Analysis of optical thickness determination of materials by THz-TDS

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Abstract. Terahertz time-domain spectrometry (THz TDS) is a sensitive probe of the complex dielectric response of materials. Methods vary for converting time-domain response into final material optical parameters together with estimation of associated uncertainties. Here we point out the importance of using an accurate extraction procedure with particular emphasis on the error introduced by associated inaccuracy in thickness determination of a sample. The Total Variation (TV) method is used to estimate sample thickness to sub-micron accuracy, by constructively using the phenomena of multiple internal reflections (‘ringing’) within a sample. The applicability and performance of the TV methodology is discussed.

1. Introduction
The THz frequency domain is defined to span from 0.1 to 10 THz [1]. THz TDS is a relatively new tool that emerged mainly due to the work of Auston in the field of photoconductive generation [2] and detection [3] of THz radiation. As a separate instrument for measuring material properties in the THz domain, THz-TDS was first used by Grischkovsky et al. [4]. Since then THz TDS has been used for characterization of a wide range of materials namely; semiconductors, biomolecules, polymers and even thin metal films [5, 6]. TDS provides a time-domain response function with the absence of the sample providing the conventional reference, system response. Conversion of time-domain traces into dispersion curves of spectral absorption coefficient and refractive index, calls for signal processing techniques to be used. Many factors compound to make signal processing an involved stage during analysis, the most important being the management of system noise. Uncertainty in sample thickness and position, besides other various sources of random and systematic errors are at issue, too. However there are still no internationally-agreed methods for processing the THz response of a sample. Constructive suggestions are therefore proffered in the use of a transfer function for optical parameter extraction and of sample-thickness determination by the total variation (TV) method [14].

2. Extraction Procedures
Determination of estimates of optical constants generally starts with ratioing the (Fourier-transformed) sample response to the reference. This constitutes an experimental transfer function (TF). A theoretical TF is then constructed, that factors in; signal attenuation and retardation of the probing THz, transmission coefficients at the sample-air interface; and internal, multi-order, reflections of the probe-pulse within the sample – a Fabry-Perot (FP) like ‘ringing’. The expression for such a TF in the case of a single-layer sample has been described earlier [7] and is formulated as:

\[
\text{TF}_{\text{single-layer}} = \frac{1}{1 - \frac{4}{r^2} \sin^2 \left( \frac{\pi d}{r} \right)}
\]
\[ \tilde{H}(v) = \tilde{\ell}_{12}(v)\tilde{\ell}_{21}(v)\exp(-i2\pi vd(\tilde{n}(v) - n_{\text{air}})/c) \cdot \sum_{m=0}^{m=M} [\tilde{r}^2(v)\exp(-i4\pi v\tilde{n}(v)/c)]^1. \] (1)

\[ \tilde{r}(v) = \frac{\tilde{n}(v)-n_{\text{air}}}{\tilde{n}(v)+n_{\text{air}}}, \quad \tilde{\ell}(v) = \frac{2n_{\text{air}}}{\tilde{n}(v)+n_{\text{air}}}, \quad \tilde{\ell}_{21}(v) = \frac{2\tilde{n}(v)}{\tilde{n}(v)+n_{\text{air}}} \]

\[ \tilde{n}(v) = n(v) - ik(v) \]

are the complex Fresnel coefficients at normal incidence; \( \tilde{n}(v) = n(v) - ik(v) \) is the complex refractive index of the sample; \( m \) is the order of FP reflections in the time-domain (TD) response and can be derived via noting that: \( t_{\text{max}} \geq nd(1 + 2m)/c \), with \( t_{\text{max}} \) being the time-elapse for a scan. Note that this model does not include scattering effects taking place on the sample, whose surfaces are assumed to be plane-parallel and flat at the THz energies concerned. The summation term of (1) accounts for FP ringing in the sample. A number of approximations are often made to simplify (1), namely; having Fresnel coefficients to be only real; ignoring higher order reflections in the sample; and, approximation of the FP term as an infinite sum of geometrical progression: \( \left(1 - r^2(v)\exp(-i4\pi vd\tilde{n}(v)/c)\right)^{-1} \). All these simplifications introduce additional errors to the final estimates of optical parameters, the last being especially problematic (it is only valid for optically thin samples, when \( m \geq 5 \)).

The structure of (1) necessitates the utilization of non-linear regression algorithm for estimation of the complex refractive index by minimization the difference error between theoretical and experimental TFs. Throughout this paper a Levenberg-Marquardt algorithm is used [8], which combines the advantages of Newtonian and gradient descent algorithms.

3. The TV methodology

Error analysis is an essential part of any THz-TDS measurements. An extensive analysis of both systematic and random sources of errors has been done previously [9]. It provides complete information about the accuracy of extracted properties and helps to track and reduce sources of uncertainty. One of the major sources of uncertainty comes from inaccurate determination of sample thickness [10]. The nature of equation (1) is fortuitous in assisting estimation of optical thickness in a very accurate manner.

Various methods have been employed for optical (also read, ‘numerical’), thickness determination. Kruger et al. [11] used two measurements of the same material but having different thicknesses. After extracting all possible solutions via TF regression analysis, the two coinciding curves corresponded to the actual thickness. The disadvantage of this method is a necessity to reproduce identical conditions for both measurements. It also has a relatively high computational cost. The approach has been developed for determination of thickness for sub-100-µm samples [12]. Its algorithm relies on an additional Fourier transform of the frequency-dependent material parameters to a quasi-space domain. Other methods [13] are based on time-domain reconstruction of the THz response, but they too suffer from high computational requirements. Here a TV method is adopted as a simple and reliable approach for numerical thickness determination [14]. The principle of TV is based on the fact that in most cases the dispersive curves of material properties are smooth, so that an incorrect determination of thickness manifests as spurious oscillations. By iteratively varying thickness, the algorithm selects the best-behaved curve, and this corresponds to the most accurate thickness estimate. The value of TV is calculated for each thickness as:

\[ TV = \sum_{m=m_1}^{m_2} \left[ \left| n_k - n_{k-1} \right| + \left| k_k - k_{k-1} \right| \right], \] (2)

where \( f_1 \) to \( f_7 \) defines the frequency domain where material parameter curves are smoothest (this typically excludes lower and higher limits of the operating frequency domain). Mathematically, a plot of TV shows the degree of fluctuation in the curves of refractive index and extinction coefficient for each separate thickness value.

Figure 1 shows an example of depth determination for a 1 mm-thick silicon plate. The TV plot clearly shows the minimum that corresponds to the actual value of thickness (the smoothest curve). Figure 1B is a high resolution about the inflection in figure 1A. It estimates the actual thickness of the plate to be 1.0319(1) mm. This is one order of accuracy above the ability of standard micrometer
gauges. Multiple micrometer gauge measurements give the thickness of the Si plate to be 1.031±0.001 mm. Note that the sample thickness found numerically corresponds to the effective thickness (taking into account surface roughness and non-uniformity), of the material in the area where the THz beam illumines the sample.

![Figure 1](image-url)  
**Figure 1.** Total variation of material parameter curves, plotted for coarse (A) and denser (B) thickness ranges. The data is shown for a silicon plate of approximately 1 mm thickness.

Successful application of the TV method requires two important conditions to be met: FP-like ringing needs exist and to be above the noise floor of the system. In fact, ringing of the THz pulse within the sample provides the physical means of accurate optical determination of the sample depth. If no internal reflections are detectable in the sample response, the TV algorithm does not work efficiently. In this case, instead of a sharp minimum in the TV curve, a broad, flat minimum extends for 10s of µm. Examination of equation (1) underscores this, where the FP term collapses to unity if no internal reflection is present. Spurious oscillations are consequently absent in parameter dispersion curves. The absence of optical ringing in the sample response can be for several reasons. In measurements of optically-thick samples employing a relatively short scan-length \( t_{\text{scan}} < 3n d/c \), when \( n^{th} \) order transmissions allied to \( n^{th} \) order back-face reflections, simply arrive at the detector after scanning has finished. Interestingly, high absorption by a sample (e.g. LiNbO\(_3\)), does not lead to a significant decrease in the amplitude of FP-reflections. The feature most responsible for attenuating \( n^{th} \) order internal reflection amplitude is scattering of THz radiation both within and at the surfaces of the sample. It is principally this feature that limits the application of the TV methodology to study of biomaterial samples like proteins, amino acids, sugars, etc. We have conducted a series of measurements of lysozyme, glycine and lactose pellets; and for all these samples there is no detectable ringing, while system dynamic range remains sufficiently high to distinguish THz pulse ringing within a 0.5 mm thick high-absorbing plate of lithium niobate.

Another significant factor influencing the performance of the TV method is sample alignment. The ideal case (as per the assumption of normal incidence in the use of Fresnel coefficients in (1)), is to set the sample-face-normal in-line with the THz beam axis. But this is not always possible, so a series of measurements have been performed to investigate the effect of face-normal deviation. Sample-tilt corresponds to an effective (secant) increase in sample thickness.

\[
d_{\text{view}} = d_{\text{actual}}/\cos(\alpha)
\]  
(3)

Figure 2 shows the results of thickness determination of the tilted sample by the TV method and that calculated by (3). Sample thickness calculated by the TV method closely follows prediction (within the angle determination inaccuracy), proving the capability of the method to extract thickness correctly. For a sample tilt of 1° the sample depth changes by 0.3 µm as determined by both TV and equation 3. According to this analysis, in order to ensure thickness-error to be less than 0.1 µm due to sample misalignment, the sample normal needs to be set to within ±0.6° deviation off from beam-axis.
Figure 2. Changes in silicon plate thickness determined from measurements using the TV method, and from equation (3).

4. Conclusions
The governing requirements have been discussed for accurate estimation of material optical properties by THz TDS. The applicability and performance of numerical thickness determination using total variation (TV) technique is assessed. Accurate information on sample-depth, being the major source of uncertainty, is necessary for reliable material parameter (i.e. optical constants) determination. The accuracy of the TV method is shown to reach ±0.1 μm if FP-like multiple-order internal reflection response is present in the sample and alignment of the sample-normal is within ±0.6° of the beam-axis.

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