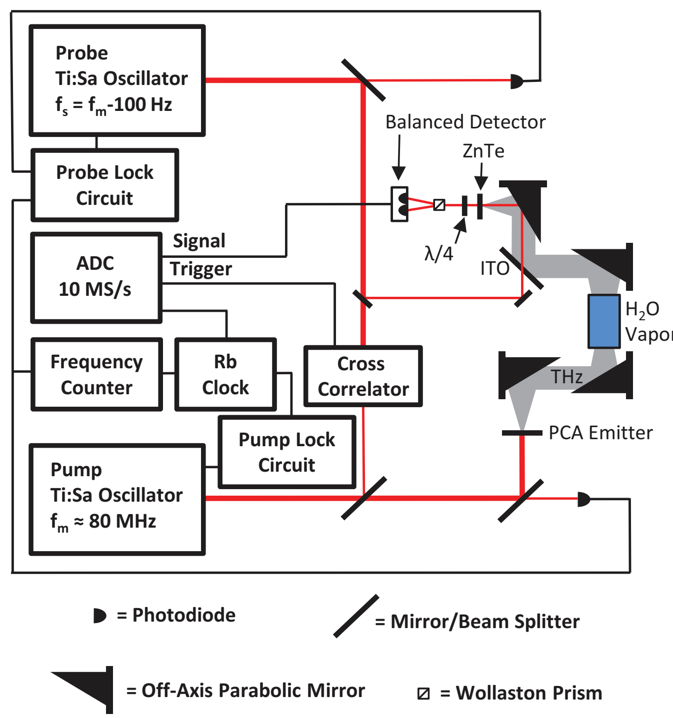

FIG. 1. The experimental layout for the generation and detection of the THz frequency comb. See text for details.

We demonstrate here the generation and coherent detection of a THz comb with both a decade-spanning bandwidth and high precision using ASynchronous OPtical Sampling THz Time-Domain Spectroscopy (ASOPS-THz-TDS) [15]. The first application of this technique to THz frequency combs was demonstrated in 2014 by Hsieh et al. using two 250 MHz Er-doped fiber lasers [12], who report a fractional accuracy of 8.4×10^{-7} for two transitions of acetonitrile at 0.64 THz with a tooth

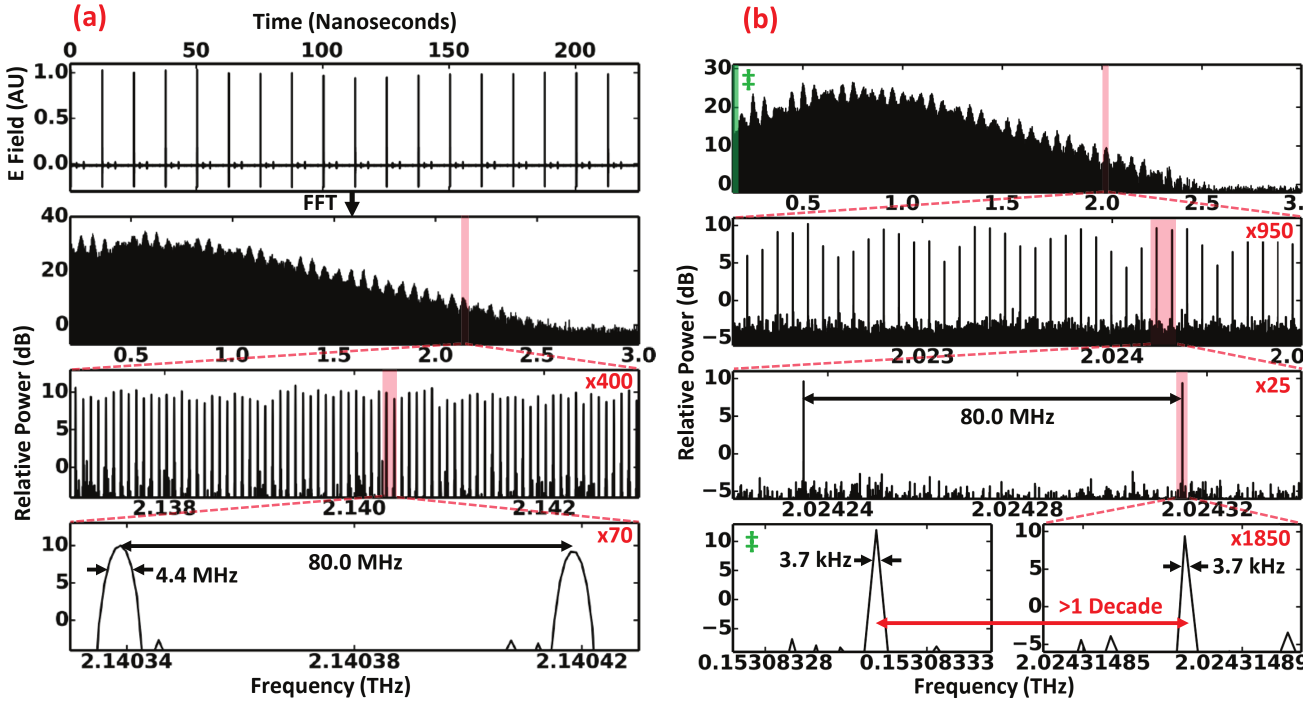

FIG. 2. (a) The Fourier transform of a 225 ns THz pulse train in delay time yields a decade-spanning frequency comb that extends up to 2.4 THz, with a 4.4 MHz tooth width. The peak dynamic range of the comb is 30 dB after 30 minutes of integration. (b) The tooth width of the THz comb can be reduced to 3.7 kHz by extending the time domain measurement to 268 μsec in delay time. The peak dynamic range is 20 dB after a single 107 s acquisition. Precision is preserved over the entire bandwidth of the comb, as shown in the bottom two panels.

width of 2.5 MHz. We have measured a 14-fold improvement in fractional accuracy and a 675-fold reduction in tooth width using a newly constructed ASOPS-THz spectrometer with two 80 MHz Ti:Sapphire femtosecond laser oscillators (Coherent Micra 5). Reaching this level of precision and accuracy is significant for THz metrology, as it is the first demonstration of broadband, Doppler-limited spectroscopy in the THz region. The Ti:Sapphire oscillators extend the bandwidth of the instrument to measure transitions between 1-2 THz, and can be further extended to 20 THz with proper optical pulse compression and THz emitter optimization, with no loss in fractional precision [16].

An instrument schematic is shown in Fig 1. A photoconductive antenna (PCA) emitter generates the comb. Since the downconversion process is dependent on the envelope of the optical pulse and not the phase, the THz pulses are effectively carrier-envelope-phase (CEP) stabilized without needing optical CEP stabilization. The repetition rate of the THz comb can be controlled by changing the repetition rate of the pump laser. Thus, a shift of 1 kHz in the 80 MHz pump laser corresponds to a shift of 12.5 MHz in the comb tooth at 1 THz. Here, the pump laser repetition rate is controlled by a piezo actuator and by changing the setpoint temperature of the water chiller, which cools the gain medium and baseplate of the modelocked oscillator.

After generation, the THz comb is detected via electro-optic (E-O) sampling by overlapping the 800 nm pulse train of a second laser, designated the probe in Fig. 1, with the THz comb in a ZnTe crystal. The THz electric field, which consists of a pulse train of $\sim$1 ps THz pulses, causes a transient birefringence in ZnTe, thus rotating the polarization of the 800 nm probe pulse train [17, 18]. The repetition rate of the probe laser is offset locked by 100 Hz from that of the pump, sweeping the probe pulse train through 12.5 ns of delay time (the round-trip time of the oscillator cavities) over 10 ms in the lab frame. A second lock circuit stabilizes the repetition rate of the pump laser over timescales $\geq$1 s. The full details of the lock circuits, and THz optics are reported elsewhere [19, 20]. The polarization of the probe light is measured by a balanced detector, and digitized.

Since the ASOPS technique ensures a repetitive phase walkout in delay time, the THz comb can be recorded in the time domain by extending the sampling window beyond the 12.5 ns interval between individual pulses. For example, the Fourier transform of a set of 18 THz pulses (covering 225 ns of delay time) yields a comb from 0.15-2.5 THz with a 4.4 MHz tooth width [30 minutes of data, Fig. 2(a)]. The averaging rate of the experiment is inversely proportional to the measurement window, so fewer averages are collected for longer pulse trains. However, an equivalent dynamic range is recovered in the

FFT, since the dynamic range increases linearly with the length of the measured waveform (assuming an adequate effective number of bits in the digitizer). Furthermore, since the frequency bins of the comb are much narrower than those of the isolated pulse experiment, we achieve a greater sensitivity to narrow-linewidth transitions. When measured for the same integration times, water vapor at 10 mTorr shows no clear absorptions in a high dynamic range, 12.5 ns window, while the lines are nearly saturated with a 225 ns-averaged comb (Fig 3(b-d)).

In the frequency domain, the experiment is equivalent to heterodyne detection with dual THz combs [12]. From this viewpoint, the THz pulse train is the signal and the probe laser acts as an effective THz local oscillator. Although the probe oscillator is not a true THz comb, it acts as one via E-O sampling, as the process is dependent on the envelope of the optical field. The mixing between the THz comb and the optical pulse train generates an RF comb in the birefringence of the ZnTe with a tooth spacing equal to the offset between the lasers, and with the nth tooth of the RF comb corresponding to the nth tooth of the THz comb. Thus, for an 80 MHz pump laser repetition rate, the 12500th tooth of the signal comb at 1 THz is detected as the 12500th tooth of the RF comb at 1.25 MHz.

To test the precision of the THz comb, we extended the measurement window to 21,470 individual THz pulses, or 268 μsec, increased the offset between the lasers to 200 Hz, and increased the sample rate of the digitizer to 20 MS/s (Fig. 2(b)). A larger laser offset lowers the ratio of lab time to delay time, such that a higher-resolution comb is acquired in an equivalent measurement time.

For short THz pulse trains, the tooth width is set by the Fourier time-frequency uncertainty relationship. However, longer pulse trains are more sensitive to various sources of jitter and drift in the system. Jitter in the offset between the pump and probe lasers, for example, will change the spacing of pulses in the time domain and thus shift the comb teeth in the frequency domain. Changes in the repetition rate of the pump laser will shift the comb teeth in the frequency domain as well. Either can broaden the teeth beyond the expected Fourier time-frequency product. For a single 107 s acquisition, we observed a transform-limited linewidth of 3.7 kHz over more than a decade of bandwidth, and thus no significant jitter in the instrument over this timescale. The tooth shown at 2.02 THz (Fig. 2(b)) demonstrates a fractional precision of $\Delta\nu/\nu = 1.8 \times 10^{-9}$.

The accuracy of our THz comb is likely limited by the lock circuit and ultimately by the Rubidium frequency reference, with a fractional uncertainty of 10^{-11}. To verify the accuracy of our instrument, we tested the 225 ns comb on rotational transitions of gas-phase water in a low-pressure cell and compared our values with the JPL spectral line catalog (Fig. 3) [21]. Initially, we took a broadband scan of water vapor at 100 mTorr, using 11 coarse steps in repetition rate to cover the spectrum between 1-2 THz, as shown in Fig 3(a). The RMS of the 16 transitions analyzed was 2.0 MHz, due to the large step size. We then performed 17 fine steps on three transitions of water vapor at 10 mTorr. Here the lines are primarily Doppler-broadened and accordingly fit to a gaussian lineshape (Fig 3(b-d)). The measured RMS of the three transitions was 92 kHz, with an average accuracy of $\Delta\nu/\nu = 6.1 \times 10^{-8}$, as shown in Table 1. Since the measured accuracy is significantly less than the Doppler linewidth, it should be possible to improve the measured RMS by reducing the linewidths of the transitions with a molecular beam. Indeed, the number density of the cell at 10 mTorr (10^{14} molecules/cm^3) and pathlength of the cell (10 cm) are similar to those achievable in molecular beams, suggesting that this comb would have sufficient sensitivity and resolution for such experiments.

TABLE I. Measured frequencies of three water vapor transitions at 10 mTorr.

$J_{K_aK_c} - J_{K_aK_c}$	This Work (MHz)	JPL[a] (MHz)	Diff. (MHz)
$3_{21} - 3_{12}$	1162911.521	1162911.602	-0.081
$2_{12} - 1_{01}$	1669904.836	1669904.775	0.061
$3_{03} - 2_{12}$	1716769.755	1716769.633	0.122
		RMS (MHz)	0.092

[a] Reference [21].

In summary, we have generated and detected a THz comb that spans over a decade in bandwidth with a transform-limited frequency precision of 1.8×10^{-9}. The average fractional accuracy (6.1×10^{-8}) of the comb was measured on three transitions of water vapor and is Doppler-limited. Although there are several approaches to Doppler-limited, far-infrared spectroscopy, we have achieved a single measurement bandwidth of >2 THz, which is an order-of-magnitude larger than any other technique. Furthermore, we are able to generate and detect the comb with all room temperature components, while many other methods require cryogenic detectors or generators.

The applications of this comb are immense, and could include THz metrology, and measurements in support of the Hershel Space Observatory, and the Atacama Large Millimeter Array, as these observatories have complementary spectral coverage and resolution to the THz comb. Broadband studies of vibration-rotation tunneling transitions in hydrogen-bonded water clusters are another exciting application; the water trimer and quatramer both have such transitions within the spectral range of this comb [22, 23]. The direct time-domain detection of the comb is also suitable for broad continuum measurements, such as pressure-broadened or condensed-phase samples. By implementing a commercially available pulse compressor on the pump and probe oscillators, and by em-

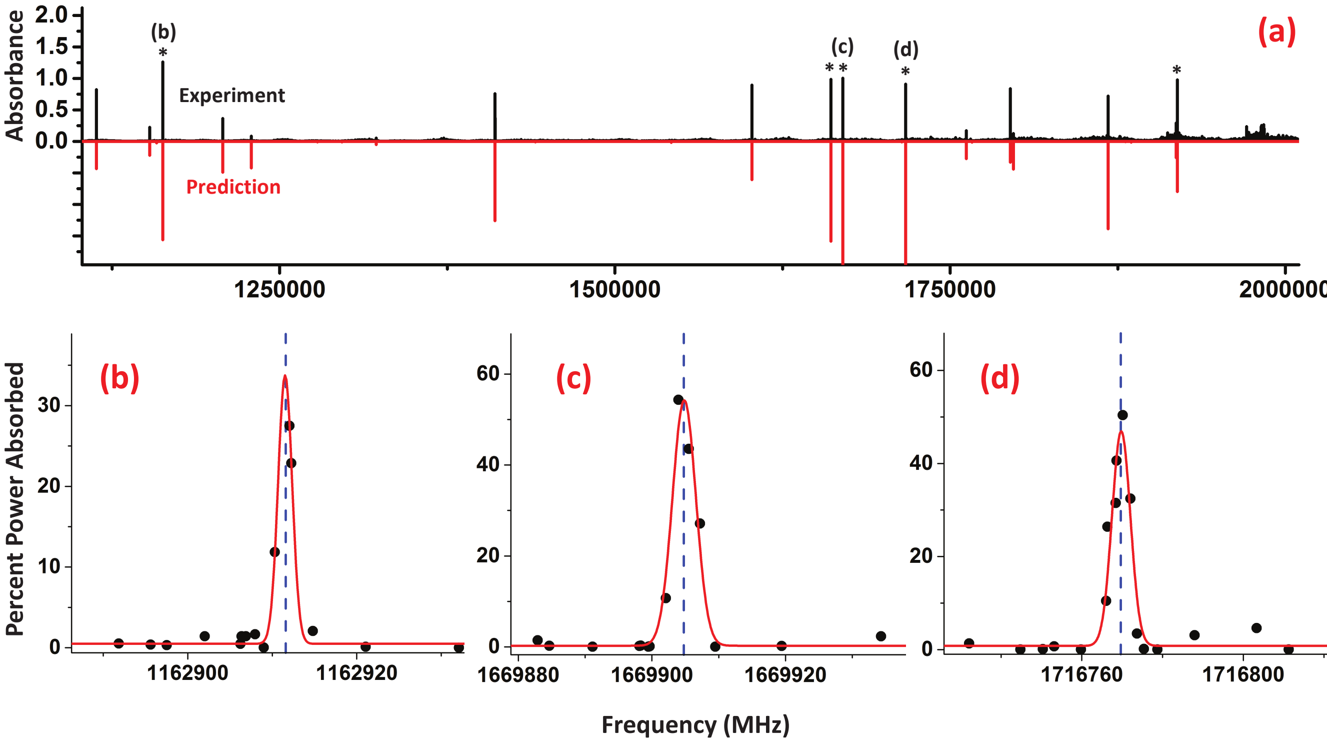

FIG. 3. (a) The broadband spectrum of water vapor at 100 mTorr with 11 repetition rate steps. The predicted spectrum from the JPL spectral line catalog [21] is shown in red, with negative intensity. The asterisks (*) indicate saturated transitions. (b-d) The gaussian fit (red line) to three transitions of water vapor at 10 mTorr. Line centers from the JPL catalog are indicated with vertical blue dashed lines.

ploying both transmission and reflection geometries for the PCA THz emitter, the comb could also be extended to 20 THz, a century in bandwidth. A comb with these properties could measure all of the intermolecular modes of hydrogen-bonded clusters in a single experiment. Since the stability of the comb is dependent on Ti:Sapphire oscillilators, many of the stabilization techniques that have been developed in optical metrology are applicable, and could improve the linewidth of the comb considerably. These improvements could open new fields of precision metrology in the THz region of the electromagnetic spectrum.

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