Reduction of Scattering Effects in THz-TDS Signals

Mayank Kaushik, *Student Member, IEEE*, Brian W.-H. Ng, *Member, IEEE*, Bernd M. Fischer, and Derek Abbott, *Fellow, IEEE*

***Abstract*— The scattering of terahertz radiation from the granular nature of a sample can potentially distort or obscure its characteristic spectral features. Several techniques have been proposed to reduce the effects of scattering in terahertz time-domain spectroscopy (THz-TDS) measurements, that usually require a complex measurement apparatus or rely on specific information about the sample under study. However, in real-world applications *a priori* information of the sample is not always known and therefore the applicability of these techniques may be limited. In this letter, we present a method for estimating and mitigating scattering effects in THz-TDS measurements for samples made of material exhibiting sharp and sparse absorption features, without requiring information of its granularity, refractive index, and density.**

***Index Terms*— Scattering, teratertz spectroscopy.**

I. Introduction

TERAHERTZ time-domain spectroscopy (THz-TDS) has emerged as an important tool for non-contact identification and classification of various materials. Particularly, in the study of solids, THz or T-ray spectroscopy is used to observe and extract absorption features that can be indicative of morphological and crystalline condition of the material under study [1], [2]. However, when the grain sizes in the solids become comparable to THz wavelengths, the consequent scattering cannot be explained by a simple Rayleigh model. This can significantly distort the spectral fingerprint of the material under study.

While several analyses have been reported for understanding the influence of scattering on the terahertz signal [3]–[6], only a few techniques that mitigate the effects of scattering have been reported. Under the assumption of weak scattering limit, Bandyopadhyay et al. [3] applied the Mie theory [7] of spherical particles to essentially separate the contributions of true absorption and scattering from the sample. On the other hand, using the knowledge of frequency dependent refractive indices of the sample and host material, Franz et al. [5] applied the theory of the Christiansen Effect to numerically eliminate the effects of scattering from the terahertz extinction spectrum. In 2008, Shen et al. [6] effectively reduced the scattering effects by averaging a large number of disjoint transmission measurements in the time domain. Recently, in 2010, Kaushik et al. [8] used the usually discarded 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. The technique relied on the availability of strong and time resolved Fresnel echoes in the measurements. Most of the approaches discussed here require either complex and time consuming measurement techniques or detailed knowledge of the sample under study.

In this letter, we introduce a method that allows the estimation of scattering for unknown sample granularity, refractive index, and particle packing density which does not require any customized measurement apparatus or procedure. We consider materials that exhibit sharp absorption features at specific frequency bins, such as α-monohydrate lactose, mixed with a transparent packing material such as granular PE.

II. Method

A set of plane waves with frequency variable ω propagating through a material with complex dielectric constant, suffers from frequency dependent attenuation due to the absorption and scattering of the signal from the material. For materials exhibiting sharp absorption features, the pattern of frequency dependence of scattering and absorption can differ markedly from each other. Generally, scattering loss depends on the dimensions and density of the internal structure of the target, and can be expressed as a linear or quadratic function over the given frequency range, depending on the medium of propagation [9]. On the other hand, absorption loss is a consequence of molecular rotations or vibrations in the media. For materials that exhibit sharp absorption features, such as $\alpha-$monohydrate lactose, the absorption features are confined to distinct frequency bins specific to the constituents of the media, and therefore we assume the sharp absorption features can be expressed as higher order polynomial functions of frequency.

In the proposed approach, we consider a set of plane waves with angular frequency variable ω propagating through a sample of material with complex dielectric constant $n + ik$, with thickness d. The Fourier transform of the transmitted signal is given by:

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

where $X(\omega)$ is the Fourier transform of the signal before propagating through the sample, $n(\omega)$ represents the frequency dependent refractive index and $k(\omega)$ represents the frequency dependent extinction coefficient of the sample, which contains contributions from both absorption and scattering [6]. The total frequency dependent attenuation $\alpha(\omega)$ suffered by the signal

Manuscript received September 21, 2011; revised October 19, 2011; accepted October 28, 2011. Date of publication November 8, 2011; date of current version January 6, 2012.

M. Kaushik, B. W.-H. Ng, and D. Abbott are with the Department of Electrical and Electronic Engineering, University of Adelaide, Adelaide SA 5005, Australia (e-mail: mayank@eleceng.adelaide.edu.au; bwng@eleceng.adelaide.edu.au; dabbott@eleceng.adelaide.edu.au).

B. M. Fischer is with the Department of Electrical and Electronic Engineering, University of Adelaide, Adelaide SA 5005, Australia. He is also with the Institut Franco-Allemand de Recherches de Saint Louis, Saint Louis 68301, France (e-mail: Bernd.FISCHER@isl.eu).

Digital Object Identifier 10.1109/LPT.2011.2174980

during transmission through the sample is given by

$$\alpha(\omega) = 2k(\omega)\frac{\omega}{c}. \tag{2}$$

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

$$\alpha(\omega) = \alpha_{\mathrm{a}}(\omega) + \alpha_{\mathrm{s}}(\omega). \tag{3}$$

In this letter, we explore the relation between the change in spectral moments and time domain features of a THz pulse when it passes through a sample material and the frequency dependent scattering $\alpha_s(\omega)$ that occurs as a result of the interaction of the THz radiation with the internal structure of the sample material. In order to do this, we have to assume an analytical expression to describe the THz signal. The exact temporal waveform of the THz pulse and hence its spectral shape depends mainly on the generation and detection mechanism, which varies from one T-ray spectrometer to another. In our experiments the THz-TDS system uses photoconductive antennas (PCAs) for both the emitter and detector. One popular analytical model for the terahertz signal generated and detected using photoconductive antennas was given by Duvillaret et al. [10]. Their expression for the amplitude spectrum of the terahertz signal mostly depends on the carrier lifetime in the antenna's semiconductor material and the laser pulse duration. For calculating the moments of a given spectrum $S(\omega)$, the following general formula can be used:

$$m_{\mathrm{n}} = \frac{\int_0^\infty \omega^n S(\omega)\,\mathrm{d}\omega}{\int_0^\infty S(\omega)\,\mathrm{d}\omega}, \tag{4}$$

where n is the order of the moment.

For the analytical model given by Duvillaret et al. [10], the above equation for calculating the spectral moments does not have a closed form solution. Thus, this analytical model cannot be used for our purpose. An alternative analytical expression for modeling the THz spectrum was given by Xu et. al [11]. This expression is entirely dependent on only one parameter, the FWHM (Full Width at Half Maximum) pulse duration of the THz time domain signal τ_r, and is given by:

$$X(\omega) = A\frac{\omega\tau_{\mathrm{r}}}{\sqrt{2}}\exp(-\omega^2\tau_{\mathrm{r}}^2/4), \tag{5}$$

where A represents the maximum amplitude of the T-ray field, and ω represents the angular frequency.

In contrast to the previous model, this analytical expression is much simpler and has a definite solution for the calculation of moments using (4). In order to compare the accuracy of this model with the model proposed by [10], we calculate the center frequency from the first spectral moment of experimentally observed amplitude spectrum of a reference measurement and compare it with the amplitude spectrum obtained from both the analytical expressions. We also compare the mean squared error (MSE) and correlation coefficient (CC) for each analytical model with the measured spectrum. The results are tabulated in Table I and Table II.

TABLE I
COMPARISON OF CENTER FREQUENCY OF ANALYTICAL MODELS AND EXPERIMENT OBSERVATIONS

Model 1 [10]	Model 2 [11]	Experiment
0.437 THz	0.426 THz	0.431 THz

TABLE II
MSE AND CC: ANALYTICAL MODELS AND MEASURED SPECTRUM

Parameter	Model 1 [10]	Model 2 [11]
MSE	0.0035	0.0042
CC	0.925	0.905

The above results show that both models explain the experimental observations to a similar level of accuracy, with the model given by Duvillaret et al. [10] showing slightly better MSE and CC parameters than the model used by Xu et al. [11]. Given the similarity in these results, it is reasonable to assume (5) to be a good approximate representation of the reference THz signal (i.e. without sample). Now using (4), the first moment for $X(\omega)$ is given by $\mu_{\mathrm{r}} = \sqrt{\pi}/\tau_{\mathrm{r}}$.

When we introduce the sample to this system, the THz signal is attenuated due to the scattering and absorption of the signal by the sample. If the frequency dependent absorption features of the sample are sharp and sparse, they do not greatly influence the center frequency of the sample spectrum and the center frequency mostly depends on the frequency dependent scattering from the sample. Modeling the scattering as a linear function of propagation length d and a quadratic over the given range of angular frequency ω, the expression for the scattering response of the sample, $Y_s(\omega)$, can be given by:

$$|Y_{\mathrm{s}}(\omega)| = A\exp(-\alpha_s d\omega^2)\frac{\omega\tau_{\mathrm{r}}}{\sqrt{2}}\exp(-\omega^2\tau_{\mathrm{r}}^2/4). \tag{6}$$

This further reduces to

$$|Y_{\mathrm{s}}(\omega)| = A\frac{\omega\tau_{\mathrm{r}}}{\sqrt{2}}\exp(-\omega^2\tau_{\mathrm{s}}^2/4), \tag{7}$$

where $\tau_{\mathrm{s}}^2 = \tau_{\mathrm{r}}^2 + 4\alpha_s d$, and the first moment of $Y_{\mathrm{s}}(\omega)$ is given by $\mu_{\mathrm{s}} = \sqrt{\pi}/\tau_{\mathrm{s}}$.

Thus it can be seen that the spectral shape of the attenuated pulse remains unchanged. However, the scattering from the sample has changed the distribution characteristic parameter τ_{r} to τ_{s} or we can say that the FWHM duration of the time domain pulse has increased from τ_r, for the reference THz signal to τ_{s}, for the sample THz signal. In terms of the frequency domain, we can say that, the mean (center frequency) of the reference spectrum μ_{r} has changed to μ_{s} for the sample spectrum. Similar observations were made by Dines and Kak [12], where they estimated attenuation of soft biological tissues using ultrasound waves. They treated the attenuation as linearly dependent on the frequency and assumed a Gaussian distribution for modeling the signal spectrum; furthermore they assumed scattering to be minimal in their experiments.

Once the means of the reference signal and the transmitted sample signal are obtained from the above equations, the unknown scattering attenuation profile $\alpha_s(\omega)$ can be calculated

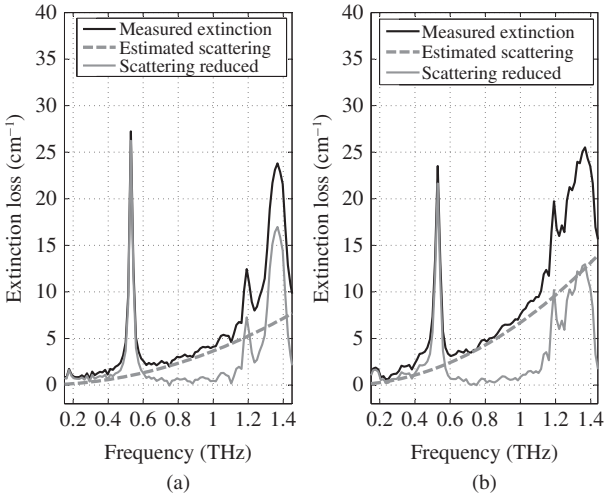

Fig. 1. Extinction loss spectra of: (a) sample pellet made of α-monohydrate lactose mixed with PE powder with avg. dia. 60 μm (black) and (b) sample pellet made of α-mono hydrate lactose mixed with PE powder with avg. dia. 360 μm (solid black), with their respective estimated scattering loss profiles (dashed gray) and scattering mitigated spectrum (solid gray).

from the first order spectral moments of the measurements:

$$\alpha_{\rm s}(\omega) = \frac{\pi}{4d}\frac{\mu_{\rm s}^2 - \mu_{\rm r}^2}{\mu_{\rm r}^2\mu_{\rm s}^2}\omega^2, \tag{8}$$

or from the FWHM pulse durations of the measurements:

$$\alpha_{\rm s}(\omega) = \frac{1}{4d}(\tau_{\rm s}^2 - \tau_{\rm r}^2)\omega^2. \tag{9}$$

Therefore, a complete scattering loss profile can be estimated for any given media using only the first spectral moments or FWHM pulse durations of the measured signals. Using a standard THz transmission spectroscopy setup, we carried 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 is comprised of PE powder with average grain diameter of 60 μm, while the average grain diameter for the PE powder of the second sample was 360 μm.

Assuming that the reflections are removed from the sample and reference data, the frequency dependent refractive index $n_{\rm s}(\omega)$ of the sample and total attenuation $\alpha(\omega)$ is given by:

$$n_{\rm s}(\omega) = n_0 - \frac{c}{\omega d}\angle H(\omega), \tag{10}$$

$$\alpha(\omega) = \frac{2}{d}\left\{\ln\left[\frac{4n_{\rm s}(\omega)n_0}{(n_{\rm s}(\omega)+n_0)^2}\right] - \ln|H(\omega)|\right\}, \tag{11}$$

where n_0 is the refractive index of free air and $H(\omega)$ is the transfer function of the sample, given by:

$$H(\omega) = \frac{E_{\rm sam}}{E_{\rm ref}}, \tag{12}$$

where $E_{\rm ref}$ and $E_{\rm sam}$ are the experimentally measured reference and sample spectrum, respectively.

III. Results

The results of our approach are shown in Fig. 1. The samples clearly show characteristic absorption features of lactose at 0.53 THz, 1.19 THz and 1.39 THz. It should be noted that along with the distinct absorption features, both samples show a baseline slope owing to scattering from the samples' granular nature. The technique described in this letter estimates the baseline slope owing to the scattering of the THz signal. The reduced scattering extinction spectrum for each sample is obtained by subtracting the estimated scattering loss, computed using (9), from the measured attenuation loss in (11). As expected, the sample with larger PE particles shows a greater baseline slope as compared to the sample with smaller PE particles, indicating stronger scattering.

IV. Conclusion

In conclusion, we report an extension of the centroid shift technique in the terahertz regime, initially proposed by Dines and Kak [12] for estimating frequency dependent attenuation of soft biological tissue using ultrasound. The method was tested on two sample pellets comprised of α-monohydrate lactose and PE with different granularity in each sample. As seen in from the results, the technique reasonably estimates scattering loss profile for both the cases without using any *a priori* information of the physical characteristics of the sample or the material, which makes it very useful for mitigating scattering effects in real world applications such as standoff measurements. It must be noted that the scattering estimate provided by this technique assumes that the material under study has sharp and sparse absorption features, which is usually the condition required for a THz spectral fingerprint.

References

[1] M. Herrmann, M. Tani, M. Watanabe, and K. Sakai, "Terahertz imaging of objects in powders," *Proc. Inst. Elect. Eng. Optoelectron.*, vol. 149, no. 3, pp. 116–120, Jun. 1, 2002.

[2] T. L. J. Chan, J. E. Bjarnason, A. W. M. Lee, M. A. Celis, and E. R. Brown, "Attenuation contrast between biomolecular and inorganic materials at terahertz frequencies," *Appl. Phys. Lett.*, vol. 85, no. 13, pp. 2523–2525, Sep. 1, 2004.

[3] A. Bandyopadhyay, *et al.*, "Effects of scattering on THz spectra of granular solids," *Int. J. Infrar. Millimeter Waves*, vol. 28, no. 11, pp. 969–978, 2007.

[4] L. M. Zurk, B. Orlowski, D. P. Winebrenner, E. I. Thorsos, M. R. Leahy-Hoppa, and L. M. Hayden, "Terahertz scattering from granular material," *J. Opt. Soc. Amer.*, vol. 24, no. 9, pp. 2238–2243, 2007.

[5] M. Franz, B. M. Fischer, and M. Walther, "The Christiansen effect in terahertz time-domain spectra of coarse-grained powders," *Appl. Phys. Lett.*, vol. 92, no. 2, pp. 021107-1–021107-3, Jan. 15, 2008.

[6] Y. C. Shen, P. F. Taday, and M. Pepper, "Elimination of scattering effects in spectral measurement of granulated materials using terahertz pulsed spectroscopy," *Appl. Phys. Lett.*, vol. 92, no. 5, pp. 051103-1–051103-3, Feb. 1, 2008.

[7] G. Mie, "Beiträge zur optik trüber medien, speziell kolloidaler metallösungen," *Ann. Phys.*, vol. 330, no. 3, pp. 377–445, 1908.

[8] M. Kaushik, B. W.-H. Ng, B. M. Fischer, and D. Abbott, "Mitigating scattering effects in THz-TDS measurements," in *Proc. 35th Int. Conf. Infr., Millimeter Terahertz Waves*, Rome, Italy, Sep. 2010, pp. 1–2.

[9] P. A. Narayana and J. Ophir, "A closed form method for the measurement of attenuation in nonlinearly dispersive media," *Ultrason. Imaging*, vol. 5, no. 1, pp. 17–21, Jan. 1983.

[10] L. Duvillaret, F. Garet, J.-F. Roux, and J.-L. Coutaz, "Analytical modeling and optimization of terahertz time-domain spectroscopy experiments, using photoswitches as antennas," *IEEE J. Sel. Topics Quantum Electron.*, vol. 7, no. 4, pp. 615–623, Jul./Aug. 2001.

[11] J. Xu, T. Yuan, S. Mickan, and X.-C. Zhang, "Limit of spectral resolution in terahertz time-domain spectroscopy," *Chin. Phys. Lett.*, vol. 20, no. 8, pp. 1266–1268, Aug. 2003.

[12] K. A. Dines and A. C. Kak, "Ultrasonic attenuation tomography of soft tissues," *Ultrason. Imaging*, vol. 1, no. 1, pp. 16–33, Jan. 1979.