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Unified description on principles of fourier transform infrared spectroscopy and terahertz time-domain spectroscopy

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ABSTRACT

Fourier transform and terahertz time-domain (TD) spectroscopic techniques are widely used in infrared and terahertz frequency bands, and they are usually considered as two different techniques. Here, however, we show that the two techniques are based on the same basic principle. In a Fourier transform infrared spectroscopy (FTIR), the infrared radiation from a continuous incoherent light source passes through a Michelson interferometer, and the interferogram as a function of optical path difference is recorded. The spectral information is decoded from the interferogram by using the Fourier transform process. In a terahertz TD spectrometer, the terahertz transient electrical pulse train from a femtosecond-laser-pumped photoconductive antenna (PCA) is sampled by another PCA (or an electro-optical crystal) gated by a probe femtosecond laser beam from the same femtosecond laser. In this paper, the principle of terahertz TD spectroscopy is described in Fourier domain. We show that the recorded TD terahertz "electrical waveform" can be regarded as an interferogram also. The FTIR and the terahertz TD spectroscopic techniques can be described within the same framework. The asynchronous optical sampling (dual comb) terahertz time-domain technique, a variant form of the traditional terahertz TD spectroscopic technique, can also be described by the same principle. Such a unified description on principles of FTIR and terahertz TD spectroscopies is useful for improving the performance of terahertz emitters and detectors, and developing new terahertz spectroscopic techniques based on femtosecond lasers.

1. Introduction

Linear response of matter to electromagnetic field is described by the complex refractive index$n = n^{'}(\omega, k) + in^{''}(\omega, k)$, where$\omega$is circular frequency, $k$is wavevector, and i is imaginary unit. The causality requires that the real part and the imaginary part of the refractive index satisfy the Kramers-Kronig relations [1]. In general, the momentum of electromagnetic field, $\hbar k$ with $\hbar$ the reduced Planck constant, is a small quantity and usually ignored in the field-matter interaction; then the refractive index is approximately expressed as$n = n^{'}(\omega) + in^{''}(\omega)$, which is known as long-wave approximation. There are various spectroscopic techniques to measure the refractive indexes of condensed matters, liquids, gases, and plasmas [2–6]. In visible and higher frequency electromagnetic bands, the different frequency components in a collimated light beam are spatially separated by dispersive devices (prisms and gratings, etc.). By rolling the dispersive devices or using linear detector arrays, the reflection and transmission spectra of samples are directly obtained, from which the refractive indexes are derived.

In infrared frequency regime, Fourier transform spectroscopic techniques are widely applied [2,3]; in comparison with the dispersive-type spectroscopies, Fourier transform infrared (FTIR) spectrometers have multiplex and throughput advantages. In a FTIR spectrometer, the spectral information of the collimated light beam from the radiation source is encoded into the interferogram from a two-arm interferometer as a function of the path difference between the two arms, and the spectrum is acquired by Fourier transforming the interferogram. However, in longer wavelength millimeter and terahertz spectral regimes, due to the decrease of spectral intensity of thermal sources, the signal to noise ratio and the multiplex advantage of FTIR systems are severely reduced [3–5].

Terahertz time-domain (TD) spectroscopy bridges the spectroscopic terahertz gap between the electrical and infrared regimes [4–7]. Because there are many low-energy excitations in semiconductors, metals, semiconductor quantum confined structures, superconductors, and biomacromolecules, terahertz TD spectroscopy is widely used to investigate the spectral fingerprints of the above materials [1,4–9]. In a traditional terahertz TD spectrometer, one collimated femtosecond

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optical pulse is divided into two parts by a beam splitter; one is named as the pump pulse, and the other is the gate pulse. The electrical pulse train from the terahertz source pumped by the pump pulse is sampled by the detector gated by the time-delayed gate pulse. Based on the effective time approximation, the whole terahertz electrical pulse is obtained in TD by continuously varying the delay time, and the spectrum is derived by Fourier transformation [4]. Biased photoconductive antennas (PCAs) or nonlinear electro-optical crystals (ZnTe, GaP, and $LiNbO_3$ etc.) are used as terahertz emitters in terahertz TD spectrometers, and zero-bias PCAs or free-space electro-optical sampling techniques are used to detect the field amplitude of the terahertz electrical pulse [9]. Here, we only consider the PCAs as the emitters and detectors in terahertz TD spectrometers for simplicity; the main conclusions are valid for the other emitters and detectors [9]. In recent years, various terahertz spectroscopic techniques based on femtosecond lasers, such as asynchronous optical sampling (ASOPS) technique [10–20], electronically optical sampling technique [21], optical sampling by cavity tuning technique [22], and single-shot detection [23], were developed as the variants of traditional terahertz TD spectroscopy.

The FTIR and terahertz TD spectroscopies are usually considered as two different techniques. The performance and characteristics of the two-type systems are systemically and directly compared by Han *et al.* [24] and Nuss *et al.* [4]. It has been shown that terahertz TD spectrometers have better performance at frequencies below 3.0 THz, but at frequencies over 5.0 THz, the opposite is true. In this paper, we first describe the principles of femtosecond-laser-pumped biased PCA emitter and femtosecond-laser-gated zero-biased PCA detector. In terahertz TD spectrometers, the PCA emitters are considered as terahertz electromagnetic (EM) combs, and the PCA detectors as terahertz photocarrier (PC) combs [9,20]. In this perspective, we show that the terahertz waveform obtained in a terahertz TD spectrometer is an interferogram, which has no difference from that measured in a FTIR spectrometer. The basic principles of terahertz TD and FTIR spectroscopies are exactly the same; the performance differences in different frequency regimes originate from the emitters and detectors used in the two-type systems. The principle of dual-comb spectroscopic technique is also considered in the same framework. We believe that the unified descriptions on principles of terahertz TD and dual-comb spectroscopies are equivalent to those described in TD framework. However, it is valuable to consider the principles of the terahertz spectroscopies in a new perspective for giving deeper understandings on these systems. Our results are helpful to improve the performance of the existing spectrometers, develop new terahertz spectroscopic techniques, and optimize the terahertz emitters and detectors used in these terahertz spectrometers.

2. Terahertz PCA emitters and detectors

The output optical field $E_O$from a femtosecond laser in Fourier space can be expressed as [25]

$$E_O = \sum_{m=-M}^{M_{max}} A_m \cos[2\pi(f_0 + mf_r)t + \phi_{CEP}], \tag{1}$$

where A_m is the amplitude of the *m-th* frequency component, M and M_{max} are the numbers of harmonic oscillations at frequencies below and above the carrier frequency f_0, respectively; f_r is the repetition frequency, and ϕ_{CEP} is the carrier-envelope phase. The EM field emitting from a femtosecond-laser-pumped PCA is [9]

$$E_{THz}(t) = \frac{\mu_0 w_0}{4\pi}\frac{\sin\theta}{r}\frac{d}{dt_r}[I_{pc}(t_r)]\hat{\theta} \propto \frac{dI_{pc}(t)}{dt} = \mu E_s \frac{dn_{pc}(t)}{dt}, \tag{2}$$

where μ_0 is the vacuum permeability, w_0 is the spot area, r is the radial distance from the radiation source,$\theta$is the azimuth angle, I_{pc} is the photon current, $t_r = t - r/c$ is the retardation time, μ is the electron mobility, E_s is the static bias electric field, n_{pc} is the photon-excited electron concentration. Because of $n_{pc} = \eta E_O E_O^*$ with η the absorption efficiency, in Fourier space the terahertz EM-field pulse E_{THz} composed of harmonic beat frequency components from the PCA can be written as

$$E_{THz}(t) = \sum_{m=N_{min}}^{N_{max}} B_m \cos(2\pi m f_r t), \tag{3}$$

where N_{min} and N_{max} are the minimum and maximum values of harmonic orders, respectively, and B_m is the amplitude of *m-th* harmonics. Due to the difference frequency process, the carrier-envelope phase ϕ_{CEP} is canceled, as shown in Eq. (3), which indicates that the frequencies of all the harmonics do not shift with time, and if the repetition frequency of pump femtosecond laser is locked to a radio frequency source with high stability, the frequency difference between every two neighboring harmonics does not vary with time. Therefore, the output of a femtosecond-laser-pumped PCA is considered as a terahertz EM comb.

The detection of terahertz field by using femtosecond-laser-gated PCAs with zero bias is based on the rectification effect of photo-excited electrons in the input terahertz field. The terahertz-field-induced current density in a PCA detector is [9,26]

$$j(t) = e\mu n_{pc}(t) E_{THz}(t), \tag{4}$$

where e is electron charge. The photon-excited electron concentration in a PCA detector can be expressed in a form similar with Eq. (3) [9,16,26],

$$n_{pc}(t) = \sum_{m=N_{min}}^{N_{max}} N_m \cos(2\pi m f_r t), \tag{5}$$

where N_m is the Fourier constant of the *m-th* harmonics. Due to the similarity between Eq. (3) and Eq. (5), a femtosecond-laser-gated PCA with zero bias is considered as a PC comb, which is a terahertz mixer with the frequency components of the PC comb as multi-frequency local oscillators.

3. Terahertz TD and FTIR spectroscopies

Fig. 1 shows the schematic diagrams of traditional terahertz TD (a) and FTIR (b) spectroscopies. In a terahertz TD spectrometer, the terahertz-field-induced current from a PCA detector can be obtained by inserting Eq. (3) and Eq. (5) into Eq. (4) and after some algebra,

$$\begin{aligned} j(t) \approx \frac{e\mu}{2}\sum_{m=M_0}^{M}\eta_m B_m \Bigg[& N_m \cos\left(\frac{2\pi m f_r L}{c} + \phi(mf_r)\right) + N_{m+1}\cos \\ & \left(2\pi f_r t - \frac{2\pi m f_r L}{c} - \phi(mf_r)\right) + N_{m-1}\cos \\ & \left(2\pi f_r t + \frac{2\pi m f_r L}{c} + \phi(mf_r)\right) + higherorderitems\Bigg], \end{aligned} \tag{6}$$

where η_m is the rectification coefficient, L is the optical path difference between the pump and gate pulses, $\phi(mf_r)$ is the phase shift introduced by the sample under measurement, and c is the speed of light in vacuum.

Eq. (6) indicates that if the optical path difference is fixed, all frequency components overlap, and the terahertz spectral information cannot be derived from the photocurrent without considering all the harmonic components. However, by sweeping the optical delay line, the zero- and the first-order harmonic components (Fig. 2) of photocurrent from the PCA detector are

$$j_0(L) = \frac{e\mu}{2}\sum_{m=M_0}^{M}\eta_m B_m N_m \cos\left[\frac{2\pi m f_r L}{c} + \phi(mf_r)\right], \tag{7a}$$

$$j_1(L, t) \approx e\mu\cos(2\pi f_r t)\sum_{m=M_0}^{M}\eta_m B_m N_m \cos\left[\frac{2\pi m f_r L}{c} + \phi(mf_r)\right]. \tag{7b}$$

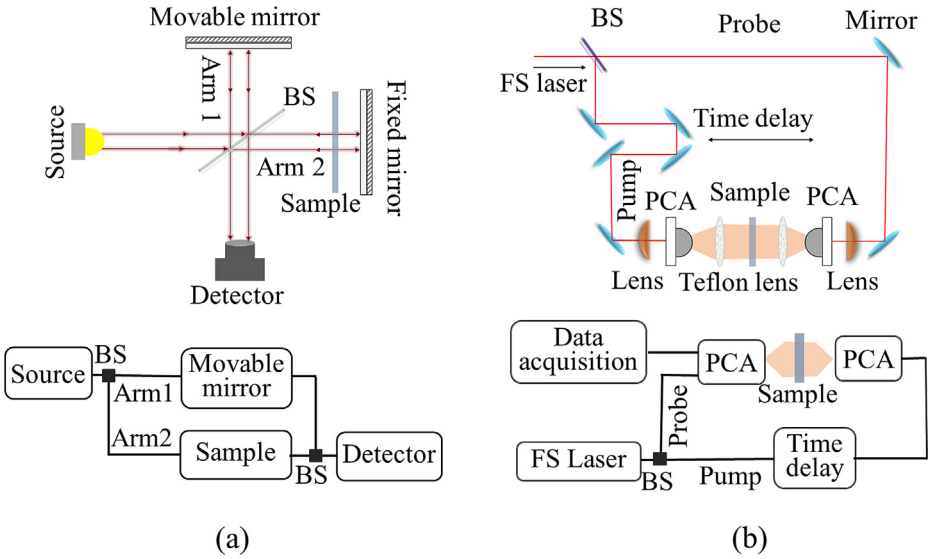

Fig. 1. Schematic diagram of principles of FTIR spectroscopy (a) and terahertz TD spectroscopy (b). BS: beam splitter, FS: femtosecond.

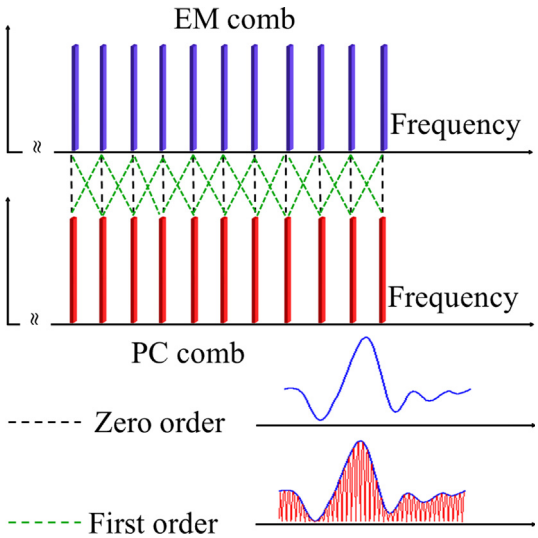

Fig. 2. Multi-frequency heterodyne detection of terahertz interferogram with a PCA detector (PC comb) in a conventional terahertz TD spectrometer.

In Eq. (7b), we make an assumption of$N_{m-1} = N_m = N_{m+1}$; the frequency difference between two neighboring comb tips is set to 100 MHz which is 10^4 smaller than the full response bandwidth of about 3.0 THz of PCA detectors, and the error introduced by such an assumption is neglectable. In a dispersive FTIR spectrometer (Fig. 1b, the sample being in one of the two arms of interferometer), the interferogram is expressed as [2,3]

$$I(L) = 2\int_0^\infty \sqrt{p_1(\nu)p_2(\nu)}\cos[2\pi\nu L + \phi(\nu)]d\nu, \tag{8}$$

where ν, $p_1(\nu)$, and $p_2(\nu)$ are frequency in unit of wavenumber (cm^{-1}), field amplitudes of the first and second arms of interferometer, respectively. Due to the similarity of Eqs. (7a) and Eq. (8), we conclude that the "TD waveform" of terahertz TD spectroscopy is an interferogram, which is exactly the same with that of FTIR spectroscopy. In both cases, the spectral information is encoded into the additional phase induced by optical delay line or interferometer, and it is recovered by Fourier transforming the interferograms (Eqs. (7a) and Eq. (8)).

Usually, for a terahertz TD spectrometer, the zero-order interferogram (Eq. (7a)) is recorded to acquire terahertz spectrum. In order to improve the signal to noise ratio, the pump femtosecond laser pulse needs to be modulated by a mechanical chopper or other type modulators, and the modulated signal is amplified by a lock-in amplifier. In comparison with the zero-order interferogram (Eq. (7a)), there is an additional factor $\cos(2\pi f_r t)$in the first-order interferogram (Eq. (7b)), which indicates that the first-order interferogram is inherently modulated with the modulation frequency f_r. With the progress of digital lock-in amplifier technique, the bandwidth of lock-in amplifier can cover the repetition frequency f_r of femtosecond laser easily. Therefore, if the repetition frequency f_r is stabilized, we can record the first-order interferogram to acquire the spectral information by using a wide bandwidth lock-in amplifier without adding an extra modulator to the system. Moreover, with the increase of modulation frequency to ~100 MHz, the flicker noise is substantially suppressed.

Eqs. (7) and (8) show that the same coding scheme is utilized to obtain the spectral information for the terahertz TD spectroscopy and the FTIR spectroscopy. Therefore, the unique characteristics owned by each of the two spectroscopic techniques originate from, to a great extent, the differences of the emitters and detectors. In terahertz regime of 0.1–5.0 THz, compared to the FTIR spectroscopic technique, the terahertz TD spectroscopy shows some advantages. Firstly, in a FTIR spectrometer, because the power of thermal blackbody emitters or high-pressure gas-discharge emitters decreases rapidly with frequency, at frequencies of below 5.0 THz, the performance of FTIR spectroscopy becomes poor. The average output power of femtosecond-laser-pumped emitters used in a terahertz TD spectrometer is at least one order larger than that of the incoherent sources used in a FTIR spectrometer. Secondly, due to the limitation of spatial coherent length of radiation from the incoherent emitters, a smaller aperture is required to reach a higher frequency resolution, which will reduce the system signal to noise ratio further for FTIR spectroscopy. However, in a terahertz TD spectrometer, due to the long spatial coherent length of laser pulse, no apertures are needed, and the system performance is insensitive to the increase of optical path difference. Thirdly, a 1:1 beam-splitter is very important for the FTIR spectroscopy. However, there are no such severely restrictions for beam-splitters used in terahertz TD spectrometers. Lastly, it is very easy to make time-resolved measurements with terahertz TD spectrometers, but it is not a trivial task to do the same measurements with traditional FTIR spectrometers.

4. Dual-comb terahertz spectroscopy

The dual-comb (or ASOPS) terahertz TD spectroscopy technique

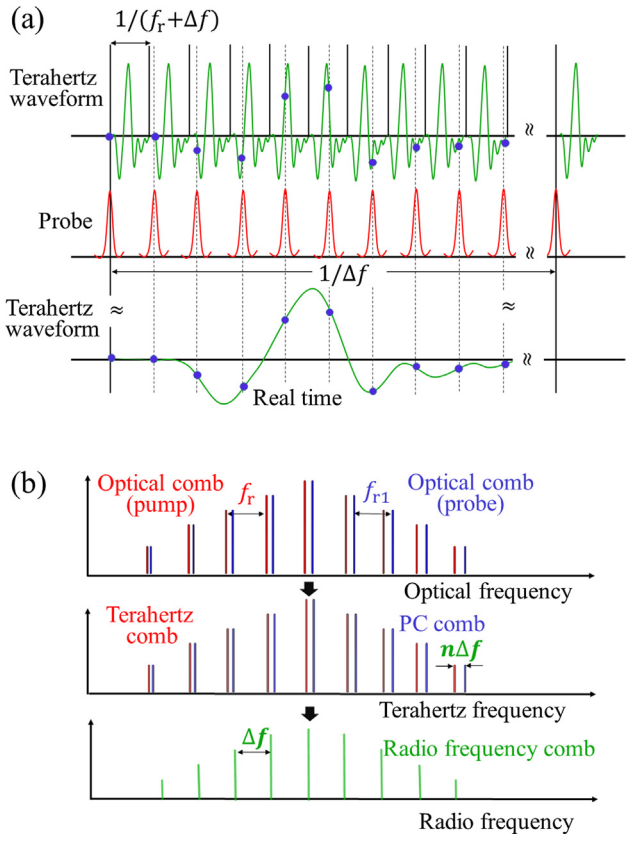

Fig. 3. Schematic diagram of principles of dual-comb spectroscopy in TD (a) and in Fourier domain (b).

show some advantages compared to the traditional TD spectroscopy with mechanical time delay line [16,26]. The schematic diagram of principle of the dual-comb terahertz spectroscopy in TD and frequency domain is shown in Fig. 3. Two repetition-frequency-locked and synchronized femtosecond lasers with repetition frequency difference of $\Delta f = f_{r1} - f_r$ are utilized to implement the ASOPS procedure, where f_r and f_{r1} is the repetition frequencies of pump and probe femtosecond pulses, respectively; one laser is used to pump a biased PCA to emit terahertz pulse train, and the other laser is used to gate an unbiased PCA detector to sample the terahertz pulse. In TD (Fig. 3(a)), the sampling time period is$T = 1/\Delta f$ that corresponds to a real delay time$\tau = 1/f_r$; in this time period, the number of equidistant sampling points is$f_r/\Delta f$, and the temporal interval between two neighboring sampling points is$\Delta t = \Delta f/f_r^2$. The bandwidth and frequency resolution of the terahertz spectra acquired with the ASOPS TD spectroscopy are$f_r^2/(2\Delta f)$and $1/\tau$, respectively. In frequency domain, the ASOPS is equivalent to a multi-frequency heterodyne process as shown in Fig. 3(b).

In a dual-comb TD spectroscopic system, similar with Eq. (7), the zero- and first-order output heterodyne signals locating in low frequency regime for the PCA detector (PC-comb) are

$$j_0(t) = \frac{e\mu}{2} \sum_{m=M_0}^{M} \eta_m B_m N_m \cos[2\pi m \Delta f t + \phi(mf_r)], \tag{9a}$$

$$j_1(t) \approx e\mu \cos(2\pi f_{r1} t) \sum_{m=M_0}^{M} \eta_m B_m N_m \cos[2\pi m \Delta f t + \phi(mf_r)], \tag{9b}$$

where $\phi(mf_r)$ is the sum of sample-induced additional phase and the fixed phase difference between the two lasers. As in Eq. (7b), we also make the assumption of$N_{m-1} = N_m = N_{m+1}$. Eq. (9) indicates that the 'terahertz waveform' measured in an ASOPS TD spectrometer can be considered as an interferogram too. Eqs. (7)–(9) clearly show that the FTIR, conventional and ASOPS terahertz TD spectroscopies are based on the same principle. However, there is a difference in signal detection. For FTIR and conventional terahertz TD spectrometers, signals are obtained through a homodyne detection process, and the spectral information is encoded in the DC signals by phase modulations. For an ASOPS spectrometer, a heterodyne detection is utilized to obtain the beat-frequency signals, from which the spectral information can be recovered.

5. Discussion

In general, they are equivalent to each other for considering the principles of FTIR and terahertz TD spectroscopic techniques in time-domain and Fourier domain. However, in some situations, more insights on the principle of terahertz TD spectroscopy can be given by considering the concerned problems in Fourier domain.

Phase error plays a seriously negative role on the performance of terahertz TD spectroscopy in a complex way [27–29]. As shown in Eq. (7), the phase error does not only originate from the delay line misplacement L, but also from the variation of repetition frequency f_r of the femtosecond laser. Moreover, the phase error is more sensitive to the above two factors in high frequency regime (larger value of multiplication factor, m). However, in TD, the variation of repetition frequency is generally not considered when analyzing the origins of phase error, because in a conventional terahertz TD spectrometer, the pump pulse and the gate pulse are from the same mother pulse, and the relative time delay between the pump pulse and the gate pulse does not change.

In the Fourier domain, we note that the terahertz spectral information is encoded into different harmonics of repetition frequency (mf_r, $m = 0, 1, 2\ldots$, the first two items being presented in Eqs. (7) and (9)). For example, for $m = 1$, the interferogram is modulated with frequency f_r (Eqs. (7b) and (9b)). Therefore, the modulated data of interferogram (modulation frequency, f_r) can be acquired by using a lock-in amplifier without introducing an extra modulator to the system. In comparison with the case of $m = 0$, it is expected that a higher signal to noise ratio can be achieved when the data acquisition is implemented at the modulation frequency of f_r, because at such a high modulation frequency the flicker noise is effectively suppressed. In addition, in Fourier domain, a femtosecond-laser-pumped (gated) PCA emitter (detector) is considered as an electromagnetic comb (photon-carrier comb), and the detection of terahertz field amplitude is considered as a heterodyne process. In this point of view, it is possible to design and optimize the PCA emitters and detectors with higher performances.

6. Conclusions

In conclusion, the basic principles of FTIR spectroscopy and terahertz TD spectroscopy are compared systemically. In a terahertz TD spectrometer, the femtosecond-laser-pumped PCA emitter is taken as an EM-comb, and the femtosecond-laser-gated PCA detector as a PC comb in Fourier domain; the detection of terahertz field by the PC comb is described by a multi-frequency heterodyne process. The "terahertz waveform" recorded by the PC-comb used in the conventional (including dual-comb) TD spectrometer is an interferogram. Therefore, The FTIR spectroscopy and the terahertz TD spectroscopy are based on the same principle. Despite the equivalence to each other between the time domain and the Fourier domain, it is possible to gain deeper understanding to the characteristics of the terahertz TD spectroscopy in Fourier domain. Firstly, we find that the spectral information is modulated to different orders (0 is the lowest order) of the laser repetition frequency. Except for the zero order, it is possible to acquire the interferogram data at other orders by using the lock-in technique without introducing extra modulators to the system. Further, the flicker noise dominated in low frequency is effectively suppressed under a high modulation frequency. Secondly, in Fourier domain, it is clear that the

phase error does not originate from the misplacement of delay line, but also from the variation of repetition frequency; and the phase error is more sensitive to the above two factors at high frequency. We believe that our work is useful for improving the performance of terahertz emitters and detectors, and developing new terahertz spectroscopies based on femtosecond laser.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Appendix A. Supplementary material

Supplementary data to this article can be found online at https://doi.org/10.1016/j.infrared.2019.06.005.

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