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Limit of Spectral Resolution in Terahertz Time-Domain Spectroscopy *

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The pulsed nature of terahertz time-domain spectroscopy (THz-TDS) sets a fundamental limit on its spectral resolution. The spectral resolution of THz-TDS can be improved by increasing the duration of the temporal measurement, but is limited by the dynamic range of the system in the time domain. This paper presents calculations and experimental results relating the temporal dynamic range of a THz-TDS system to its spectral resolution. We discuss three typical terahertz sources in terms of their dynamic range and hence achievable spectral resolution.

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Terahertz time-domain spectroscopy (THz-TDS)[1,2] is an innovative sensing and imaging technology that provides information unavailable through conventional methods, such as microwave and x-ray techniques. The THz wave technology will impact communications, imaging, medical diagnosis, health monitoring, environmental control and chemical and biological identification.[3,4] To optimize THz-TDS, efforts have been made to extend the bandwidth[5,6] and to improve the signal-to-noise ratio (SNR) of the system.[7,8] The third desirable characteristic is a high spectral resolution, which is critical for observing resonances with narrow line widths, associated with distinct energy transitions at THz frequency range.

To fully exploit the potential of THz-TDS there must be a thorough understanding of the limits of its spectral resolution. The THz spectrum is calculated in THz-TDS by numerical Fourier transformation of the measured temporal waveform. From Fourier theory, a higher spectral resolution is obtained by extending the temporal measurement window, which corresponds to the scanning distance. The spectral resolution in Fourier transformation spectroscopy is fundamentally limited by the duration of the time-domain measurement, as expressed in Heisenberg's uncertainty principle. The other major technical limitation on the spectral resolution of THz-TDS is the repetition rate of the laser source used to generate and detect THz pulses. The longest time duration that can be sampled is equal to the time between consecutive pulses. These two limitations on spectral resolution can be improved upon by: (i) using alternative time scanning methods to replace distance scanning, and (ii) using a lower repetition rate laser amplifier rather than higher repetition rate laser oscillator. In this Letter, we discuss how the pulsed nature of time-domain spectroscopy raises another fundamental limitation. We also discuss the relationship between the temporal dynamic range and spectral resolution, and demonstrate the calculated and measured dynamic range versus temporal scanning range.

In THz-TDS, the temporal waveform of the THz pulse $S(t)$ is sampled at temporal intervals of δt over a range of T. The noise and interference with THz-TDS have been previously discussed.[10] Instead of discussing the physical properties of the noise, we focus on its performance in the THz frequency domain. The sampled waveform $S(t)$ includes the electric field of THz radiation $E_{\rm THz}(t)$, noise due to fluctuations in the THz field $N_{\rm THz}(t)$ and background noise from the THz detection (probe) beam $N_{\rm B}(t)$. $N_{\rm THz}(t)$ is presented by $N_{\rm THz}(t) = R(t)E_{\rm THz}(t)$, where $R(t)$ is a dimensionless random ratio.

If THz-TDS is stable over the measurement period, both $R(t)$ and $N_{\rm B}(t)$ are considered stationary, ergodic random data. $R(t)$ and $N_{\rm B}(t)$ have a mean of 0, and a standard deviation of σ_R and $\sigma_{\rm B}$ respectively. The standard deviation is defined as the square root of the sum of $(R(t))^2$ or $(N_{\rm B}(t))^2$ respectively, and represents the noise level in the measurement. Additionally, as they are fluctuations at different sampling points, to simplify the discussion and without affect the result we assume that both $R(t)$ and $N_{\rm B}(t)$ have no frequency dependence in the range of 0 to $\pi/\delta t$. $S = 1/\sigma_R$ and $D = A/s_{\rm B}$ are defined as the temporal measurement SNR and dynamic range respectively, where A is the maximum amplitude of THz field in time domain. The spectrum of the THz radiation is extracted from its temporal waveform in the period of 0 to T by Fourier transformation. The spectral resolution of THz-TDS is improved by long temporal scanning, but is limited by its dynamic range in frequency domain.

In a typical THz-TDS experiment, the temporal waveform of the THz pulse concentrates within a time interval (0 to T') and its spectrum has a finite band-

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width (0 to Ω'). The spectrum of THz radiation $E(\omega)$ will not change with increasing T or the decrease δt as $T > T'$ and $\Omega = \pi/\delta t > \Omega'$. In other words, the integration of $E(t)$ and $N_{\mathrm{THz}}(t)$ from 0 to T leads to the same result when integrating them from minus infinity to plus infinity.

The noise related to the THz field described in the frequency domain $N_{\mathrm{THz}}(\omega)$ is presented by a cross correlation of the spectrum of the THz field $E(\omega)$ and the spectrum of the random ratio $R(\omega)$. When $\Omega \gg \Omega'$ is realized in the measurement, $N_{\mathrm{THz}}(\omega)$ will be a frequency independent quantity within the range of 0 to Ω', because $R(\omega)$ is independent of frequency. The power of R should be the same when it is described in time-domain as well as in frequency domain, then we have

$$\rho_{\mathrm{THz}} = \sqrt{\frac{\delta t}{2\pi}}\sigma_R A\kappa, \tag{1}$$

where κ is defined by $\kappa = A^{-1}\sqrt{\int_{-\infty}^{+\infty}|E(t)|^2\mathrm{d}t}$. The background noise can be considered as $N_{\mathrm{B}}(t)$ times of a rectangular function ($X(t) = 1$, $0 < t < T$; otherwise $X(t) = 0$), so that the background noise level in the frequency domain is

$$\rho_{\mathrm{B}} = \sqrt{\frac{T\delta t}{2\pi}}\sigma_{\mathrm{B}}. \tag{2}$$

Here we note that unlike ρ_{THz}, which does not relate to the measurement duration, the background noise level ρ_{B} is proportional to the square root of the measurement duration, and will increase for longer temporal measurements. On the other hand, when T is fixed, both ρ_{THz} and ρ_{B} are proportional to the square root of the temporal resolution δt. As a result, a higher temporal resolution reduces the noise in the frequency domain.

The dynamic range of the THz time-domain spectrometer in the frequency domain $D(\omega)$ is determined by the spectrum of THz radiation over the noise level, which includes the noise arising from the THz field and background noise. $D(\omega)$ is then expressed by

$$D(\omega) = \frac{k(\omega)}{\sqrt{\frac{\delta t}{2\pi}\left(\frac{\kappa}{S}\right)^2 + \frac{T\delta t}{2\pi}\frac{1}{D^2}}}, \tag{3}$$

where $k(\omega)$ is a factor that depends on the actual THz waveform and is defined by $k(\omega) = E(\omega)/A_{\mathrm{THz}}$ (see Table 1). In a typical THz-TDS experiment with $S \sim 100$ to 1000, D can have a value of 10^5. When a temporal measurement duration only slightly longer than $T'T$ is used in TDS, the noise $N_{\mathrm{THz}}(t)$ related to the THz field dominates, the background noise $N_{\mathrm{B}}(t)$ can be ignored. In this case, the dynamic range of the spectrometer is linearly related to the SNR of the temporal measurement, which limits the maximum dynamic range of the THz spectrum, and the spectral resolution of the THz spectrometer is independent of the SNR of the temporal measurement.

Table 1. $E(t)$, $k(\omega)$ and κ of THz pulses with different waveforms for three sources of pulsed THz radiation.

Source form	Temporal waveform	$k(\omega)$	κ^2
Photoconductive dipole antenna Bi-polar	$2A\frac{t}{\tau^2}\mathrm{e}^{-t^2/\tau^2}$	$\frac{\omega\tau}{\sqrt{2}}\mathrm{e}^{-\omega^2\tau^2/4}$	$\frac{\sqrt{2\pi}}{2\tau}$
Surface field single-polar	$\frac{2A}{\tau^2}\mathrm{e}^{-t^2/\tau^2} - 4A\frac{t^2}{\tau^4}\mathrm{e}^{-t^2/\tau^2}$	$\frac{\omega^2\tau}{\sqrt{2}}\mathrm{e}^{-\omega^2\tau^2/4}$	$\frac{3\sqrt{2\pi}}{2\tau^3}$
Optical rectification	$A\sin(\omega_0 t)\mathrm{e}^{-a\|t\|}$ $t > 0$	$\sqrt{\frac{\omega_0^2}{[a^2+(\omega_0-\omega)^2][a^2+(\omega_0+\omega)^2]}}$	$\frac{\omega_0^2}{4a(a^2+\omega_0^2)}$
Decayed oscillation	0 $t < 0$		

However, when a longer measurement duration is set with the intention of increasing spectral resolution, the background noise $N_{\mathrm{B}}(t)$ dominates the THz electric field noise $N_{\mathrm{THz}}(t)$. As a result, the dynamic range of THz-TDS in the frequency domain is proportional to the temporal measurement dynamic range and inversely proportional to the square root of the temporal measurement duration. A longer temporal measurement leads to a smaller dynamic range in the frequency domain. Hence the longest achievable temporal measurement duration of THz-TDS is reached when the dynamic range in the frequency domain reaches the basic requirement in the experiment. Consequently, the finite temporal measurement dynamic range limits the spectral resolution of THz spectrometers.

We note that, regardless of the total temporal measurement duration, a higher temporal resolution enhances the dynamic range in the frequency domain by smoothing the data over more sampling points.

To experimentally demonstrate the relationship between the dynamic range in the frequency domain and temporal scanning range, we record a THz waveform generated from a 2-mm-thick ZnTe crystal using a femtosecond laser.[11] The emitted THz wave is collimated and focused by two parabolic mirrors and detected using electro-optic (EO) sampling by a 5-mm-thick ZnTe crystal.[12] The entire THz beam path is purged by dry air and the humidity is limited to less than 6%; the water vapour absorption will not be significant in our experiment. The delay stage resolution in the experiment is 10 μm (66.7 fs) and the temporal measurement dynamic range is 1000. A series of THz spectra are extracted by the Fourier transform of the temporal waveform in the range of 0 to $2^n T_0$, where $T_0 = 8.53\,\text{ps}$ and $n = 0, 1, \cdots\cdots 11$. The dynamic range at the frequency with the maximum amplitude as a function of temporal scanning range is presented in Fig. 1. The solid dots are experimental data, and the curve is fitted by Eq. (3). Temporal resolutions, rather than 66.7 fs, are used in the experiment where the SNR in the frequency domain is inversely proportional to the square root of the temporal resolution.

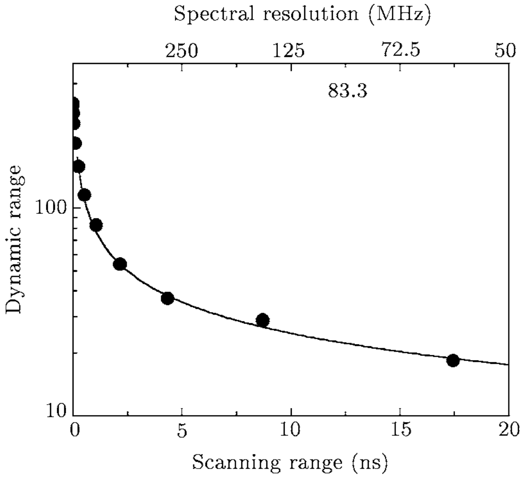

Fig. 1. Dynamic range of THz-TDS (central frequency) according to the temporal scanning range. Dots: experimental result; solid line: calculated result from Eq. (1).

If a sample has absorption of $M(\omega)$ for the THz spectrum, the required dynamic range of the THz spectrometer should be $1/M(\omega)$. Then the highest spectral resolution of the THz spectrometer is extracted from Eq. (3) by $D(\omega) = 1/M(\omega)$ and $\delta\nu = 1/T$:

$$\delta\nu \geq \frac{1}{2\Omega}\left(\frac{1}{M(\omega)k(\omega)D}\right)^2. \tag{4}$$

If $2\Omega = 10\,\text{THz}$, $k(\omega) = 1\,\text{ps}$, $M(\omega) = 1\%$ and $D = 10^5$ are applied to Eq. (4), the highest spectral resolution of the THz spectrometer is 0.1 MHz.

To understand the relationship between the spectral resolution of the THz spectrometer and its temporal measurement dynamic range, $k(\omega)$ and κ for the actual THz pulse are discussed, where $k(\omega)$ and κ can be extracted from the temporal waveform of the THz pulse, which depends mainly on the generation and detection mechanism and varies from one THz spectrometer to another. THz waveforms may be simulated by using several simple formulas. Currently, THz radiation for most TDS systems is generated by photo-switching[13] or optical rectification.[11] The temporal waveform of the THz pulse generated by photo-switching may be described as either bipolar (with an outer electrical field) or unipolar (with an inner electrical field). The temporal waveform of the THz pulse generated by optical rectification is considered as a damped oscillation. Here $k(\omega)$ and κ of the three most widely used THz sources are presented in Table 1.

In summary, we have demonstrated that the spectral resolution of THz-TDS depends not only on the duration of the sampled time-domain waveform $S(t)$, but also on the dynamic range of the THz spectrometer. A high-dynamic range system is required for high-resolution spectroscopy.

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