# Asynchronous optical sampling terahertz time-domain spectroscopy for ultrahigh spectral resolution and rapid data acquisition

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We propose a terahertz time-domain spectrometer based on asynchronous optical sampling. The spectrometer features ultrahigh spectral resolution equivalent to a laser mode-locked frequency and rapid data acquisition. The proposed method requires no mechanical translation stages for time-delay scanning, and hence, overcomes the inherent tradeoff between frequency resolution and data acquisition time. Time evolution of the picosecond terahertz pulse with a temporal window of 12.1 ns is measured directly on an oscilloscope using a time-scale magnification of 764 815. A frequency resolution of 82.6 MHz is achieved at a measurement time of 10 s. The effectiveness of the proposed method is confirmed by comparing it with conventional stage-scanning terahertz time-domain spectroscopy. © *2005 American Institute of Physics.* [DOI: 10.1063/1.2008379]

Terahertz time-domain spectroscopy (THz-TDS) has been attracting much attention as the basic measurement method in THz science and technology. In conventional THz-TDS, temporal profiles of a pulsed THz electric field are first measured by scanning the time interval between a THz pulse and a probe pulse using a mechanical translation stage. Then, broadband spectra of THz amplitude and phase are obtained through a Fourier-transform procedure. Frequency resolution and frequency range in THz-TDS are defined as reciprocals of an overall window and step increment in the time delay, which correspond, respectively, to the stroke length and step displacement in the mechanical stage. Thus, there is an inherent tradeoff between the frequency resolution and the measurement time in conventional THz-TDS. The problem would be largely solved if we could do away with the time-consuming mechanical stage. An effective method to realize a stage-free configuration involves the asynchronous optical sampling (AOS) technique, often employed in optical communication,[1,2] pump-probe spectroscopy,[3,4] Fourier-transform infrared spectroscopy,[5] and electronic THz spectroscopy[6] for rapid data acquisition. In this method, the use of two asynchronous mode-locked lasers magnifies the time evolution of the ultrafast transient signal arbitrarily in the time scale. The resultant signal can be measured directly and quickly on a conventional oscilloscope. The most attractive advantage of the AOS method in connection with THz-TDS is that it helps realize a time delay much longer than that achievable by the conventional motor-driven translation stage. When a temporal window equivalent to the THz pulse period is obtained, the theoretical limit of frequency resolution in THz-TDS is achieved. Thus, application of the AOS concept to THz-TDS provides an additional advantage besides rapid data acquisition: an ultrahigh spectral resolution equivalent to the laser mode-locked frequency. In this letter, we apply the AOS technique to THz-TDS (AOS-THz-TDS) to drastically improve the frequency resolution and decrease the measurement time.

The experimental setup is shown in Fig. 1. We used two femtosecond lasers for AOS-THz-TDS: a 60-fs Kerr-lens-mode-locked Ti:sapphire laser (ML-Ti:S-1) and a 10-fs Kerr-lens-mode-locked Ti:sapphire laser (ML-Ti:S-2). The

mode-locked frequencies of the two lasers ($f_1$ and $f_2$) are set approximately at 82.6 MHz. The difference in mode-locked frequency ($\Delta f = f_1 - f_2$) must be stabilized at a certain frequency to achieve the AOS method. Hence, a ring-type piezoelectric transducer (PZT, stroke=6 $\mu$m; resonance frequency=6 kHz) is attached to an output coupler in ML-Ti:S-1 for controlling $f_1$ within a ±125-Hz range. The difference-frequency signal (frequency=$10f_1 - 10f_2 = 10\Delta f$) between the tenth-harmonic components of the mode-locked frequency is extracted from two signals detected with photodiodes (PD) through a series of electronic heterodyned processes that employ a voltage-controlled oscillator (VCO), two double-balanced mixers (DBM), three low-pass filters (LPF), and an analog mixer ($M$). Finally, $f_1$ is controlled to keep $\Delta f = 108$ Hz via a phase-locked loop (PLL, reference frequency=1.08 kHz) and a PZT controller. The
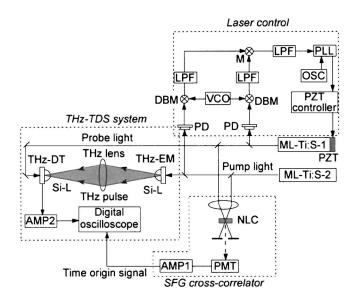


FIG. 1. Experimental setup. ML-Ti:S-1 and ML-Ti:S-2: mode-locked Ti:sapphire lasers; PZT: piezoelectric transducer; PD: photodiode; DBM: double-balanced mixer; VCO: voltage-controlled oscillator; LPF: low-pass filter; $M$: analog mixer; PLL: phase-locked loop. OSC: oscillator; NLC: LiB$_3$O$_5$; PMT: photomultiplier; AMP1 and AMP2: current preamplifiers; THz-EM: THz emitter; THz-DT: THz detector; Si-L: silicon lens.

resulting fluctuation of $\Delta f$ is suppressed within $108 \pm 0.1$ Hz at a gate time of 1 s. Portions of the two laser lights are fed into a SFG (sum frequency generation) cross correlator using a nonlinear optical crystal ($LiB_3O_5$, thickness$=2$ mm). The resultant SFG cross-correlation light is detected with a photomultiplier (PMT), amplified with a current preamplifier (AMP1), and then used as the time origin in AOS-THz-TDS. In THz-TDS, ML-Ti:S-1 and ML-Ti:S-2 are used as the probe light and pump light, respectively. We use two bowtie-shaped, low temperature grown GaAs photoconductive antennas (bowtie length$=1$ mm; gap$=5$ $\mu$m) as a THz emitter (THz-EM) and THz detector (THz-DT) to improve the detection sensitivity of a weak THz signal. The THz pulse radiating from the THz emitter is detected with the THz detector via two hemispherical silicon lenses (Si-L) and a THz lens ($f=50$ mm). After passing through another current preamplifier (AMP2; sensitivity$=10^6$ V/A, bandwidth $=200$ kHz), the signal can be directly observed at a scan rate of $\Delta f$ with a digital oscilloscope (bandwidth$=500$ MHz) triggered by the time-origin signal from the SFG cross correlator.

Figure 2(a) shows the temporal profile of a pulsed THz electric field measured by AOS-THz-TDS, in which signal averaging of 1000-sweep sequences was carried out by a digital oscilloscope (required time$=10$ s). The observed temporal profile is expanded by a temporal magnification factor of 764 815 ($=f_1/\Delta f=82\,600\,000/108$) according to the principle of the AOS method.[4] Furthermore, the temporally magnified THz pulse is sampled as a burst of the probe pulse with a period of $1/f_1$, which is equivalent to a step increment of 15.8 fs in real time delay [$=\Delta f/f_1 f_2=108/(82\,600\,000 \times 82\,600\,108)$]. The upper and lower horizontal scales in Fig. 2(a) indicate, respectively, the time scale on the oscilloscope and the *real* time scale translated using the above magnification factor. The temporal wave form of the pulsed THz electric field was measured within a temporal window of the pulse period ($=12.1$ ns). In contrast to conventional THz-TDS, AOS-THz-TDS can acquire the temporal wave form at a fixed scan rate of $\Delta f$ ($=108$ Hz) regardless of the size of the overall temporal window. A temporal region of 100 ps enlarged from Fig. 2(a) is shown in Fig. 2(b). The transient evolution of the THz electric field in the picosecond region is clearly observed. In the present system, the bowtie-shaped photoconductive antennas used as a THz emitter and detector limit the bandwidth of the THz pulse to a considerably narrow range, resulting in the long pulse width of the THz electric field ($=6.5$ ps). To compare with AOS-THz-TDS, we obtained the temporal profile of the THz electric field by conventional THz-TDS using a motor-driven mechanical stage and a lock-in amplifier, as shown in Fig. 2(c) (measurement time$=5$ min). The comparison indicated that AOS-THz-TDS reproduces the temporal profile of a pulsed THz emission accurately. The slight distortion of the observed THz pulse profile is due to the narrow bandwidth of the AMP2 and/or insufficient temporal magnification of the AOS method. It is important to emphasize that the signal-to-noise ratio observed in AOS-THz-TDS for a measurement time of 10 s is almost the same as that observed in conventional THz-TDS in 5 min. Furthermore, the proposed method requires no lock-in amplifier, which is usually used in THz-TDS for sensitive detection of a weak THz electric field. Figure 3(a) shows the THz amplitude spectrum calculated
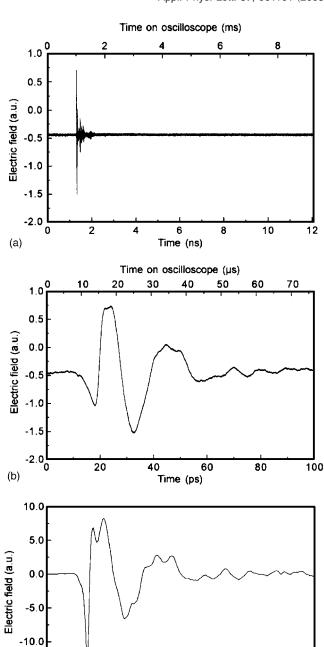


FIG. 2. Temporal wave forms of pulsed THz electrical field. (a) AOS-THz-TDS (full scale$=12.1$ ns), (b) AOS-THz-TDS (full scale$=100$ ps), and (c) stage-scanning THz-TDS (full scale$=100$ ps). The upper horizontal scales indicate the time scale on oscilloscope.

from the Fourier transform of the wave form in Fig. 2(a). The frequency resolution and frequency span are calculated to be 82.6 MHz and 68 THz, respectively, from an overall window of 12.1 ns and a step increment of 15.8 fs, if fluctuation in $\Delta f$ is neglected. However, the actual instability in $\Delta f$ fluctuates the frequency resolution and frequency span. For example, a fluctuation of 0.1 Hz in $\Delta f$ leads to those of 11.2 ps in the overall window and 0.015 fs in the step increment, resulting in those of 76.4 kHz in frequency resolution and 69.3 GHz in frequency span. The spectral structure in Fig. 3(a) is due to multiple reflection of THz pulse in a hemispherical silicon lens. The THz amplitude spectrum in Fig. 3(b) was obtained through conventional THz-TDS, by
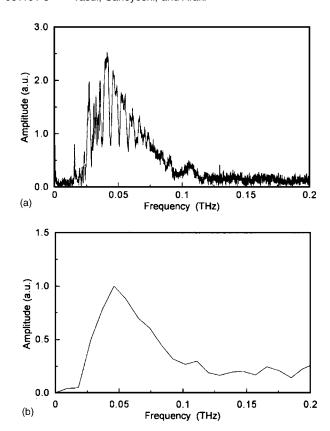
FIG. 3. Spectra of THz amplitude. (a) AOS-THz-TDS (frequency resolution=82.6 MHz) and (b) stage-scanning THz-TDS (frequency resolution=10 GHz).

taking the Fourier transform of the wave form in Fig. 2(c). A comparison between the two profiles reveals a remarkably improved frequency resolution in the case of AOS-THz-TDS. The frequency resolution of 82.6 MHz can be achieved in conventional THz-TDS if a long stage scanning equivalent to a laser cavity length (=1.82 m) is adopted. However, this result is extremely time consuming; over 10 h under the experimental conditions used in Fig. 2(c).

In conclusion, using the AOS-THz-TDS technique, we have obtained a frequency resolution of 82.6 MHz at a data acquisition time of 10 s. This method not only achieves the theoretical limit of frequency resolution in THz-TDS at the short measurement time but also opens the door to a compact, simple, and stable THz-TDS system with no mechanical parts by the combination of femtosecond mode-locked fiber lasers and fiber-coupled photoconductive antennas. The use of a broadband THz emitter and detector, adjustment of the temporal magnification factor, and/or use of broadband THz detection electronics will enable further expansion of the spectral bandwidth in AOS-THz-TDS.

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