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Dynamic range in terahertz time-domain transmission and reflection spectroscopy

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We present a quantitative method for identification of the dynamic range of the detectable absorption coefficient in the analysis of transmission terahertz (THz) time-domain spectroscopy data. In transmission measurements the largest detectable absorption coefficient is determined by the dynamic range of the THz signals, whereas in reflection measurements the largest detectable absorption coefficient is determined by the scan-to-scan reproducibility of the signal. © 2005 Optical Society of America

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In 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 number of THz spectra being published in the literature and collected in databases, it is becoming more and more important to address issues of data reproducibility and sources of error to avoid misinterpretation of experimentally obtained data. In a dielectric spectroscopy study of the polymer poly(methyl methacrylate), Kojima *et al.* may have interpreted the roll-off of the measured absorption coefficient at high frequencies as a manifestation of the boson peak.[8]

In this Letter we describe quantitatively and in a 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 the 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 was recognized in a qualitative way by Walther *et al.*[9] To the best of our knowledge this important effect has to date not been described in a quantitative way.

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

$$n(v) = 1 + \frac{c}{2\pi v d}\,\phi(v), \tag{1}$$

$$\alpha(v) = -\frac{2}{d}\,\ln\!\left\{ A(v)\,\frac{[n(v)+1]^2}{4n(v)} \right\}. \tag{2}$$

Owing to the typical single-cycle nature of the THz pulse, its frequency spectrum extends from the low gigahertz 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 normally independent of frequency and corresponds to the spectrum recorded with a completely blocked THz beam path. The origin of this noise is of an electronic nature, whereas fluctuations of the THz signal itself are caused mainly by laser intensity fluctuations.

Figure 1 shows a THz pulse 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. Inspection of Eq. (2) shows that 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:

$$\alpha_{\max} d = 2\ln\!\left[\mathrm{DR}\,\frac{4n}{(n+1)^2} \right]. \tag{3}$$

In other words, if the absorption coefficient exceeds $\alpha_{\max}$, the analysis will show an apparent absorption coefficient equal to $\alpha_{\max}$. In transmission THz-TDS the measurement of the refractive index is not subject to the same principal limitations as the measurement of the absorption coefficient.

In Fig. 1(c) we show the frequency dependence of $\alpha_{\max} d$ in a transmission experiment based on the THz pulse in Fig. 1(a), 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 $280 \mathrm{~cm}^{-1}$ in a 0.5-mm-thick sample. The DR of αd decreases in an almost linear fashion toward higher frequencies, until the cutoff frequency, which in this case is 4.6 THz. Above this frequency the apparent absorption coefficient will always be zero.
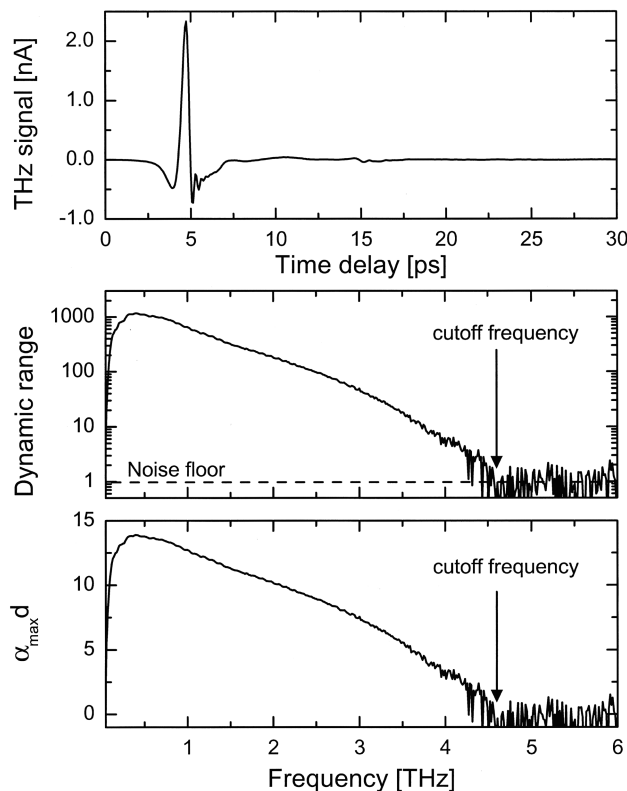
Fig. 1. (a) Typical THz pulse, (b) corresponding frequency spectrum, shown here relative to the noise floor. The upper limit of the detectable absorption is shown in (c).

In Fig. 2(a) the measured 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. The 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 figure α_{max} 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 such that without a quantitative analysis of the DR of the spectrometer it is not obvious that the absorption coefficient above 2.2 THz is not valid.

In Fig. 2(b) the measured absorption spectra of two different samples of the polar polymer polylactide is shown. This polymer has a glass transition temperature of 32.9 °C and changes its internal structure above this temperature. We show the absorption coefficient at room temperature, slightly below the glass transition temperature. One of the samples had a thickness of 0.58 mm, and the other sample had a thickness of 2.79 mm. Again the DRs of the absorption coefficients in the two measurements are shown in the same figure. At frequencies up to 1.25 THz the two measurements are identical except for the slightly stronger etalon effects in the thinner sample. The absorption spectrum recorded on the thick sample follows the DR at frequencies above 1.25 THz, with a relatively low noise level up to 4 THz. However, measurement of a thinner sample shows that the absorption in reality contin-

ues to grow, at least until the DR of the experiment is reached, this time at a frequency of 1.9 THz. The very sharp bend of the absorption curve, especially that of the thick sample, shows that the measured absorption coefficient can be trusted until the detection limit set by the DR is met.

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 from a silver mirror surface with a reflection coefficient 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.[10] Alternatively, the reference and sample signals are obtained as reflections from the front and the backside of a thick window in front of the sample.[11]

Here we consider only the case of normal incidence with a metal mirror as a reference surface. In the frequency domain the ratio between the sample and the reference spectra is then

$$|r|\exp(i\phi) = \frac{\hat{n} - 1}{\hat{n} + 1} = \frac{n + i\kappa - 1}{n + i\kappa + 1}. \quad (4)$$

Equation (4) can be inverted, and simple expressions for the index of refraction and absorption coefficient can be found:

$$n = \frac{1 - |r|^2}{1 + |r|^2 - 2|r|\cos\phi}, \quad (5)$$

$$\alpha = \frac{4\pi v}{c} \frac{2|r|\sin\phi}{1 + |r|^2 - 2|r|\cos\phi}. \quad (6)$$
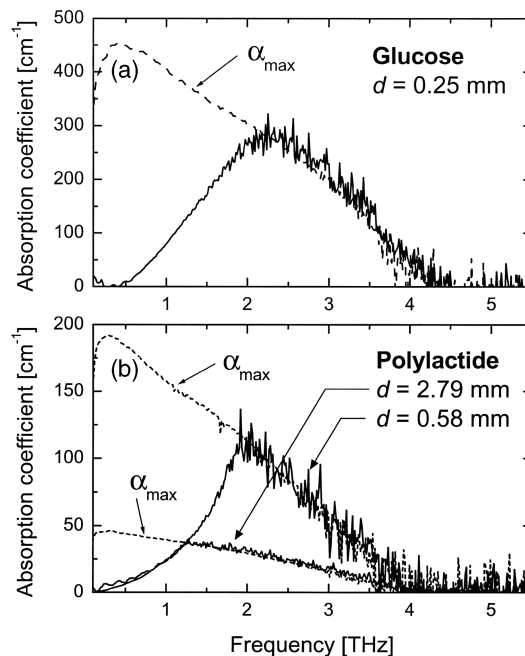

Fig. 2. (a) Measured absorption spectrum of amorphous glucose (solid curve). The dashed curve indicates the DR of the measurement. (b) Apparent absorption spectrum of polylactide recorded on samples with thicknesses of 2.79 and 0.58 mm (solid curves). The dashed curves indicate the DR of each experiment.
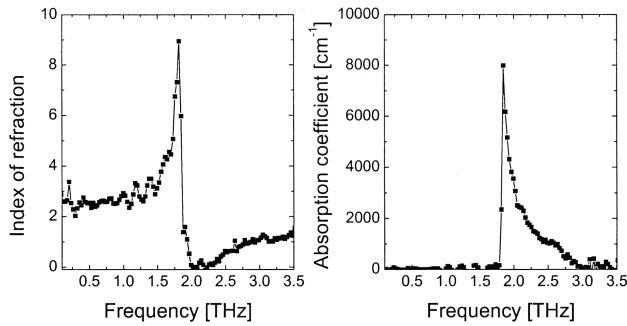
Fig. 3. Index of refraction and absorption coefficient of polycrystalline CsI, measured by reflection THz-TDS. The strong phonon resonance at 1.84 THz is clearly resolved.

The largest absorption coefficient is measured in the vicinity of $r = 1$ and $\phi = 0$. In this region the absorption coefficient can be approximated as

$$\alpha \approx \frac{4\pi v}{c} \frac{2\phi}{(1 - |r|)^2 + \phi^2}. \tag{7}$$

In a reflection experiment the measurement $|r| = 1 \pm s_r$, and hence $\phi = 0 \pm s_\phi$ corresponds to an absorption coefficient larger than

$$\alpha_{\max} \approx \frac{4\pi v}{c} \frac{2s_\phi}{s_r{}^2 + s_\phi{}^2}. \tag{8}$$

If we assume a phase accuracy $\delta\phi = 1°$, an amplitude accuracy $\delta r = 0.01$, and a frequency of 1 THz, the maximum absorption coefficient that can be measured is $\alpha_{\max} = 3.7 \times 10^4$ cm^{-1}, well above absorption coefficients that are normally encountered in dielectric and semiconductor materials. In Fig. 3 we illustrate the capability to measure large absorption coefficients with THz-TDS reflection spectroscopy with a measurement of the dielectric function of polycrystalline cesium iodide (CsI). CsI has a strong fundamental phonon mode at 1.84 THz. Therefore the dielectric function is difficult to measure with transmission techniques in this frequency range.[12] In this reflection measurement, on the other hand, the full dielectric function is measured across the fundamental phonon band, including the Reststrahlen band above the resonance. We are working on a study of the detailed, temperature-dependent properties of the dielectric function of the CsI phonon line, together with a detailed analysis of the important signal-to-noise aspects of reflection THz-TDS measurements.

In conclusion, we have shown that the dynamic range of a THz-TDS spectrometer is strongly frequency dependent. In transmission experiments its origin is found in the finite level of electronic white noise in the detection system. We have shown two examples where the frequency dependence of the DR may lead to artifacts in the absorption spectrum and obscure the interpretation of the experimental data. While the DR of a THz-TDS experiment limits an absorption measurement, the examples presented show the important fact that, if the measured absorption is within the DR, it is indeed a faithful representation of the real absorption coefficient. Larger absorption coefficients can be measured reliably with THz-TDS reflection spectrometers, where the limiting factor in absorption measurements is the noise of the phase and amplitude change upon reflection, which is determined mainly by laser fluctuations.

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