Overview of Data Analysis Techniques of Frequency-Domain Terahertz Spectroscopy

Blaž Pongrac, Dušan Gleich
University of Maribor, Faculty of electrical engineering and computer science, Koroška cesta 46, 2000 Maribor, Slovenia
blaz.pongrac1@um.si

***Abstract*—Chemical analysis is widely used in different fields. It is based on spectroscopy and different methods. One of the upcoming technology is terahertz (THz) spectroscopy, but it is not yet well researched. This paper presents opto-electrical methods for generating THz waves and it presents frequency-domain spectroscopy as the possible solution to problems, that are presented in infrared, visible-light, ultraviolet, x-ray and gamma spectroscopies. Basic analysis techniques are presented, including deriving absorption coefficient and refractive index. Both are interconnected with Kramer-Kronig relation and are fundamental sample characteristics that can be measured with spectrometers. Example of measuring transmittance and refractive index are presented in experiment, made with frequency-domain spectroscopy platform TeraScan 1550.**

***Keywords*—Terahertz; Frequency-Domain; Spectroscopy; Data Analysis; Absorption; Refractive index**

I. INTRODUCTION

Chemical analysis is becoming one of the most important tools for material analysis in different fields, such as environment protection, agriculture, food industry, safety, pharmaceutical and medicine. Leading method for material and substance analysis is spectroscopy. Spectroscopy is study of impact of electromagnetic radiation on the substance. It originated from studying of visual light dispersion to its wavelengths. Now, spectroscopy is divided by type of radiative energy and frequency band or wavelength regions it is operating on. Electromagnetic radiation energy is commonly available and widely used from spectrometers to radars.

Spectroscopy based on electromagnetic energy is divided in microwave, terahertz, infrared, visible light, ultraviolet, x-ray and gamma spectroscopy. Microwave spectroscopy is suitable for analyzing bigger targets. Good example of microwave spectrometry is ground penetrating radar or GPR. By sending microwaves into the ground and analyzing their reflection, one can determine structure of the ground and can detect objects buried in the ground. Gamma, x-ray and ultraviolet spectroscopies are able to measure nanometer to sub-nanometer particles, but because they uses ionizing waves, they are invasive and dangerous for biological matter. Visual light spectroscopy is commonly used for biological matter analysis, but it has one major drawback – it is opaque for dielectrics, therefore dielectric materials must be dissolved in fluids. Based on electromagnetic radiation characteristic, both infrared and terahertz spectroscopies offer some solutions to the problems all other spectroscopies have. Nevertheless, we yet do not know, what can be achieved with terahertz spectroscopy, since it is not well known field.

Terahertz (THz) frequency band is located between microwaves and far-infrared spectrum (100 GHz – 10 THz) [1]. THz waves are considered non-invasive, since they are non-ionizing waves. They are suited for measuring content of the sample of dielectric and biological materials and are opaque for metals. Most materials, such as paper, cardboard, textile and plastics, have spectral footprints in THz band. In addition, THz wave propagation is sensible to gases, which makes THz spectroscopy suitable for gas detection.

Generating and detecting terahertz waves is challenging. In later years, new methods were developed. From the three methods, we can recognize for generating/detecting terahertz waves (optical method, opto-electrical method and electrical method), opto-electrical is the most promising. It combines advantages from both, optical and electrical methods. THz waves can be generated in whole THz spectrum relatively high bandwidth (up to few THz), which we cannot say for electrical methods (limited by frequency limitations of semiconductors [2]) and optical methods [3] (limited by cooling of the lasers used in generating THz waves [1]). In addition, technology costs less, because more common and cheaper components can be used.

II. METHODS OF GENERTING THZ WAVES

There are two opto-electrical methods of generating THz waves, from which we can derive two spectroscopies: time-domain spectroscopy and frequency-domains spectroscopy. Basic principle is the same for both methods. At its hearth, there are photoconductive antennas (PCA). Photoconductive antennas are built on photoconductive substrates with subpicosecond carrier lifetime. One of such substrate is InGaAs or InGa(Al)As. Optical pulse is directed onto photoconductive antenna. When it hits the photoconductive substrate, it propagate into substrate, where it is absorbed and photocarriers are generated. Bias field accelerates photocarriers and transient photocurrent is generated, which is driving dipole antenna and emits THz pulse. Basic principle is presented on Figure 1.

For receiving THz wave, photoconductive antenna can also be used. Only this time, it should not be voltage biased, but same optical pulse as the one directed onto emitting antenna should be directed onto receiving one. When THz pulse hits the receiving photoconductive antenna, it overlaps with photocarriers, generated by optical signal. Photocurrent is generated and it can be measured.

978-1-7281-2181-9/19/$31.00 ©2019 IEEE

Basic setup for time-domain spectrometer and frequency-domain spectrometer are presented in Figure 2 and Figure 3, respectively. Time-domain spectrometer is build around femtosecond laser. Emitted laser pulse is split into two beams. First one is directed to emitting photoconductive antenna, second one is directed through adjustable delay stage to receiving photoconductive antenna. With the delay sweep, emitted and received signals can be convoluted. Frequency domain spectrometer is built around two DFB lasers, which emits optical signals with different wavelengths, and optical coupler. Both optical signals are mixed in optical coupler and directed to the emitting and receiving antenna. Mixed optical signal can be perceived as train of optical pulses. Lock-in detection amplifier can be used for detecting pulses.

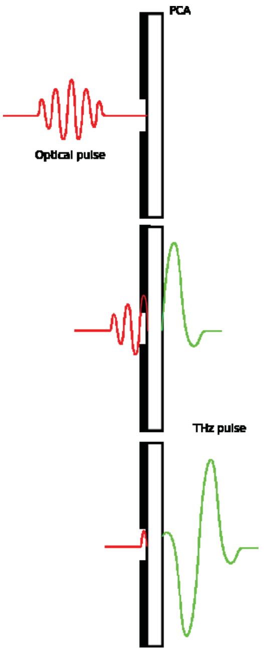

Figure 1. Principle of generating THz waves with PCA [4]

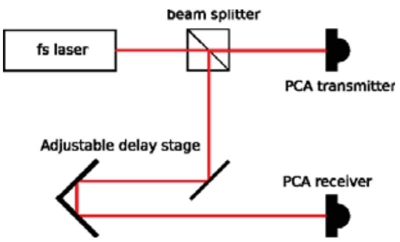

Figure 2. Time-domain THz spectrometer structure [9]

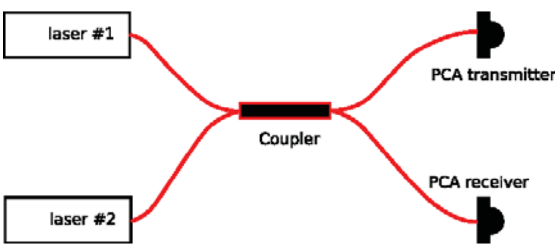

Figure 3. Frequency-domain THz spectrometer structure [9]

Photoconductive antennas are small in size and are not omnidirectional. However, they emit poorly directed beam. One solution for that problem is to collimate the beam with mirrors. Collimation of the terahertz beam can be performed based on the application. If the application predicts measuring THz waves that propagate through measured sample (transmission spectroscopy), setup as presented in Figure 4a must be used. If the application predicts measuring THz waves that are reflected from the surface of measured sample (reflection spectroscopy), setup as presented in Figure 4b must be used.

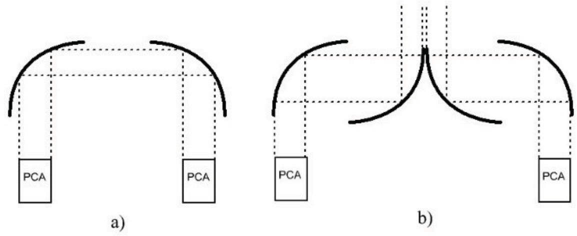

Figure 4. Two different methods for collimation of THz wave: a) transmission spectroscopy, b) reflection spectroscopy

III. FREQUENCY DOMAIN SPECTROSCOPY

Frequency-domain spectroscopy is capable of measuring frequency depended characteristic of certain material. It works like step frequency continuous wave radar – spectrometer generates THz wave of certain frequency and measures amplitude and phase of the received signal. Based on the collimation techniques, presented in chapter Methods of generating THz waves, experimental setup for frequency-domain spectroscopy should look like one in Figure 5.

Experimental setup, used in the experiment in this paper, is build around Toptica Photonic's TeraScan 1550 frequency-domain spectroscopy platform and its proprietary software. It has two tunable 1550 nm DFB lasers (one is cooled, other is heated in order to achieve difference in wavelengths), InGaAs photoconductive antennas and frequency range from -50 to 1210 GHz (2.7 THz is reachable with extension). Setup is presented in Figure 5. THz beam is collimated with Thorlabs tunable parabolic mirrors in transmission spectroscopy setup. THz optics are presented in Figure 6.

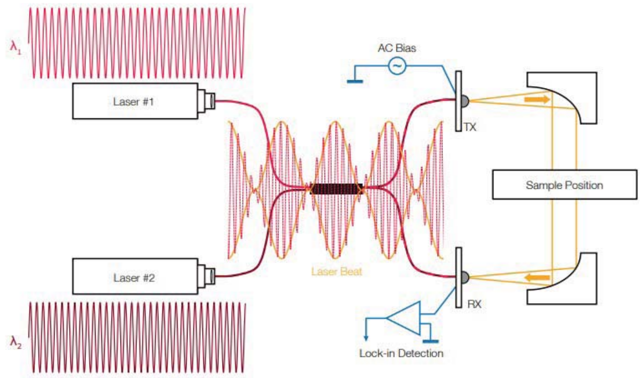

Figure 5. TeraScan 1550 platform configuration, typical configuration for transmission spectroscopy [5]

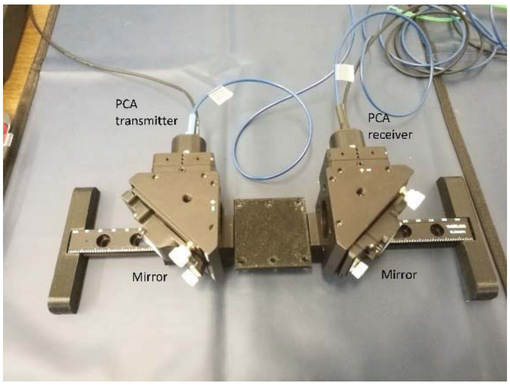

Figure 6. THz optical components setup

IV. Data analysis methods

Data analysis methods for transmission spectroscopy are mainly based on measuring absorption and transmittance. Raw measured data in frequency domain are phase fringes of received signal. With basic mathematical operations, we can determine measured photocurrent envelope. With comparing data to reference measurement, we can calculate transmittance, which is calculated with Lamper-Beer law (1), where I_{ref} is reference signal (photocurrent) and I_m is measured signal (photocurrent). From transmittance, absorbance A and from it absorption coefficient α can be calculated (2).

$$T = \frac{I_m}{I_{ref}} = 10^{-A} \tag{1}$$

$$\alpha = \frac{2.303 \cdot A}{d} \tag{2}$$

From phase fringes we can also determine phase of the received signal, from which phase length can be derived. From difference in phase length of measured spectrum and of reference spectrum (optical path length, OPL) and thickness of measured sample d, refractive index can be calculated (3).

$$n = \frac{OPL}{d} \tag{3}$$

Refractive index and absorption coefficient are linked with Kramers-Kronig relation, which states, that both coefficients are linked together as they represent real and imaginary part of the same function [7]. In case of absorption coefficient and refractive index that function is complex dielectric constant. (4) is presenting relation between refractive index n and absorption coefficient α, where c is speed of light, γ is Cauchy principal value and Ω is angular frequency.

$$n[\omega] = 1 + \frac{c}{\pi}\gamma \int_0^{+\infty} \frac{\alpha(\omega)}{\Omega^2 - \omega^2} d\Omega \tag{4}$$

Further analysis methods are divided into identification techniques and quantization techniques. Identification technics consist of feature comparison between reference database and measured material, which can result in substance detection, but are unable to quantify substance in the sample. One of widely used identification techniques is measuring absorption in material and comparing it to the samples from database [6]. [8] is proposing using chemometrics methods, such as principal component analysis and even provides example. Quantization techniques are able to detect and to quantify substance in measured sample. They can be based on preliminary calibration or modeling of the THz spectrum.

V. Experimental setup and results

In order for later development of data processing techniques, one must first understand basic operations of calculating absorption coefficient and refractive index from measured phase fringe in frequency-domain spectroscopy. Simple experiment was derived to do that.

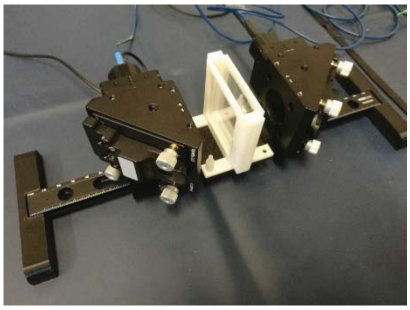

Figure 7. Casing for granular and powdered samples with thin plastic film as walls

As the measured sample, mixed soil was used. For the purpose of the experiment, casing for granular and powdered samples was designed and 3D printed, which is presented in Figure 7. Casing is 10 mm wide. Thin plastic film is used on the sides. Characteristics of the casing were measured and are presented in Figure 8. Calculated refractive index of the casing walls was close to 1 while thickness of both walls is well below 1 mm. Because of that, $n = 1$ was used for further reference.

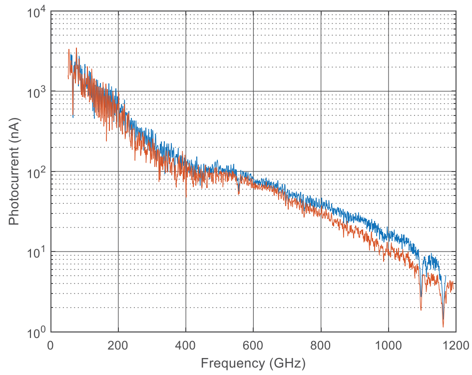

Figure 8. Reference amplitude spectrum (blue) and amplitude spectrum of empty casing (orange)

Measurement of the soil spectrum was performed in frequency range from 50 to 1200 GHz with resolution of 0.05 GHz, sample compare of 15 samples and integration constant of 5.04 ms. Figure 9 is presenting experimental results. We can determine, that mixed soil samples 10 mm wide is opaque for frequency above 400 GHz. Nevertheless, below that value, results are good enough for further processing.

Calculated refractive index and transmittance are presented on Figures 10 and 1, respectively. We can conclude, that our sample really is opaque for the frequencies above 400 GHz, since transmittance is almost zero and refractive index is at least ten times bigger.

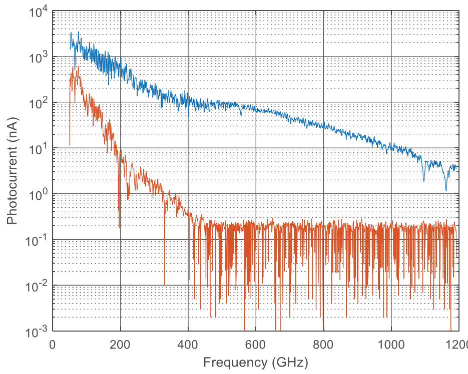

Figure 9. Amplitude spectrum of empty casing (blue) and soil amplitude spectrum (orange)

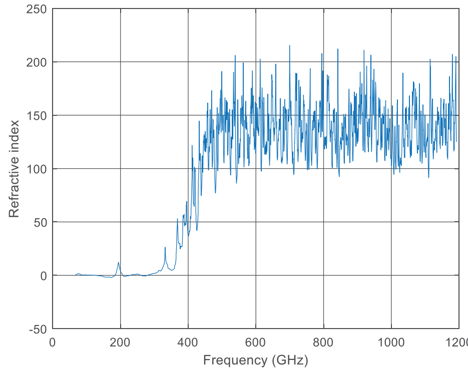

Figure 10. Refractive of the soil sample

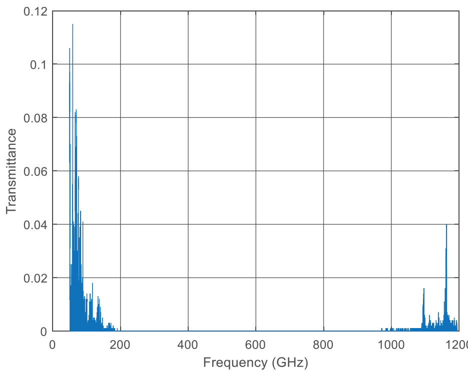

Figure 11. Transmittance of the soil sample

VI. Conclusion

Terahertz spectrometry is showing great promise in analyzing material samples, since THz wave have wavelengths in millimeter and submillimeter range and are non-ionizing (do not affect biological tissue). Opto-electrical methods for generating THz waves were presented. Both methods are based on photoconductive antennas, difference is only in THz wave detection. Time-domain spectrometer uses delay line and convolution, while frequency domain spectrometer uses two lasers, optical mixer and lock-in amplifier for measuring amplitude and phase.

Basic processing methods for frequency-domain spectroscopy were presented. Raw data, measured with frequency domain spectrometer are phase modulated fringes. By calculating fringes envelope, one can calculate amplitude spectrum. Two fundamental characteristics that can be measured with frequency-domain spectrometer are absorbance/absorption coefficients and reflectance/refraction index. They both represent complex dielectric function and are interconnected with Kramer-Kronig relation.

In order to present both fundamental characteristics, experiment was designed and carried out. Most of the problems come from collimated beam calibration and from temperature instability of frequency-domain system. Nevertheless, the experiment offers great foundation on which later experiment can be build.

In future work, both identification and quantization methods will be tested. In order to achieve that, database of absorption coefficients and refractive indexes should be constructed. Database will offer us methods of fast application development. It would also be necessary to search and to test for other data processing methods, that are used in other spectroscopies, mainly infrared spectroscopy. Both, constructing the database for substances and implementation of spectroscopic algorithms are next stage in our research and are currently being realized.

Acknowledgment

This research was carried out and made possible within Slovenian Research Agency project number J7-9408.

References

[1] M. Lang and A. Deninger, “Laser-based terahertz generation and aplication,”, in Photonik international, Fellbach, vol. 4/2012, pp. 36-38, November 2012

[2] U. K. Mishra and J. Singh, Semiconductor Device Physics and Design, Dordrecht: Springer Science ‚ Business Media, pp. 415-416

[3] A.A Danylov, T. M. Goyette, J. Waldman, M. J. Coulumbe, A. J. Gatesman, R. H. Giles, X. Qian, N. Chandrayan, S. Vangala, K. Termkoa, W. D. Goodhue and W. E. Nixon, “Terahertz inverse synthetic aperture radar (ISAR) imaging with a quantum cascade laser transmitter”, in Optics Express 18(15):16264-72, July 2010

[4] N. M. Burford and M. O. El-Shenawee, “Review of terahertz photoconductive antenna technology”, in Optical Engineering 56(1), 010901, January 2017

[5] Terahertz Technologies, Systems and Accessories: Product Brochure, Toptica Photonics

[6] A. Roggenbuck, H. Schmitz, A. Deninger, I. C. Mayorga, J. Hamberger, R. Güsten and M. Grüninger, “Coherent broadband continuous-wave terahertz spectroscopy on solid-state samples”, in New Journal of Physics 12, 043017 (13pp), April 2010

[7] C. Bédard, A. Destexhe, “Kramers-Kronig Relations and the Properties of Conductivity and Permittivity in Heterogeneous Media”, in Journal of electromagneticAnalysis and Applications, Vol.10 No.2, 2018.

[8] J. El Haddad, B. Bousquet, L. Canioni and P. Mounaix, “Review in terahertz spectral analysis”, in Trends in Analytical Chemistry, Elsevier, 2013, 44, pp. 98-105

[9] B. Pongrac, D. Gleich, “Overview of Opto-electrical Terahertz Spectroscopy”, in IWSSIP 2019 international conference, “unpublished”