# Asynchronous optical sampling with GHz repetition rate femtosecond lasers for high precision Terahertz spectroscopy

***T. Dekorsy*** ***G. Klatt, R. Gebs, C. Janke, and A. Bartels***
Department of Physics and Center for Applied Photonics, University of Konstanz, D-78457, Germany,
thomas.dekorsy@uni-konstanz.de

## Abstract

We report a terahertz time-domain spectrometer with 6 THz spectral coverage and 1 GHz resolution which is based on high-speed asynchronous optical sampling. High-speed asynchronous optical sampling is based on two femtosecond lasers with approximately 1 GHz repetition rate. The two lasers are stabilized in their repetition rate to an off-set frequency of approximately 2 kHz. The time delay between a pump pulse exciting a photoconductive THz emitter and the probe pulses is hence scanned at 2 kHz scan rate without employing a mechanical delay stage. The timing jitter between the pump and the probe pulses is reduced to 40 fs over the full scan range giving a high time-resolution. We demonstrate the capabilities of this system for gas spectroscopy and the characterization of frequency selective surfaces with characteristic frequencies in the THz range.

## 1. Introduction

In the past years terahertz time-domain spectroscopy (THz-TDS) has evolved as important tool for applications in THz science and technology, e.g. including drug detection and food inspection, detection of explosives, or environmental gas analysis and sensing [1,2]. The established techniques for THz-TDS use ultrashort laser pulses from a single mode-locked laser for the generation and a delayed probe pulse for detection of THz radiation. Such systems have been demonstrated covering a frequency range from tens of GHz well into the mid-infrared [3-6]. One disadvantage of such conventional systems is the fact that they are employing mechanical delay stages to adjust the timing between the THz generation and detection pulses. These delay stages are result in a low acquisition rate for THz transients. As an example, to detect a 1 ns long THz transient requires a delay stage travel distance of 15 cm. This time delay would yield a 1 GHz resolution in the THz spectrum after a Fourier transform. Such a resolution is typically needed to resolve spectral absorption features of environmental gases. The time required for accelerating the mechanical delay stage and to average out laser noise requires an acquisition times in the range of a few tens of minutes. This long data acquisition time is a strong restriction for applications where the acquisition of a THz spectrum must be completed in a time below a second. Such applications include the fast readout of THz sensors investigations of the structural dynamics in biomolecules or experiments under rapidly varying environmental conditions [7]. Faster mechanical scanning approaches using rotating mirrors have been demonstrated with up to 400 Hz scan rate and up to 1 ns time delay [8,9].These results present an improvement with respect to linear stages, however, disadvantages remain such as a significant acoustic noise due to the rotating devices. We also like to mention that all mechanical delay stages are a source of calibration errors of amplitude and time-delay due to potential misalignment.

Asynchronous optical sampling (ASOPS) is a time-domain spectroscopy technique first introduced with picosecond lasers in the late 80's that does not need a mechanical delay stage and thus eliminates the problems discussed above [10]. Recently we introduced high-speed ASOPS which uses two Ti:sapphire femtosecond lasers whose repetition rates of 1 GHz are linked with an offset $\Delta f_R$ of a few kilohertz [11]. As a result of the offset, the delay between pairs of pulses from the lasers is scanned between zero and the inverse laser repetition rate at a scan rate equal to $\Delta f_R$. The main advantage of high-speed ASOPS is the scan rate of several kilohertz. This yields a performance that is impossible with conventional systems. It allows us completing the acquisition of a single time-domain trace before any technical noise of a femtosecond laser with significant Fourier components in the range up to 1 kHz can affect the signal. This permits measurements at the shot-noise limit without the use of lock-in amplifiers or other noise suppression techniques.

In order to realize a THz-TDS system, one laser is used to generate the THz radiation, the second laser is used to detect the THz waveform in the same detection geometry as in a conventional THz-TDS setup [4-7]. The THz waveform is mapped onto the intensity of the detection laser beam which is measured and digitized as a function of real-time $t$ and recalibrated to a time-delay scale $\tau$ using a factor $\Delta f_R/f_R$, where $f_R$ is the repetition rate of the THz generation laser. High-speed ASOPS has been realized previously with a time resolution of approximately 160 fs, limited by timing jitter between the offset-locked lasers. This has enabled THz-TDS at 1 GHz resolution and 10 kHz scan rate without mechanical delay stage at a spectral coverage of 3 THz [11]. Here we report a high-speed ASOPS based THz-TDS

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system with a significantly improved time resolution of 40 fs [12,13]. This time resolution is determined by the laser pulse duration, leading to a spectral coverage of more than 6 THz. To assess the utility of the system for rapid and precise spectroscopy, we have carefully compared a measured water vapor absorption spectrum to data reported in the HITRAN database [14,15].

## 2. High-speed asynchronous sampling set-up

The laser system and optical setup are equivalent to a previously reported system [11]. Two fs Ti:sapphire ring-lasers with $f_R = 1$ GHz are offset-locked at the third repetition rate harmonic with $\Delta f_R = 10$ kHz. The schematic of the set-up is sketched in Figure 1. Four improvements were implemented in order to enhance the bandwidth performance [12]: i) The phase-locked loop electronics which is used to offset-stabilize the two lasers operate at the tenth harmonic of the repetition frequency. Hence the feedback loop is able to perform a tighter offset lock with a reduced timing jitter due to an enhanced phase-noise sensitivity. The frequency shifter used to define the offset frequency is replaced by a direct-digital synthesis electronics which allows for a better calibration of the time-delay axis [13]. ii) The repetition rate difference is reduced to 2 kHz. The reduced repetition rate difference in combination with the 1 GHz repetition rate and the 100 MHz detector and A/D-converter bandwidth yields an enhanced density of the data points, i.e. one point every 20 fs. iii) The trigger signal for the data acquisition is generated optically via two-photon absorption in a GaP photodiode instead of electronically via mixing of electronic signals at the repetition rates. iv) The ZnTe crystal used for electro-optic detection of the THz transients via the Pockels effect was replaced with a 400 µm thick (110)-GaP crystal. Due to the high TO phonon frequency of GaP a larger bandwidth is achieved [16]. The THz transients are generated in a highly efficient large area THz emitter based on inter-digitated electrode design on a GaAs substrate [17].

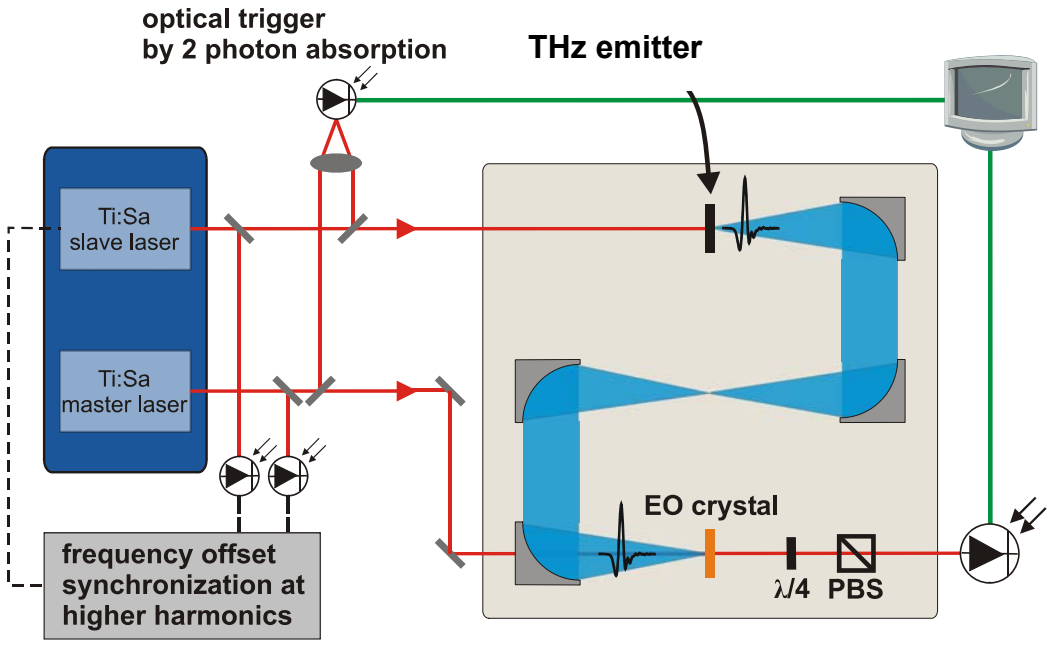


Fig. 1: Schematic of the ASOPS TD-THz spectrometer. Optical paths are red lines, THz paths are blue and electronic signals are dashed black lines.

## 3. High-resolution THz spectroscopy of water vapor absorption

To test the spectral accuracy of our THz-TDS system, atmospheric water vapor is one of the best suited samples [18]. Between 0.2-6.5 THz more than 1000 absorption lines are well known from experimental and theoretical

investigations and are documented in the HITRAN database [14,15]. The reported frequency error of the HITRAN data for water vapor is between 300 kHz and 3 MHz, enabling us to test our system at this accuracy level. The width of each absorption line at atmospheric pressure conditions lies in the range of 10 GHz. This is well-matched to the 1 GHz resolution of our THz-TDS system determined by the maximum time delay of 1 ns. Figure 2 shows the comparison between a measured and calculated water vapor absorption spectrum. The acquisition time for the trace with water vapor was 60 s, the reference data are the 9-minute data shown in Fig. 1. The data for the calculation are taken from the latest version of the HITRAN database [15]. The calculation of the spectrum is performed according to the formulas of reference [14]. During the 60 s measurement the ambient parameters were kept constant at a temperature of 20°C, a pressure 1015 hPa, and a relative humidity of 26%. Up to a frequency of 2.5 THz the line positions as well as the line strength agree very well. For greater frequencies absorption measurements are limited by the dynamic range of the spectrometer. Lines with an absorption higher than 0.15 $cm^{-1}$ cannot be fully resolved due to the limited signal-to-noise.

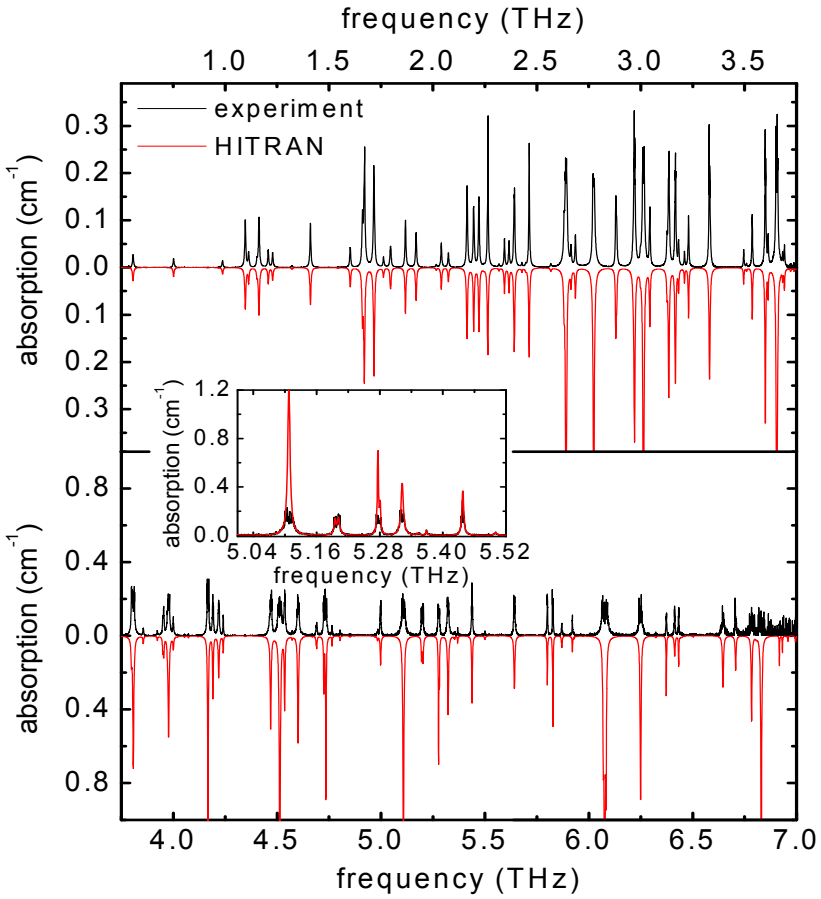


Fig. 2: Experimentally determined absorption of water vapor compared to the absorption data of the HITRAN data base. The inset shows details a part of the spectrum where the absorption is too high to determine the full line shape experimentally.

## 4. Conclusion

We have demonstrated a high-speed ASOPS based THz-TDS spectrometer which operates without a mechanical delay. At a scan rate of 2 kHz, the spectrometer offers more than 6 THz spectral coverage and 1 GHz spectral resolution. The precision of the spectrometer has been evaluated by measuring water vapor absorption and comparing it to data published in the HITRAN database. The mean deviation for the line positions is 142 MHz and the maximum deviation is 461 MHz at an acquisition time of 60 s and only slightly larger at 0.6 s acquisition time. The presented instrument should be useful for all applications where rapid data acquisition in combination with a high frequency accuracy and high resolution is important.

## 5. Acknowledgments

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