Scattering Robust Features for Classification of Materials Using Terahertz

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***Abstract*—Terahertz spectroscopy has emerged as an important tool for identification and classification of materials, which exhibit absorption features at specific and distinct frequency bins in the THz spectrum. The scattering of terahertz radiation from granular substances can significantly distort the spectral fingerprint of the material under study. In this paper we propose a signal processing based technique to mitigate the effects of scattering from the measured terahertz spectrum to produce features that can be used for scattering invariant classification of material using THz-TDS.**

I. Introduction

Scattering is a major problem in the study of polycrystalline material samples at THz frequencies. The cause of scattering in this case is inhomogeneities in the sample. The variations are roughly of the scale of tens of microns [1]. For scattering centers that have sizes comparable to THz wavelengths, the scattering process becomes quite complex and cannot be explained by simple Rayleigh scattering.

In order to resolve the problem of scattering in polycrystalline materials, a common approach is to grind the sample material to a fine powder before mixing it with fine powder of polyethylene or HDPE. This minimises the number of coarse grains in the sample and thus limits the scattering. Another approach applied by Shen *et al.* [2] effectively reduced the scattering effects by time domain averaging a large number of disjoint transmission measurements. Recetly in 2008, Franz *et al.* [3] applied the theory of the Christiansen Effect to numerically eliminate the effects of scattering from the terahertz extinction spectrum using the knowledge of frequency dependent refractive indices of the sample and host material. In 2010, Kaushik *et al.* [4] introduced a new technique that used Fresnel echoes generated during the THz-TDS process to estimate and mitigate the scattering effects in the measured extinction spectrum of the sample under study. This technique relied on the availability of strong and time resolved Fresnel echoes for estimating the scattering loss. All these approaches either require a complex and time consuming sample preparation and measurement process or rely on specific information about the sample under study. However, in real-world applications such as stand-off measurements, *a priori* information of the the sample is not always known and therefore limits the applicability of these techniques.

In this paper, we introduce a new signal processing based technique for mitigating effects of scattering in THz-TDS measurements that does not require any *a priori* information of the sample and produces features that are robust against various scattering conditions and can be used for material classification using THz-TDS.

II. Background

When a plane wave of frequency f propagates through a material with complex dielectric constant $n + ik$, it is subjected to frequency dependent propagation effects, such as attenuation and dispersion. The total attenuation suffered by the signal is mainly due to absorption and scattering of the signal from the material. For materials that exhibit sharp absorption features that are present in specfic and distinct freqency bins, the frequency dependence of absorption and scattering can differ markedly from each other. Generally, scattering loss depends on the dimensions, density of the internal structure and refractive index of the medium, and can be expressed as a low order polynomial over the given frequency range, depending on the medium of propagation [5]. On the other hand, absorption loss is a consquence of intermolecular vibrations in the medium. For materials that exhibit sharp absorption features, such as $\alpha-$monohydrate lactose, a commonly used biochemical ingredient of various pharmaceutical tablets, the absorption features are confined to distinct frequency bins specific to the constituents of the medium, and therefore we assume the sharp absorption features can be expressed as higher order polynomials over the frequency range.

III. Method

Consider a plane wave of angular frequency ω propagating through a sample of material with complex dielectric constant $n(\omega) + ik(\omega)$, with thickness d. The Fourier transform of the transmitted signal is given by:

$$Y(\omega) = X(\omega) \exp\left[in(\omega)\omega\frac{d}{c}\right] \exp\left\{-\alpha(\omega)\frac{d}{2}\right\}, \quad (1)$$

where $X(\omega)$ is the Fourier transform of the signal before propagating through the sample, $n(\omega)$ represents the frequency dependent refractive index and $\alpha(\omega) = 2k(\omega)\omega/c$ represents the frequency dependent total attenuation coefficient of the material. The magnitude response of the sample is given by:

$$|Y(\omega)| = |X(\omega)| \exp\left\{-\alpha(\omega)\frac{d}{2}\right\}, \tag{2}$$

or

$$\log(|Y(\omega)|) = \log(|X(\omega)|) - \alpha(\omega)\frac{d}{2}. \tag{3}$$

As mentioned earlier, $\alpha(\omega)$, the total frequency dependent attenuation consists of two contributing terms namely the frequency dependent absorption loss $\alpha_{\mathrm{a}}(\omega)$ and the frequency dependent scattering $\alpha_s(\omega)$. This relationship can be expressed as:

$$\alpha(\omega) = \alpha_{\mathrm{a}}(\omega) + \alpha_{\mathrm{s}}(\omega). \tag{4}$$

Thus (3) can be expressed as:

$$\log(|Y(\omega)|) = \log(|X(\omega)|) - \alpha_{\mathrm{a}}(\omega)\frac{d}{2} - \alpha_{\mathrm{s}}(\omega)\frac{d}{2}. \tag{5}$$

We model the scattering as a function of the angular frequency ω as

$$\alpha_{\mathrm{s}}(\omega) = \alpha_0 \left[\frac{\omega}{\omega_0}\right]^A, \tag{6}$$

where ω_0 is the angular frequency at which $\alpha_s = \alpha_0$, α_0 and A are unknown parameters that depend on the sample granularity. A similar model was proposed by Shen *et al.* [2] for modelling the observed extinction spectra obtained from THz-TDS of various samples made of non-absorbing granular PE particles.

Thus the expression for the magnitude response of the sample can now be expressed as:

$$\log(|Y(\omega)|) = \log(|X(\omega)|) - \alpha_{\mathrm{a}}(\omega)\frac{d}{2} - \alpha_0 \left[\frac{\omega}{\omega_0}\right]^A \frac{d}{2}, \tag{7}$$

or

$$\begin{aligned} Z(\omega) &= \left[\frac{\omega_0}{\omega}\right]^A \log(|Y(\omega)|) \\ &= \left[\frac{\omega_0}{\omega}\right]^A \log(|X(\omega)|) - \left[\frac{\omega_0}{\omega}\right]^A \alpha_{\mathrm{a}}(\omega)\frac{d}{2} - \alpha_0\frac{d}{2}, \end{aligned} \tag{8}$$

where (8) shows that the spectral shape of $Z(\omega)$ does not change with the scattering coefficient α_0, but is merely shifted in overall level by $\alpha_0\frac{d}{2}$. In order to eliminate this scattering dependent level shift, we initialize the value of A with 0 and increment it in steps of 0.01 to iteratively solve the following equation:

$$Z_0(\omega) = Z(\omega) - \overline{Z(\omega)}, \tag{9}$$

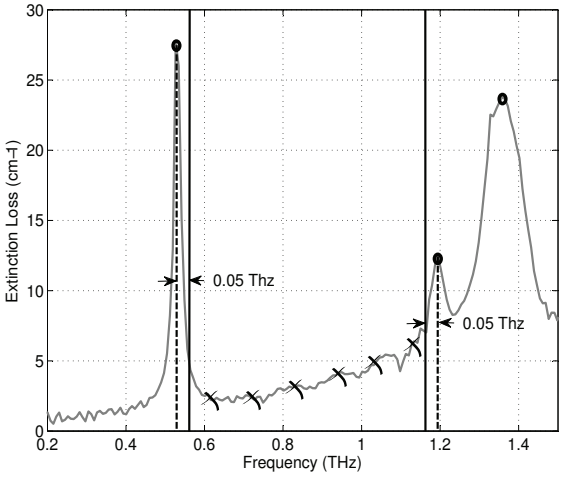

Fig. 1. Application of a peak detection algorithm to the experiment measurements to identify the absorption peak locations (marked as ecllipses), the black lines (dotted and solid) represent the measurement setup frequency resolution limits, and the region ω_{s}, where all the attenuation is due to scattering only (marked with crosses). The unknown parameter A is estimated over the frequency range ω_{s}.

till, for some defined frequency region ω_{s} in which $|Y(\omega)|$ shows no absorption features and all the attenuation is due to scattering, the value of $Z_0(\omega_{\mathrm{s}}) - (\omega_0/\omega)^A \log(|X(\omega_{\mathrm{s}})|)$ is minimized, where $\overline{Z(\omega)}$ represents the mean of $Z(\omega)$.

Accordingly, the spectral function $Z_0(\omega)$ does not depend on scattering, and depends only on the reference signal and absorption from the sample. One could obtain spectral features for scattering invariant material classification from $Z_0(\omega)$. Our focus here is on the absorption features, which can be obtained from $Z_0(\omega)$ as:

$$\alpha_{\mathrm{a}}(\omega) = -\frac{2}{d}\frac{(\omega/\omega_0)^A Z_0(\omega)}{\log(|X(\omega)|)}. \tag{10}$$

In the above procedure an important step is to find the frequency region ω_{s} in which the measured spectrum shows no absorption features. We know that for materials like α-monohydrate lactose, absorption features are sharp and present in distinct frequency bins. Thus, we used a threshold based peak detection algorithm for identifying the location of the absorption features. Once the location of the abosrption features are determined, the region between two consecutive absorption peaks can be identified as ω_{s}, in which the measured spectrum shows no absorption features and all the attenuation is due to scattering. In our measurement setup the frequency resolution is approximately 0.1 THz, thus when determining the region ω_{s}, based on the detected absorption peak location, we exclude the regions in the vicinity of the absorption peaks that falls within the frequency resolution limits. This is shown in the Fig. 1.

IV. Experimental Details

Transmission mode terahertz time-domain spectroscopy through two samples made of α-monohydrate lactose and

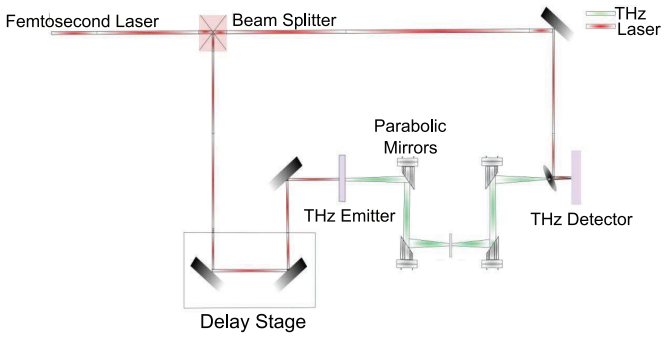

Fig. 2. An illustration of the THz-TDS pump probe system used for our experiments.

granular PE powder is carried out at the National ARC T-ray Facility at the University of Adelaide, South Australia. In the apparatus shown in Fig. 2, a MiraSeed (Coherent) Ti:Sapphire femtosecond mode-locked laser is used as the source of the optical pulses. It is pumped by a Verdi (Coherent) V6 with a wavelength of 532 nm. The femtosecond (fs) laser produces output pulses of duration less than 50 fs at a repetition rate of 76 MHz. The laser has an output power of 0.75 W with a center wavelength of 800 nm. The femtosecond laser beam then is split into the probe beam and the pump beam using a beam splitter. At the pump beam, the laser is modulated at 300 Hz by an optical chopper before being focused onto a photoconductive emitter that is biased at 90 V dc. The generated THz pulse is focused onto the detector using four off-axis gold plated parabolic mirrors. The probe beam is designed to gate the incoming THz pulse at the detector. The entire THz waveform is sampled by varying the optical length of the delay stage.

Using the setup described above and shown in Fig. 2, we carry out transmission measurements for two samples comprised of α-monohydrate lactose powder and PE powder mixed in a 1:1 ratio and pressed to form a pellet. The PE powder used for each sample had different grain sizes. The first sample comprised of PE powder with average grain diameter of 60 m while the average grain diameter for the PE powder of the second sample was 360 m. Due to the scattering effects, the usable bandwidth of our system was limited to 1.6 THz. Therefore, at this stage we were only able to test the proposed technique on samples made of α-monohydrate lactose, as it exhibits various absorption bands in the given frequency range.

V. Data Aquisition

In this analysis, we assume a plane wavefront for the terahertz radiation, in the far field. By analysing the sample propagation geometry, and assuming that the reflections are removed from the sample and reference data, the measured optical attenuation $\alpha_{\mathrm{m}}(\omega)$ for the α-monohydrate lactose and PE samples is given by:

$$\alpha_{\mathrm{m}}(\omega) = \frac{2}{d}\left\{\ln\left[\frac{4n_{\mathrm{s}}n_0}{(n_{\mathrm{s}}(\omega)+n_0(\omega))^2}\right] - \ln|H(\omega)|\right\}, \quad (11)$$

where n_{s} is the refractive index of the sample, n_0 is the refractive index of free air and $H(\omega)$ is the transfer function of the sample.

Both the samples clearly show characteristic absorption features of α-monohydrate lactose at 0.53 THz, 1.19 THz and 1.39 THz. It should be noted that along with the distinct absorption features both the samples show a baseline slope owing to scattering of the terahertz signal due to the granular nature of the samples. As expected, the sample with larger PE particles scatter the terahertz waves more as compared to the smaller PE particle sample and hence shows a greater baseline slope.

VI. Results

In order to obtain the scattering invariant features from the experiment measurments, we apply the processing described in Section III and use (10) to obtain $\alpha_{\mathrm{a}}(\omega)$, the scattering removed absorption spectrum of the samples. Fig. 3 and Fig. 4 show the comparison of the scattering removed absorption features with the measured extinction spectrum.

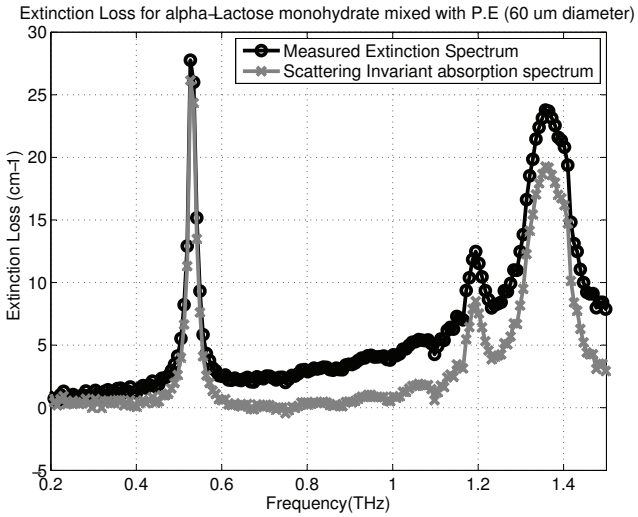

Fig. 3. Measured Extinction loss spectrum of sample pellet made of α-monohydrate lactose mixed with PE powder with avg. dia. 60 μm (black with circles) vs the scattering removed absorption spectrum of sample pellet made of α-monohydrate lactose mixed with PE powder with avg. dia. 60 μm obtained using (10) (Grey with cross).

VII. Conclusion

Sensing and imaging with terahertz frequency for biomedical applications faces various hurdles. Scattering with its ability to obscure the absorption fingerprint of the material under study remains a common problem for many spectroscopic and imaging applications. Nevertheless, T-rays exhibit significantly reduced scattering in human tissue in comparision to near-infrared optical frequencies due to the increased wavelength.

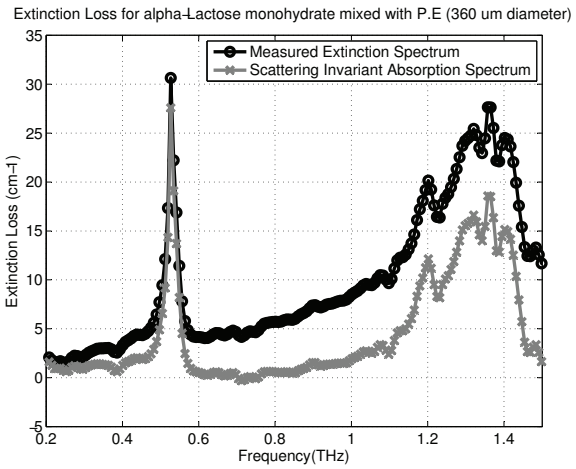

Fig. 4. Measured Extinction loss spectrum of sample pellet made of α-monohydrate lactose mixed with PE powder with avg. dia. 360 μm (black with circles) vs the scattering removed absorption spectrum of sample pellet made of α-monohydrate lactose mixed with PE powder with avg. dia. 360 μm obtained using (10) (Grey with cross).

However, scattering remains an important concern because it is neglected by common signal processing algorithms in this field.

In conclusion, we introduce for the first time a signal processing based technique for enabling scattering invariant classification of materials using THz time domain spectroscopy. The method was tested on two sample pellets comprised of α-monohydrate lactose commonly used as an ingredient of various pharmaceuticle tablets and PE with different granularity in each sample. As shown by the results presented here, the method reasonably eliminates the sample's scattering contribution from the measured extinction spectrum to reveal true absorption features of the sample, without requiring any special sample preperation techniques, measurement setup and any *a priori* information about the sample material. This make the method very useful for mitigating scattering effects in real-world applications such as in standoff measurements, food, and pharmaceutical quality control.

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