

Dynamic range and numerical error propagation in terahertz time-domain spectroscopy

Bernd M. Fischer, Matthias Hoffmann, and Peter Uhd Jepsen

*Freiburg Materials Research Center, University of Freiburg, Stefan-Meier-Strasse 21, D-79104 Freiburg, Germany
Bernd.fischer@physik.uni-freiburg.de, matthias.hoffmann@physik.uni-freiburg.de, jepsen@uni-freiburg.de*

Abstract: We discuss the influence of electronic noise and laser fluctuations on the largest detectable absorption and data accuracy in terahertz time-domain spectroscopy. Error propagation from the time domain to the frequency domain is discussed.

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1. Introduction

In the recent years the technique of static and transient terahertz time-domain spectroscopy (THz-TDS) has developed into a versatile spectroscopic technique for characterization of the complete dielectric function of a wide range of materials in the far-infrared spectral region. The technique has found application in a wide range of disciplines [1-7].

With an increasing amount of THz spectra published in the literature and collected in databases it becomes more and more important to address issues of data reproducibility and sources of error in order to avoid misinterpretation of experimentally obtained data.

In this presentation we describe quantitatively and in a very fundamental way the limits of the measurable absorption coefficient in transmission- and reflection THz-TDS. The analysis is based on the determination of the signal-to-noise ratio and dynamic range of a THz-TDS experiment. Our considerations show that interpretation of data from transmission THz-TDS especially on the high-frequency wing of the spectrometer bandwidth requires caution. This limitation has earlier been recognized in a qualitative way by Walther *et al.* [8].

2. Dynamic range in THz-TDS transmission and reflection measurements

The dielectric function of a material is in transmission THz-TDS determined by measuring a reference pulse $E_{\text{ref}}(t)$ propagating through the empty spectrometer and a sample pulse $E_{\text{sam}}(t)$ propagating through the spectrometer with the sample of thickness d placed in the beam path. These two pulses are transformed to the frequency domain, and the amplitude $A(\nu)$ and phase $\phi(\nu)$ of the ratio of the two spectra $E_{\text{sam}}(\nu)/E_{\text{ref}}(\nu)$ is calculated and analyzed to yield the spectra of the absorption coefficient $\alpha(\nu)$ and index of refraction $n(\nu)$.

Due to the typical single-cycle nature of a THz pulse its frequency spectrum extends from the low GHz region to several THz. At high frequencies the spectrum is characterized by a gradual roll-off, until the detected signal level approaches that of the noise floor of the experiment. The noise floor is independent of frequency, and corresponds to the spectrum recorded with a completely blocked THz beam path. The origin of this noise is of electronic nature, whereas fluctuations of the THz signal itself are mainly caused by laser intensity fluctuations.

In panels (a) and (b) of Fig. 1 a THz pulse is shown together with its frequency spectrum, normalized to the noise floor of the experiment. This normalized spectrum is a good measure of the frequency-dependent dynamic range (DR) of the experiment. The largest absorption coefficient that can be measured reliably with a given DR corresponds to the situation where the sample signal is attenuated to a level equal to the noise floor,

$$\alpha_{\text{max}} d = 2 \ln \left(DR \frac{4n}{(n+1)^2} \right) . \quad (1)$$

In panel (c) of Fig. 1 we show the frequency dependence of $\alpha_{\text{max}} d$ in a transmission experiment based on the THz pulse in panel (a) of Fig. 1, assuming that the sample under investigation has an index of refraction $n = 3$. At the peak of the spectrum we can measure values of αd up to 14, corresponding to an absorption coefficient of 280 cm^{-1} in a 0.5-mm thick sample. The dynamic range of αd decreases in an almost linear fashion towards higher

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frequencies, until the cutoff frequency, which in this case 4.6 THz. Above this frequency the apparent absorption coefficient will always be zero.

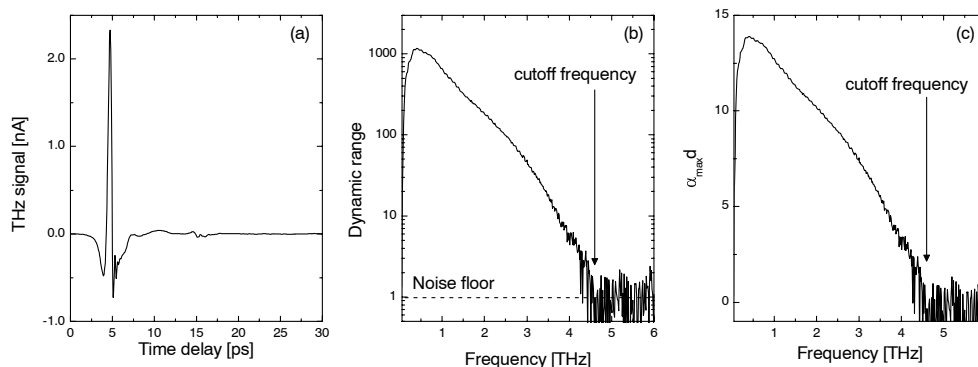


Fig. 1. (a) A typical THz pulse and (b) its corresponding frequency spectrum relative to the electronic noise floor of the experiment. The upper limit of the detectable absorption with this pulse is shown in (c).

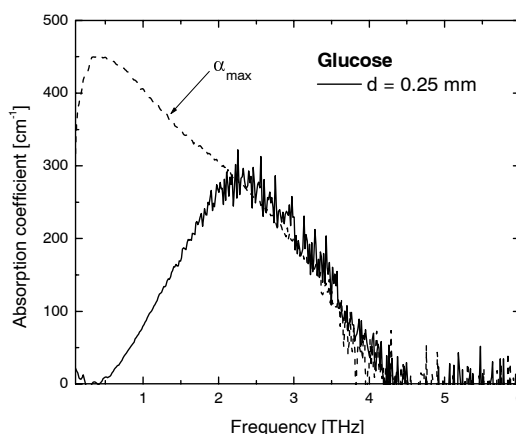


Fig. 2. Measured absorption spectrum of amorphous glucose (solid line). The dashed line indicates the dynamic range of the measurement.

In Fig. 2 a measurement of the absorption spectrum in the range 0.1-5 THz of amorphous glucose is shown. The sample was prepared by melting of polycrystalline glucose followed by rapid cooling of the sample material between two cold glass plates. This rapid cooling inhibits recrystallization of the sugar. The absorption spectrum recorded on the 0.25-mm thick sample is dominated by a broad peak centered at 2.2 THz. In the same graph $\alpha_{\max} d$ is shown. Comparison between the two curves makes it clear that the absorption measurement is valid for frequencies below 2.2 THz. In this example the quality of the absorption data is so that without a quantitative analysis of the dynamic range of the spectrometer it is not obvious that the absorption coefficient above 2.2 THz is not valid.

A method to circumvent the limitations of THz-TDS described above is to operate the THz spectrometer in the reflection mode. In this case the reference signal typically consists of the THz pulse reflected on a silver mirror surface with a reflection coefficient very close to minus unity. The sample signal consists of the THz pulse reflected from the plane sample surface, positioned at the same plane as the reference surface [1]. Alternatively, the reference- and sample signals are obtained as the reflections from the front- and backside of a thick window in front of the sample [9]. The reflection coefficient of absorbing samples is complex. Hence the recorded phase shift at the sample interface can be used to extract the absorption coefficient. In this case the largest detectable absorption coefficient is determined by the fluctuations of the laser intensity rather than the dynamic range of the experiment, thereby

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circumventing the limitations posed on a transmission experiment. THz-TDS in the reflection geometry has been used to characterize strongly doped semiconductors [1], polar liquids [9], and fundamental phonon resonances in ionic crystals [10].

3. Error propagation from the time domain to the frequency domain

The accuracy of the absorption coefficient and index of refraction determined in a THz-TDS measurement is given by the noise present in the experiment. The most straightforward way to consider the influence of this noise is to carry out several measurements and perform a statistical analysis of the result. As discussed above the dynamic range of the experiment is determined by electronic noise. The dynamic range can be increased by repetitive measurements and averaging of the time-domain signals *before* further processing. Averaging of signals after transformation to the frequency domain will not increase the dynamic range of the experiment. This procedure raises the question of how to propagate the experimental noise, or standard deviation of the measurement to the frequency domain.

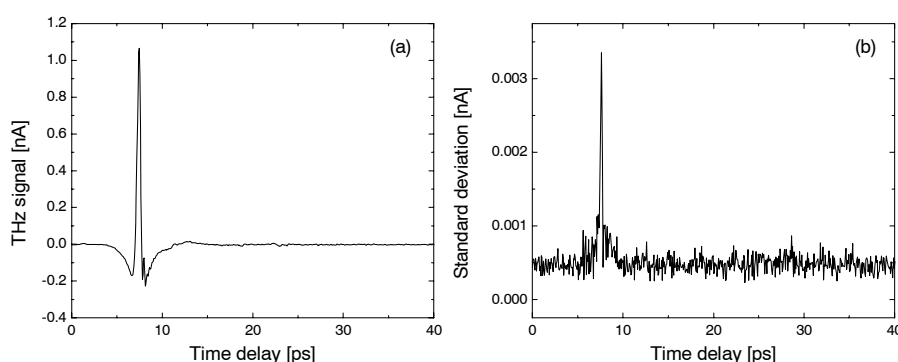


Fig. 3. (a) A time trace of a typical THz pulse, obtained by averaging of 8 individual traces. (b) The time trace of the standard deviation of the THz pulse in (a).

In part (a) of Fig. 3 we show a THz pulse obtained by averaging of 8 individual recorded time traces. In part (b) of Fig. 3 the corresponding standard deviation of the averaged signal is shown. The standard deviation of the trace is constant except in the region where the THz signal is present. This illustrates the difference between the electronic noise and the noise introduced by laser fluctuations.

Propagation of the standard deviation shown in part (b) of Fig. 3 through the Fourier transformation determines the frequency-dependent standard deviation of the amplitude and phase of the THz signal. Subsequent error propagation then allows us to link the time-domain noise to a frequency-dependent standard deviation of the measured dielectric spectrum of the sample.

4. References

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