Integrated system of traditional THz time-domain spectroscopy and asynchronous optical sampling

Jing Ding(丁晶), Qing-Hao Meng(孟庆昊), Yan Shen(沈妍), Chen-Xin Ding(丁晨鑫), Bo Su(苏波)†, Hai-Lin Cui(崔海林), and Cun-Lin Zhang(张存林)

Department of Physics, Capital Normal University, Beijing Advanced Innovation Center for Imaging Theory and Technology, Beijing Key Laboratory for Terahertz Spectroscopy and Imaging, Key Laboratory of Terahertz Optoelectronics, Ministry of Education, Beijing 100048, *China*

(Received 18 June 2022; revised manuscript received 28 August 2022; accepted manuscript online 9 September 2022)

Terahertz time-domain spectroscopy (THz-TDS) system, as a new means of spectral analysis and detection, plays an increasingly pivotal role in basic scientific research. However, owing to the long scanning time of the traditional THz-TDS system and the complex control of the asynchronous optical scanning (ASOPS) system, which requires frequent calibration, we combine traditional THz-TDS and ASOPS systems to form a composite system and propose an all-fiber trigger signal generation method based on the time overlapping interference signal generated by the collinear motion of two laser pulses. Finally, the time-domain and frequency-domain spectra are obtained by using two independent systems in the integrated systems. It is found that the full width at half maximum (FWHM) of the time-domain spectra and the spectral width of the frequency-domain spectra are almost the same, but the sampling speed of the ASOPS system is significantly faster than that of the traditional THz-TDS system, which conduces to the study of the transient characteristics of substances.

Keywords: terahertz time-domain spectroscopy (THz-TDS), asynchronous optical scanning (ASOPS) system, integrated system, trigger signal

PACS: 87.50.U–, 43.60.Qv **DOI:** 10.1088/1674-1056/ac90b2

1. Introduction

Terahertz (THz) wave is located between microwave and infrared, and the frequency ranges from 0.1 THz to 10 THz.[1] The data rates of this band can be expanded to terabits per second.[2] The discovery of laser-induced ultrafast demagnetization provides a perfect platform to manipulate spins on a sub-picosecond timescale and opens up a promising route to developing future information technology at THz speed.[3–5] In the past two decades, THz electromagnetic spectrum technology has advanced in both basic and applied research fields. Compared with traditional light sources, THz pulses have the characteristics of transient, broadband, coherence, and low energy, which are crucial for exploring material structures.[6] THz time-domain spectroscopy (THz-TDS) systems have been employed in the scientific research field for nearly 30 years. These systems are based on ultrafast optoelectronic technology to generate and detect THz waves, and the laser power ranges from a few mW to more than 3 W.[7,8] A traditional THz-TDS system divides a beam from a femtosecond laser into two beams, one is used as a pump light to generate THz waves, and the other is adopted as detection light to detect THz signals. The optical delay line is used to control the different optical paths of the two laser beams to adjust the time when the femtosecond laser reaches the antenna. Each step of the optical delay line only completes the sampling of one point in a THz pulse. As such, to obtain a complete THz pulse, the motor needs to step tens of thousands of times, which takes several minutes to more than ten minutes. Therefore, the delay device-based THz-TDS systems have limitations, *i.e.*, they cannot quickly detect THz pulses. The study of physical processes such as material transient changes relies on the detection of ultrafast pulse transient characteristics, so improving the pulse detection speed has become the only solution. The rapid detection of THz pulses by using asynchronous optical sampling (ASOPS)[9,10] technology can solve the above problems. The ASOPS system does not need a mechanical delay line, so its acquisition time is short, and it has high time resolution and frequency resolution. However, the ASOPS system usually uses piezoelectric ceramics to precisely control the repetition frequency. Since piezoelectric ceramics have high requirements for the ambient temperature, it needs to be calibrated regularly when the environmental conditions change. In addition, in the ASOPS system, the acquisition of high-stability trigger signal (TS)[11] determines the accuracy of the collected signal. Double-balanced mixer and two-photon absorption are two common methods of generating TS. However, these two methods have two main disadvantages. First, they require the two laser beams used to generate the TS should have high stability and directivity, which makes the optical path more complicated; second, the generation of TS needs to consume more laser power, which leads the pump power and detection power to account for only 60% of the total power, thus affecting the signal strength. In this work, we propose an all-fiber laser pulse coherence method, which can obtain high-

†Corresponding author. E-mail: subo75@cnu.edu.cn

© 2023 Chinese Physical Society and IOP Publishing Ltd

stability TS with only a few milliwatts of power. Furthermore, since the laser propagates in the optical fiber, there is no requirement for the directivity of the laser beams. Finally, we establish an integrated system of the traditional THz-TDS system and ASOPS system. The ASOPS system can significantly shorten the acquisition time of THz time-domain spectra, and the traditional THz-TDS system can provide the necessary calibration for the ASOPS system.

2. ASOPS principle

The ASOPS is a recent ultrafast THz-TDS technology independent of a machine translation platform.[12,13] It uses two femtosecond lasers to generate pump and detection pulses. The difference in repetition frequency between the two lasers is controlled by a high bandwidth feedback control circuit, ranging from less than 1 Hz to tens of kHz.[14,15] If the THz and detection pulses coincide at a certain time, there will be a difference in time between the two pulses when the next pulse arrives, and each pulse thereafter will have the same time difference until they coincide again.[16] Because the ASOPS system does not need a mechanical delay device, the mechanical error is reduced, and the THz spectra obtained have high accuracy. The sampling principle is shown in Fig. 1.[17]

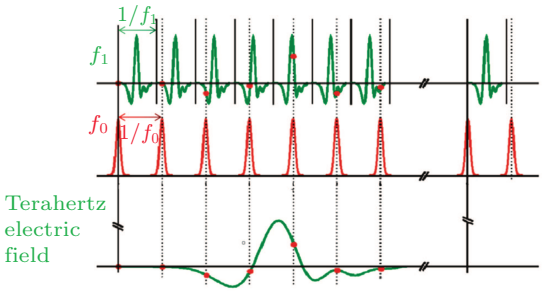


Fig. 1. Sampling principle of ASOPS system.

If the repetition frequency of the master laser and slave laser are f_1 (pump pulse) and f_0 (detection pulse), respectively, the time interval of the pump pulse is expressed as follows:

$$T_1 = \frac{1}{f_1}. \tag{1}$$

If the time difference between the detection pulse and the pump pulse arriving at an antenna each time is Δt, the time interval of the detection pulse is expressed as follows:

$$T_0 = T_1 + \Delta t. \tag{2}$$

Then, the repetition frequency of the detection pulse is expressed as

$$f_0 = \frac{1}{T_0} = \frac{1}{T_1 + \Delta t}. \tag{3}$$

Further, the difference in repetition frequency between the detection laser and the pump pulse is given by

$$\Delta f = f_1 - f_0 = \frac{\Delta t}{T_1(T_1 + \Delta t)}. \tag{4}$$

The total time of each signal acquisition is

$$T = \frac{1}{\Delta f}. \tag{5}$$

When the pump and detection pulses coincide at a certain time, a high-level pulse signal, which is also known as a TS, will be generated, and its period is the reciprocal of the difference in repetition frequency. When each TS is generated, the acquisition begins to collect the signal, and when the next TS is generated, the acquisition ends. Therefore, T is also the total time of each THz signal acquisition.

3. Acquisition of TS

3.1. Trigger mode of TS

In the ASOPS system, a data acquisition TS is required to start to scan each data. At present, there are two common techniques to generate ASOPS TSs: one is to convert the laser pulse generated by the master–slave laser into an electrical signal through a photodiode, which then generates a TS through a double-balanced mixer, and the other uses two-photon absorption to generate a TS. These two methods have the problems of unstable TSs and difficult adjustment of two-photon frequency doubling signal in the experimental process. Therefore, in this study, we design an all-fiber TS generation method based on the time overlapping interference signal generated by the collinear motion of two laser pulses. Compared with the double-balanced mixing detector, it effectively avoids the instability of the TS caused by the introduction of electrical signals many times and reduces the jitter by approximately 100 fs–200 fs. In addition, compared with the two-photon TS, the trigger mode of the proposed TS effectively avoids the problem that the frequency doubling signal is difficult to send out in the process of two-photon adjustment, thereby greatly simplifying the optical path design and facilitating the further integration of ASOPS system. Further, this provides a good premise for obtaining stable THz signals with high signal-to-noise ratios.

3.2. TS generation principle

The pump and probe femtosecond laser pulse are simplified into two monochromatic Gaussian pulses, $E_{\mathrm{m}}(t)$ and $E_{\mathrm{s}}(t)$, respectively,[18] *i.e.*,

$$E_{\mathrm{m}}(t) = E_{\mathrm{m}} \exp[-t^2/(2T_0^2)] \exp(-\mathrm{i}\omega t), \tag{6}$$

$$E_{\mathrm{s}}(t) = E_{\mathrm{s}} \exp[-(t-\tau)^2/(2T_0^2)] \exp[-\mathrm{i}\omega(t-\tau)], \tag{7}$$

where $E_{\mathrm{m}}(t)$ and $E_{\mathrm{s}}(t)$ each denote the peak intensity of the Gaussian pulsed electric field, T_0 is the full width at half maximum (FWHM), ω represents the angular frequency of the two Gaussian electric fields, and τ refers to the time delay of the

detection pulse relative to the pump pulse. The default initial phases of the two Gaussian electric fields are both zero, *i.e.*, $\varphi_1 = \varphi_2 = 0$.

The total intensity $I(\tau)$ of the two Gaussian pulses can be expressed as follows:

$$\begin{aligned} I(\tau) = {} & E_{\mathrm{m}}^2 \int_{-\infty}^{+\infty} \exp(-t^2/T_0^2)\,\mathrm{d}t \\ & + E_{\mathrm{s}}^2 \int_{-\infty}^{+\infty} \exp(-(t-\tau)^2/T_0^2)\,\mathrm{d}t \\ & + 2E_{\mathrm{m}}E_{\mathrm{s}}\cos(\omega\tau) \int_{-\infty}^{+\infty} \exp[(-t^2/2T_0^2) \\ & - (t-\tau)^2/2T_0^2]\,\mathrm{d}t. \end{aligned} \tag{8}$$

The FWHM of the femtosecond fiber laser pulse is 140 fs, whereas the photodiode response time of the balanced detector (BD) is greater than 2 ns. Therefore, the current output of the photodiode can be regarded as a simple Gaussian impulse response, and the peak amplitude is proportional to the laser pulse energy. By setting $E_{\mathrm{m}} = E_{\mathrm{s}} = 1$, initial $\tau = 0$, then $\tau \geq 10T_0$, equation (8) yields a ratio of 2:1 for the maximum optical overlapping pulse intensity compared with the simple summation of the temporally distinct laser pulses ($10T_0 \leq \tau \ll 2$ ns). Consequently, as long as there is a mechanism for removing the laser pulse sequence from the signal, the data acquisition TS can be generated from the optical temporal coincidence of laser pulses.[19]

3.3. Generation of TS

A schematic diagram of the TS generation is shown in Fig. 2. The entire system includes two fiber femtosecond lasers, fiber couplers, and BDs. The function of the BD is to convert the optical signal into an electrical signal and further amplify it. The repetition rates of the two femtosecond lasers are about $f_1 = 100$ MHz and $f_0 = 100$ MHz$-\Delta f$, respectively, where Δf is adjustable, and the adjustment range can vary from less than 1 Hz to 10 kHz. The power of the master and slave lasers are 120 mW and 135 mW, respectively. The pulse width and center wavelength of the two lasers are 75 fs and 1550 nm, respectively. A small part of the power of the master slave laser is sent to the polarization-maintaining fiber coupler, in which the optical interference signal is generated. The signal is input to two stacked reverse-biased photodiodes of the BD to convert the optical signal into an electrical signal. The BD can remove all simple summation signals; however, it cannot completely remove coherent interference signals. In addition, because there is always a certain deviation between the lengths of the two optical fibers input to the BD, the beat signal and TS generated by interference are obtained from the BD, and then the beat signal is filtered to obtain a stable TS. Figure 3 shows the stable beat signal and TS displayed on the oscilloscope, and figure 4 shows the pure TS.

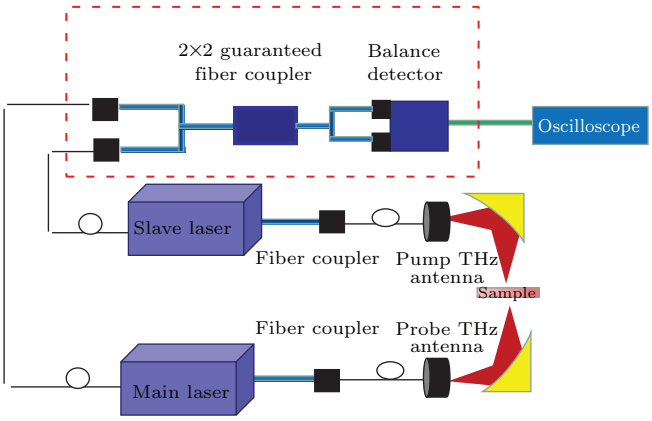


Fig. 2. Schematic diagram of TS generation.

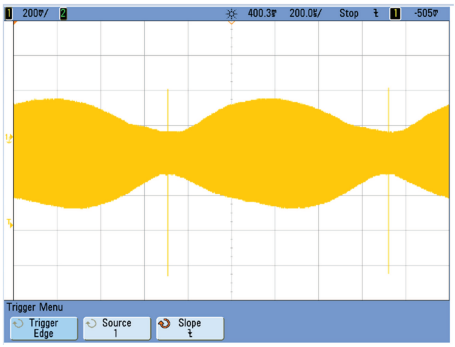


Fig. 3. Beat signal and TS displayed on oscilloscope.

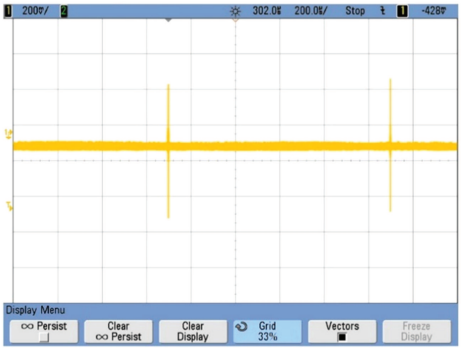


Fig. 4. TS after filtering beat signal.

4. Integration of traditional THz-TDS system and ASOPS system

In this study, we establish an integrated system of the traditional THz-TDS system and ASOPS system (as shown in Fig. 5).

The traditional THz-TDS system can provide a calibration scheme for the ASOPS system to further optimize the THz signal. The specific process is as follows. First, when the master laser works alone, it is a traditional THz-TDS system. A femtosecond pulsed laser is divided into two beams with equal power through a half-wave plate and a polarizing beam splitter prism. One is coupled into the first optical fiber photoconductive antenna as pump light through gold mirrors M_1 and M_2 and reaches optical fiber port coupler 1 for generating THz waves, and the other is coupled into the second op-

tical fiber photoconductive antenna through gold mirrors M_5, M_3, and M_4 and optical fiber port coupler 2 to detect THz waves. Finally, the obtained signal is amplified by a phase-locked amplifier and collected by using a computer. Then, when the master laser and slave laser work simultaneously, it is an ASOPS system. A femtosecond laser pulse of the master laser passes through the half-wave plate and polarization beam splitter prism. At this moment, the half-wave plate is adjusted to make its transmitted light intensity decrease to zero; the reflected light is coupled through gold mirrors M_1 and M_2, and then enters into optical fiber port coupler 1 and finally enters into the optical fiber photoconductive antenna to generate THz waves. A femtosecond pulse emitted from the slave laser is coupled into the optical fiber photoconductive antenna through optical fiber port coupler 3 to detect THz waves. Part of the femtosecond laser pulse of the master laser and slave laser enters into the fiber coupler to generate TS waves. Finally, the detected signal is amplified by the amplification circuit andrecorded by an oscilloscope.

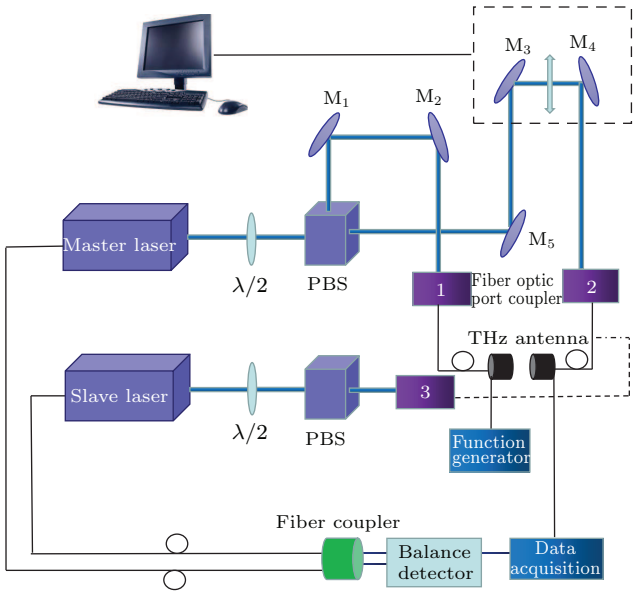


Fig. 5. Integrated ASOPS and traditional THz-TDS system.

5. Experimental results

Since the difference in frequency between the master laser and the slave laser of the ASOPS system is adjustable, we first study the TSs when the frequency difference is 50 Hz and 500 Hz respectively. The TSs recorded by the oscilloscope are shown in Figs. 6(a) and 6(b). It can be seen that the TSs under the emerging trigger mode is relatively stable and easy to implement, which further contributes to the miniaturization and integration of the system.

Figure 7(a) shows the THz time-domain spectrum obtained from the ASOPS system. We set Δf to be 2 Hz, and the temporal window of THz waveform is only 0.5 s. In addition, for the ASOPS system, the sampling interval is determined by $T_0 = 1/f_0$, so the value is about 1×10^{-8} s. To further verify the stability and accuracy of the system, the entire system is switched to the very mature traditional THz-TDS working mode based on a mechanical delay line. The sampling interval is determined by the step size, total moving distance of the delay line, and the temporal window of the THz waveform. In our experiment, these corresponding parameters are 0.01 mm, 10 cm, and 5 minutes respectively, so the sampling interval is 0.03 s. The frequency-domain spectra shown in Figs. 8(a) and 8(b) are obtained by the Fourier transform of the two THz time-domain spectra. We find that the FWHM of the time-domain spectra and the spectrum widths of the frequency-domain spectra obtained from the two systems are almost the same. However, the ASOPS system is dominant at the time of obtaining the spectrum.

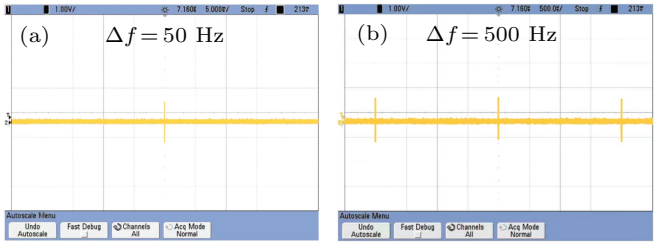


Fig. 6. TSs obtained under differences in frequency: (a) 50 Hz and (b) 500 Hz.

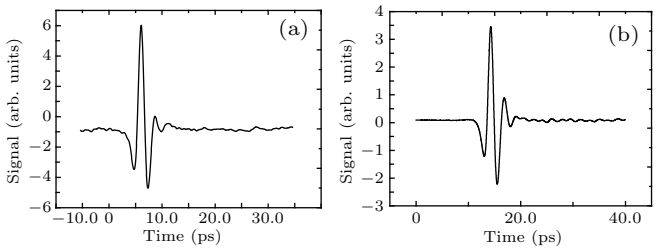


Fig. 7. THz time-domain spectra of (a) ASOPS system and (b) traditional THz-TDS system.

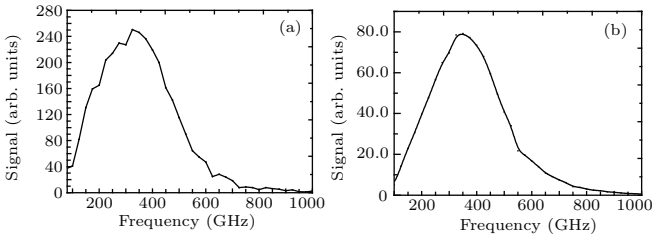


Fig. 8. THz spectra of (a) ASOPS system and (b) traditional THz-TDS system.

6. Conclusions

In this study, we established an integrated system of traditional THz-TDS and ASOPS systems. The traditional THz-TDS system can calibrate the optical path of the ASOPS system to ensure the accuracy of the collected signal. An all-fiber TS generation method based on the time overlapping interference signal generated by the collinear motion of two laser

pulses is adopted. Finally, the time-domain and frequency-domain spectra are obtained by using two independent systems in the integrated system. It is found that the FWHMs of the time domain spectrum and the spectral widths of the frequency domain spectrum of the two independent systems in the integrated system have similar spectral forms, but the sampling speed of the ASOPS system is significantly faster than that of the traditional THz-TDS system. The integrated system provides a reliable guarantee for the accurate study of the transient characteristics of substances, and has potential advantages in the study of molecular dynamics.

Acknowledgements

The authors would like to thank "Enago" for English polishing.

Project supported by the National Key Research and Development Program of China (Grant No. 2021YFB3200100) and the National Natural Science Foundation of China (Grant No. 61575131).

References

[1] Zhang Y, Liu S and Huang W 2022 *Chin. Phys. B* **31** 068702
[2] Piyush A, Lisen H, Sze T L and Ran J S 2022 *Nat. Commun.* **13** 4072
[3] Piyush A, Rohit M and Abhishek K 2021 *Adv. Funct. Materials* **31** 2010453
[4] Dieny B, Prejbeanu I L and Garello K 2020 *Nat. Electron.* **3** 446
[5] Piyush A, Yang Y S and James L 2022 *Appl. Phys. Lett.* **120** 161104
[6] Zhang Y, Zhang X and Li S 2016 *Sci. Rep.* **6** 26949
[7] Björn G, Roman J B D, Thorsten Göbel, Martin Schell, Werner Bohmeyer, Ralf Müller and Andreas Steiger 2015 *Opt. Lett.* **40** 3545
[8] Zhang Z, Liu Y and Zhu X 2021 *J. Phys.: Conf. Ser.* **1865** 022010
[9] Bartels A and Dekorsy T 2008 *Technisches Messen.* **1** 75
[10] Zhang H, Su B and Yang X 2018 *Rev. Sci. Instrum.* **89** 113108
[11] Puc U, Abina A and Rutar M 2015 *Appl. Opt.* **54** 4495
[12] Yasui T, Saneyoshi E and Araki T 2005 *Appl. Phys. Lett.* **87** 061101
[13] Bartels A, Thoma A and Janke C 2006 *IEEE Quantum Electronics and Laser Science Conference*, 21–26 May 2006, Long Beach, CA, USA
[14] Bartels A, Cerna R and Kistner C 2007 *Rev. Sci. Instrum.* **78** 2357
[15] Klatt G, Gebs R and Janke C 2009 *Opt. Express* **17** 22847
[16] Gebs R, Klatt G and Janke C 2010 *Opt. Express* **18** 5974
[17] Duan G T, Li L and Cui H L 2016 *Transactions of Beijing Institute of Technology* **36** 170
[18] Zhang H, Su B and Yang X 2018 *Rev. Sci. Instrum.* **89** 113108
[19] Yang X, Su B and Wu Y 2019 *Measurement Science and Technology* **30** 045204