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Multilayer Extraction of Complex Refractive Index in Broadband Transmission Terahertz Time-Domain Spectroscopy

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***Abstract*—In terahertz spectroscopy, multi-layered samples often need to be measured, for instance in a liquid flow cell, and this complicates the extraction of material parameters. We present a spectroscopic parameter extraction algorithm for multilayer samples that can also be used to extract the thickness of an unknown sample layer.**

I. INTRODUCTION

Unlike most vibrational spectroscopic techniques, terahertz time-domain spectroscopy (THz-TDS) allows the full electric-field of the THz pulse to be measured coherently in the time-domain. This gives a precise determination of the phase delay and absorption in the form of a complex refractive index (ñ). To calculate ñ, a model is fitted to a sample's transfer function [1].

A sample of interest can consist of multiple layers, such as those undertaken on liquids in which a flow cell with two THz transparent windows. The undetermined layer's ñ cannot then be accurately extracted without recourse to modelling the entire structure. Here, we present an algorithm capable of extracting both ñ, and the thickness for an undetermined layer in any multi-layered structure, using transmission THz-TDS [2].

II. MODELLING OF A MULTILAYER SAMPLE'S TRANSFER FUNCTION

To extract ñ for a sample, we numerically fit a transfer function model to the experimental data by minimizing an error function [1]. This function will converge to a global solution provided phase unwrapping is correctly accounted for. In this work we ensure the refractive index "wraps" around a region based on an initial estimate. This is based on the approximate periodicity of the real refractive index:

$$n = n \pm a\frac{c}{fl} \qquad (1)$$

Where c, f, a and l are the free space velocity of light, frequency, an arbitrary integer and thickness of the layer respectively. This finds the local solution to the error minimization in the region of an initial estimate of ñ and allows us to make considerable performance optimizations in the algorithm.

To numerically fit a model, we use the Nelder Mead [1] algorithm to minimize the error function (ε) with respect to ñ:

$$\varepsilon = \left(\hat{H} - H(\tilde{n})\right)\left(\hat{H} - H(\tilde{n})\right)^{\dagger} \qquad (2)$$

Where $\hat{H}$ is our measured transfer function, deconvolved from reference (air) and sample measurements, $H(\tilde{n})$ is our transfer function model for the undetermined layer's ñ and † is the conjugation operation.

To calculate $H(\tilde{n})$, we assume that the THz radiation is a parallel beam passing through the sample, which consists of a number of homogenous layers of dielectric. We further assume that layers other than the undetermined layer have a known ñ and thickness, which are all measured independently. The beam partially reflects at each interface, leading to resonances between interfaces. Importantly, these resonances are not independent and have inconsistent length within layers owing to attenuation. If the sample consists of more than one layer this can quickly lead to a complex time response (Fig. 1).

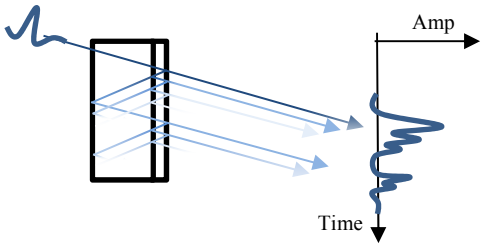

Fig. 1: A THz pulse interacts with a two layer sample, the beam forks at each layer interface to produce a composite of THz pulses (angle offset for clarity). Each path has decreasing amplitude, thus resonances within the same layer are not the same length.

To build a transfer function model, we split the THz beam into discrete segments between interfaces. We use a recursive process to build a "*resonant tree*" out of these segments (Fig. 2), as at each interface the beam path splits into a reflection and transmission path.

The key to the recursive process is that a resonance tree can be considered to be a combination of two sub trees, and thus, two sub transfer functions, joined at an interface. The process only considers its segment of the beam and the effect of the reflection or transmission on the beam at each interface. If any beam is thought to fall outside of the time window of the experimental measurement, or attenuated to a level that is below the dynamic range of the instrument, then termination conditions have been met and that branch will be terminated. In the special case where a pulse being considered has passed through the entire multi-layer structure, and is propagating towards the detector this forms part of the final transfer function. If not, two child copies are created and form a reflection and transmission subtree (see Fig. 2), Each is then multiplied by an appropriate reflection or transmission coefficient [1] for the interface, and then the sum is multiplied by the current segment's propagation coefficient [1]. This, allows us to construct the THz beam path through any multi-layered sample, which can be visualized as a tree structure. As an example of a simple system, Fig. 2 shows how a simple resonant tree can recursively calculate the samples transfer function. An initial calculation process (I) creates a reflection child process (1R1), and a transmission child process (1T1). These in turn create multiple child processes until termination conditions are met. Layer information and termination conditions are propagated to each child process (blue arrows), the resulting calculated transfer function propagated back up (yellow arrows) to the parent, this way the complete transfer

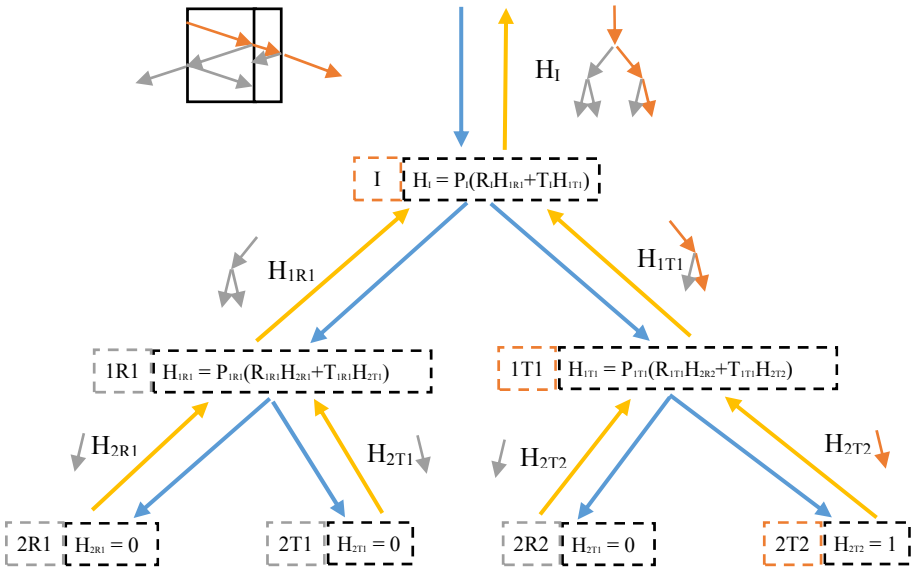

Fig. 2: A recursive process builds a "resonant tree" (show next to yellow arrows) and calculates the corresponding transfer function. The above transfer function is calculated for the simple case where only a direct pass through occurs (see top left, diagram of beam propagation – angle offset for clarity only). Starting with an initial process (I), each process (boxes) will create two child processes to calculate the reflection (R) and transmission (T) sub trees. Information about the model is propagated to the child process (blue arrows) from the parent process. This includes spectral information about the layers but also the layer location for a child process, a time limit and associated dynamic range limit. These are used to form termination conditions. Child processes will return their calculated H to their parent process (yellow arrows). A parent process will combine the reflection and transmission H, using the current layer's propagation coefficient (P), the interface's reflection coefficient (R) and transmission coefficient (T). This way a complete transfer function (H_I) is calculated (and hence a full tree is formed) and returned to the error minimization algorithm.

can be constructed.

After calculating the transfer function, we apply a correction for air displaced, which would otherwise be present during a reference measurement. Unlike previous methods [3], our algorithm can extract ñ without predetermining the THz beam paths through the sample.

Resonance within an undetermined layer can be used to extract the thickness. Etalons will form within the extracted ñ if the thickness is incorrect, by minimizing these etalons the thickness can be extracted. We use total variance (TV) [1], to quantify and minimize these etalons in real refractive index.

III. Results

We have considered a three-layer sample, consisting of a flow cell comprising two 2.1 mm *z*-cut quartz windows with an ~180-µm-thick cavity between for a liquid sample. We used an empty flow cell (no windows or sample) as our reference, and a flow cell with propan-2-ol as a sample. We used TV in combination with smoothed measurements of ñ, Fig. 3, to verify the channel thickness. By using our algorithm, we are capable of extracting both the real, n, and imaginary, κ, parts of ñ of propan-2-ol while also confirming the thickness of the sample.

IV. Summary

We have developed an algorithm that is capable of extracting a sample's ñ within a multi-layered structure, and which does not require predetermined beam paths. We have used this methodology to extract the ñ of a liquid within a flow cell.

V. Acknowledgements

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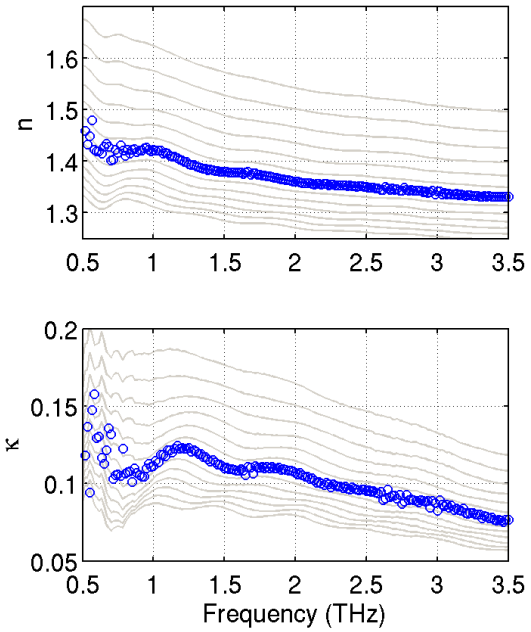

Fig. 3: Propan-2-ol measurements taken from a flow cell measurement, top – real refractive index (n), bottom – extinction coefficient (κ). Blue dots – raw data at the extracted thickness of 180 µm, grey curves – smoothed data used for thickness extraction (120 – 240 µm, 10 µm steps).

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