Accurate Material Parameter Extraction from Broadband Terahertz Spectroscopy

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***Abstract*—We demonstrate how a transfer function model based parameter extraction method, combined with total variance analysis, allows the extraction of both the complex refractive index and the thickness of a sample over a bandwidth of > 6 THz from THz time-domain spectroscopy measurements. We discuss how the techniques developed have been applied to absorbent powders measured at variable low temperatures.**

I. Introduction and Background

Typical terahertz time domain spectroscopic (THz-TDS) systems make use of a biased, photoconductive (PC) emitter illuminated using a femtosecond, pulsed laser. The THz radiation emitted is collected either from the rear of the emitter, after transmission through the emitter [1], or from the front in reflection mode [2]. The former technique is typically limited in bandwidth (up to 4 THz) owing to absorption in the PC material. In the latter arrangement, the collected radiation typically a has broader bandwidth (0.3 – 8 THz). To measure the broadband THz pulse, a thin electro-optic crystal (such as 150-μm-thick GaP) [2] is often used. However extraction of accurate optical parameters can be compromised by the generation of high amplitude, time–domain signal reflections from the thin detection crystal, which in turn can limit the subsequent frequency resolution if oscillation–free sample parameters are to be obtained. This is pertinent when extracting thickness of a sample due to the longer time window required, we demonstrate in this work that these system reflections do not impede thickness extraction. Furthermore correct determination of the sample thickness is critical in order to extract accurate optical parameters, and can constitute the greatest systematic source of uncertainty in measurements [3]. This is of particular importance when measuring of irregular samples, such as powdered materials, and when the sample thickness might change, for example under variable temperature conditions.

In this work, we obtain the complex refractive index, $\tilde{n}$, from samples by fitting a model to the transfer function (TF) of a sample. Provided both the model and TF contain sample reflections the thickness can also be extracted. First, by Fourier deconvolution, we calculate the TF of sample from our reference (dry air). We then estimate $\tilde{n}$ of the sample from the TF, by using a measured thickness [4]. A time window can include sample reflections of the THz signal which will lead to frequency-domain oscillations in $\tilde{n}$ [3]. In this work, we use a Fabry-Perot resonator based model (equation 1) which accounts for this effect and reduces the oscillations in $\tilde{n}$, as a function of thickness accuracy [4].

$$H_{sam}(\omega) = \frac{4\tilde{n}}{(\tilde{n}+1)^2} e^{-j\frac{\omega l}{c}(\tilde{n}-1)} \sum_{\delta=0}^{\delta=m} \left(\frac{1-\tilde{n}}{1+\tilde{n}} e^{\frac{\omega l}{c}\tilde{n}} \right)^{2\delta} \tag{1}$$

where l is the assumed thickness, ω is the angular frequency, c is the free space velocity of light and m is the number of sample reflections in the time window. We combine this model with measurements of the total variance [5] which provides a measure of the amplitude of the oscillations in $\tilde{n}$. We define total variance as:

$$TV = \sum |n[i] - n[i-1]| \tag{2}$$

Where n is the real part of refractive index and i is the frequency index. We use the real part of refractive index as it is less variant than the imaginary part [6], and thus increases the sensitivity of total variation to oscillations. An accurate thickness can therefore be obtained by minimizing the total variance as a function of assumed thickness.

α_{max} is a useful measure for estimating bandwidth [7], it defines a maximum absorption coefficient, α, based on the dynamic range of the reference. The bandwidth is therefore defined as the frequency range where α is not capped by this limit. We define a generalization for α_{max} for each discrete pulse of the sample response:

$$\alpha_{max,\delta} = \frac{2}{(2\delta+1)l} ln \left| DR \frac{4n(1-n)^{2\delta}}{(n+1)^{2(\delta+1)}} \right| \tag{3}$$

Where n is the real refractive index, l is the sample thickness (for this purpose, measured thickness is used), DR is the dynamic range and δ indicates the origin of the pulse, i.e. $\delta = 0$ is the initial pulse, $\delta = 1$ is the first reflection, etc. By calculating $\alpha_{max,\delta}$ for $\delta = m$, we can estimate the bandwidth which our model is valid over, and thus the bandwidth which we should perform total variance analysis over.

II. Results

In Figure 1 and Table I, we demonstrate the use of total variance as a measure of thickness accuracy for several common reference samples used in THz-TDS; lactose anhydrous (LA), lactose monohydrate (LM), quartz and silicon. In all

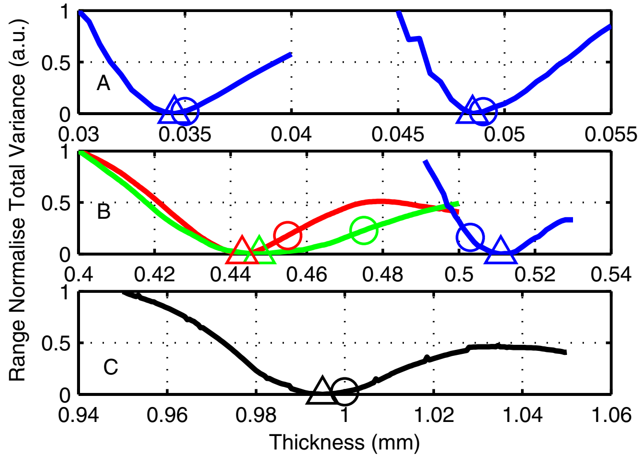

Fig. 1. Range Normalized Total Variance - the triangle indicates the local minimum and the circle indicates the measured thickness. Blue - Quartz, Red - Lactose Monohydrate, Green - Lactose Anhydrous and Black - Silicon

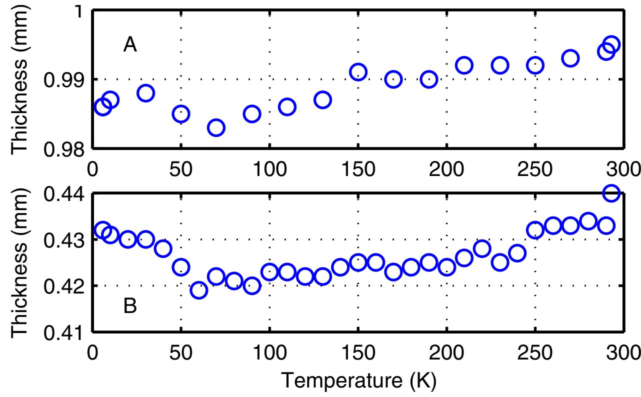

Fig. 2. Thickness as function of temperature, A) 1 mm Silicon, B) 0.455 mm Lactose Monohydrate

these measurements a coarse measurement was initially made to locate the local minimum of the total variation, from 90 % to 110 % of the measured thickness in 1 % steps, which was then repeated with a finer 0.5 μm step size across an appropriate range. The samples of quartz with thicknesses below 50 μm were prepared in house by lapping. As such, the thickness is known to a high degree of precision ($\pm$2.5 μm) and, as shown in Table I agrees very well with our total variance method. Compared to these measurements, thicker samples were only be measured with lower precision ($\pm$10 μm), which therefore results in a larger difference from the calculated values. The Lactose pellets were diluted with PTFE, to a concentration of 10 % by mass, and hydraulically pressed into pellets. These samples have shown the largest discrepancy between the

Sample	Extracted Thickness (mm)	Measured Thickness (mm)
Quartz	0.0345, 0.0485, 0.511	0.035, 0.049, 0.503
LM	0.443	0.455
LA	0.4475	0.475
Silicon	0.995	1

TABLE I
MEASURED AND EXTRACTED THICKNESS'S OF THE SIX SAMPLES

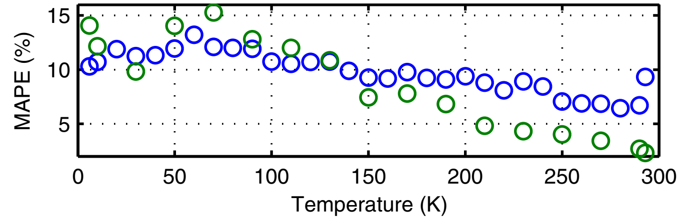

Fig. 3. Mean Absolute Percentage Error as a function of temperature, Blue - Lactose Monohydrate, Green - Silicon

measured and extracted thicknesses. This is attributed to local variations in thickness which are detected by the THz probe, which are averaged out during mechanical measurements of the thickness. We see good agreement between our extracted thicknesses and the thickness uncertainty of the thick Quartz and Silicon samples. A common assumption in variable temperature measurements is constant thickness. We demonstrate here, however, (Figure 2) that significant thickness change can occur, leading to significant errors in absorption coefficient (Figure 3) at low temperatures if changes in thickness are not considered. The increased accuracy which our technique provides will be crucial when extracting accurate low temperature parameters from THz-TDS across a wide range of samples.

III. Conclusion

In conclusion we present the use of a transfer function based parameter extraction technique on a variety of samples, and variable temperature controlled conditions. We have demonstrated that this methodology is a robust technique not only for measuring thickness, but also for correcting for changes in thickness during variable temperature measurements.

Acknowledgment

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