Homeland security enforcement using novel terahertz technology · IV
–Final report–

This is the final report for Grant FA2386-08-1-4055 AOARD 084055. The main aspects of the project — aim, theory, experiments and results — will be presented.

On behalf of my colleagues and myself, I would like to thank AOARD for continuing to support our research.

Contents

1. Project

This project aims to find a method of identifying liquid samples by using terahertz radiation. Achieving this goal would have an impact on various activities, such as homeland security, pharmaceutical, food industry and other fields where it is important to identify liquids and/or analyze their composition.

Based on our previous experience with the interaction between the terahertz radiation and liquids, we proposed an approach relying on silicon prism device. Undoped silicon is transparent in the terahertz range; a prism made of such silicon can be used to manipulate the terahertz waves to make them interact with a liquid layer placed on one of the prism surfaces. By analyzing the changes that follow this interaction, data can be obtained about the optical properties of the liquid, which in turn allow the identification of the liquid and the measurement of other of its properties, such as concentration.

The results will be detailed below, in a separate section. Here are the main points at the end of the current stage:

1. Increasing the measurement accuracy requires a longer measurement time or a larger prism. To allow for practical real-time applications we chose the second option.

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14. ABSTRACT
This project aimed to find a method of identifying liquid samples using terahertz radiation. By obtaining optical properties of the liquid, it would be possible to identify the liquid and other properties, such as concentration.

15. SUBJECT TERMS

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2. Formulas that transform measured data into complex refraction indices can be obtained, but a more suitable way is to use computer calculations.

3. Tests were performed on several liquids and no overlaps have been observed. If by continuing the measurements on other liquids such overlaps are found, then another terahertz frequency can be used to distinguish between liquids.

4. In the current configuration, measuring liquid samples enclosed in plastic containers is still difficult.

2. Theory

A. Principle

The optical properties of the liquid sample that can be obtained by this approach are the refraction index and the absorption coefficient. If these two parameters — or another pair of parameters related to them — are measured at one or more frequencies, the liquid can be identified by comparing them with those of other substances in a database.

To measure its optical properties, the liquid sample is placed on a surface of a prism made of undoped prism, that has high electric resistivity, which means that it is transparent to the terahertz waves. This high-purity silicon is the most transparent material in the terahertz range.

A parallel terahertz beam is passed through the prism, as shown in Figure 1. At the interface between the prism and the sample, a partial reflection takes place, which changes the intensity and the polarization properties of the wave depending on the optical parameters of the liquid. If no sample is placed on the prism, then the reflection is total, meaning none of the incoming energy is lost above the surface; when a liquid is present, part of this energy is absorbed in the liquid. Additionally, the polarization state of the wave is changed. By comparing the measurements (with and without sample), the optical constants of the liquid can be determined.

Each liquid, at each electromagnetic frequency, has a specific complex refraction index, written as

$$n = n_{re} - i \cdot n_{im} \quad (1)$$

where n_{re} is the usual refraction index and n_{im} reflects the absorption properties of the substance. The pair (n_{re} n_{im}) is then characteristic for each liquid, and by measuring it we can identify liquids.

B. Method

After considering several methods of measuring the effect of the liquid sample on the terahertz wave, we decided that the most promising would be the so-called *ellipsometric technique*. Conventionally, the ellipsometry is used mostly in the visible range of the electromagnetic spectrum, in applications such as analyzing the structure of complex thin

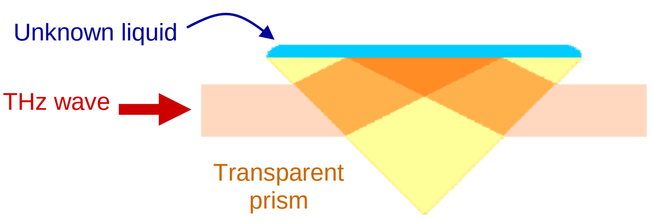


Fig. 1. Schematic of the measurement principle. The terahertz wave is reflected at the interface between the prism and the liquid in a way that depends on the liquid's optical properties.

films.

In principle, the ellipsometry consists in using two polarizers: one with a fixed angle placed before the interaction with the sample, and one rotatable, placed after the sample, for analyzing the polarization state of the wave. In general, after the interaction takes place, the polarization state of the wave is elliptical (hence the name given to the method) and the optical properties of the sample are reflected in the parameters of this elliptical polarization.

At the interface between the prism and the sample, the internal reflection is given by the following amplitude reflection coefficients, for each polarization (parallel and perpendicular):

$$r_{\parallel} = \frac{n_1 \cos\theta_0 - n_0 \cos\theta_1}{n_1 \cos\theta_0 + n_0 \cos\theta_1}$$
$$r_{\perp} = \frac{n_0 \cos\theta_0 - n_1 \cos\theta_1}{n_0 \cos\theta_0 + n_1 \cos\theta_1} \qquad (2)$$

where subscripts 0 and 1 represent the prism and the sample, respectively, and the quantities have the following meanings: the n's are the complex refraction indices, and the θ's are the complex angles inside each medium.

The refraction indices and the angles are linked through the Snell's law (all quantities are complex numbers):

$$n_0 \sin\theta_0 = n_1 \sin\theta_1 \qquad (3)$$

The ellipsometric technique is applied as follows. The input beam is polarized at 45°, that is, halfway between the parallel and the perpendicular polarization. The output beam is passed through a second polarizer, whose orientation can be adjusted. By rotating the second polarizer, a sine-shaped intensity signal is recorded:

$$I = I_0 \left(1 + a \cdot \cos 2\alpha + b \cdot \sin 2\alpha\right) \qquad (4)$$

The ellipsometric parameters Ψ and Δ are calculated using the relations:

$$\cos 2\Psi = -a$$
$$\cos\Delta = \frac{b}{\sqrt{1 - a^2}} \qquad (5)$$

Finally, the sample refraction index is calculated:

$$n_1 = n_0 \frac{\sin\theta_0}{\sqrt{1 + \left(\frac{1-\rho}{1+\rho}\right)^2 \tan^2\theta_0}} \qquad (6)$$

where ρ is defined as

$$\rho = \tan\Psi \cdot e^{i\Delta} \qquad (7)$$

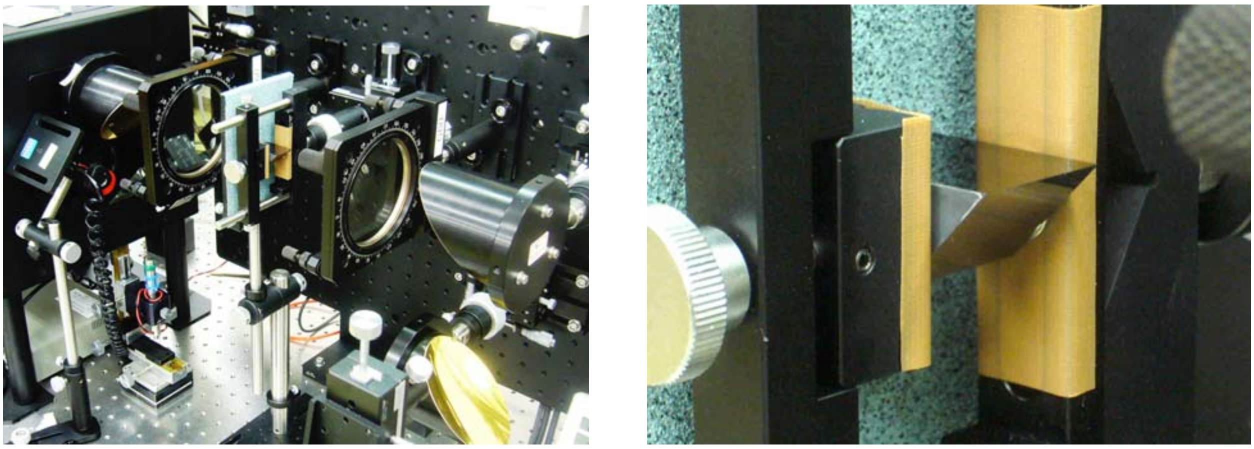
In practice, the input and output surfaces of the prism intervene with their own additional effects upon the polarization state of the beam, and these must be taken into account in calculating the refraction index.

3. Experiments

The experimental setup consists of the following parts:

- the source, a backward-wave oscillator, which produces terahertz waves with a frequency adjustable between approximately 400 and 700 GHz;
- a chopper for modulating the terahertz beam, to increase the signal-to-noise ratio by the use of a lock-in amplifier;
- collimation and focusing optics, consisting in four gold-plated off-axis parabolic mirrors;
- a plane dichroic mirror, made of indium-tin oxide on a glass substrate, to combine a guiding visible beam with the terahertz beam, which facilitates alignment and adjustments;
- the silicon prism;
- two polarizers, made of free-standing wire grids, to polarize and then analyze the polarization state;
- a room-temperature DLATGS pyroelectric sensor, to detect the terahertz radiation.

Additionally, the setup includes several electronic devices and a computer for controlling and measurement, as well as several other secondary accessories.



(a) (b)

Fig. 2. (a) The optical setup, excluding the detector, showing the placement of the silicon prism between polarizers, in the collimated beam. (b) The silicon prism.

4. Results

To verify the theory and confirm that different liquids have indeed different effects on the terahertz wave, preliminary measurements were performed. Three liquids — water, ethanol, and their mixture in equal volumes — were placed on the silicon prism, one after another. The angle of the second polarizer was changed in four equally spaced positions and the intensity at the detector was recorded. The data were processed as shown in Figure 3.

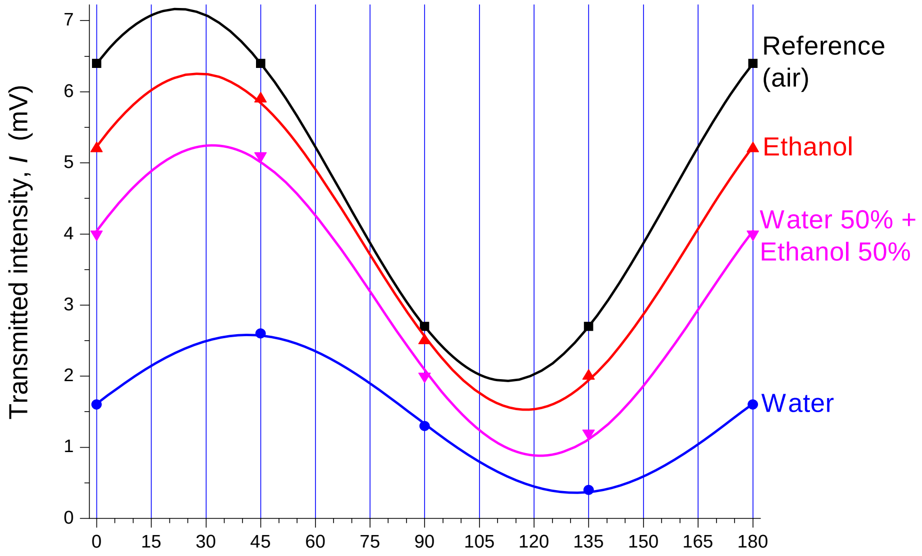


Fig. 3. Test measurements. Each curve represents the detected signal intensity as a function of the output polarizer angle. The continuous lines are best fits to the experimental data, which are represented by dots. (The data at 180° are identical with those at 0°, as the polarizer behaves identically at these angles.)

An important setback in the progress of our research was the finding that the signals depend strongly on the optical alignment of the setup. Figure 4 shows the effect. After the measurements labeled “Air 1”, “Ethanol”, and “Water” were taken, the setup was realigned and another reference measurement was taken with air as sample. This is shown in the same figure as “Air 2”, clearly in a displaced position from “Air 1”.

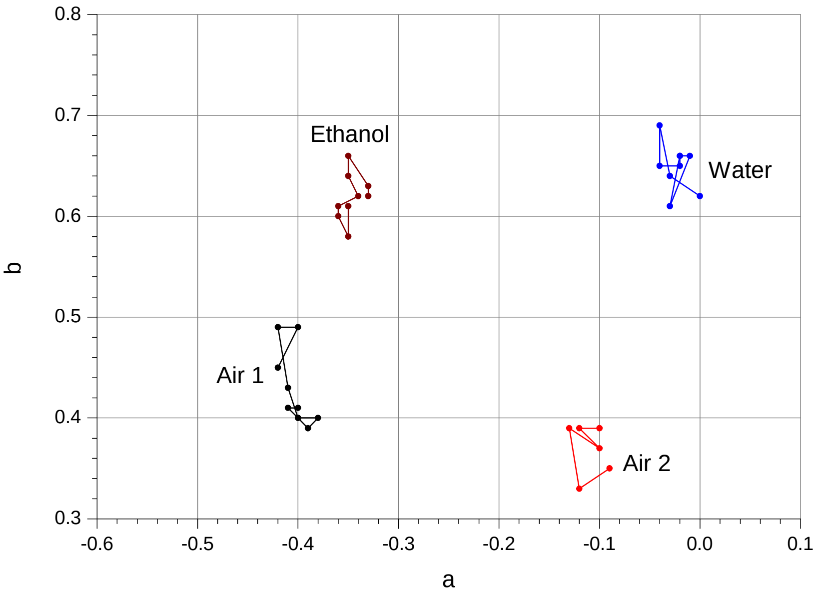


Fig. 4. Processed results obtained from sine waves such as those in Fig. 3. This map of the coefficients a and b from Eq. 4 allows a visualization of different liquids and measurements as clouds of points.

The reason for this strong dependence is still under investigation. One way of finding it is experimental, by checking the dependence between the prism position and the parameters *a* and *b*. Another is by computer simulation, which requires building a program that follows step by step the behavior of the terahertz wave and calculates the changes of the output beam depending the prism alignment.

5. Publications

The results obtained during this research were published as:

■ Adrian Dobroiu, Chiko Otani, "Ellipsometric measurements on liquids in the terahertz range," Extreme Photonics Symposium 2009, Wako, Japan, May 20-21, 2009

They are also being prepared to be presented as:

■ Adrian Dobroiu, Chiko Otani, "Ellipsometry in the terahertz range for liquid identification," The 34th International Conference of Infrared, Millimeter, and Terahertz Waves, Busan, Korea, September 21-25, 2009

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Poster presented at the Extreme Photonics Symposium 2009:

Extreme Photonics
Wako, May 2009

Ellipsometric measurements on liquids in the terahertz range

Adrian Dobroiu, Chiko Otani
RIKEN, Terahertz Sensing and Imaging Laboratory

Purpose

- Liquid identification
- Liquid analysis
- Concentration measurement of solutions

The technique could be applied wherever we need to identify or analyze liquids, for example in homeland security, chemistry, food production, pharmacy, etc.

We have described a similar technique, but with a different principle, in:

A. Dobroiu, R. Beigang, C. Otani, and K. Kawase, "Monolithic Fabry-Perot resonator for the measurement of optical constants in the terahertz range," *Applied Physics Letters* **86**, 231107 (2005)

Theory

A terahertz wave is sent through a transparent silicon prism. In the absence of the liquid sample, a total reflection occurs on the upper surface. The output beam intensity and polarization are measured for reference.

When a sample is placed on top of the prism, the total internal reflection becomes partial reflection, and the intensity and polarization state of the wave are changed, allowing the measurement of the liquid optical constants.

The complex refraction index is $n_{re} - i \cdot n_{im}$, where n_{re} is the usual refraction index and n_{im} is proportional to the absorption coefficient. The pair (n_{re}, n_{im}) depends on the frequency and is characteristic for each liquid.

At the interface between the prism and the sample, the internal reflection is given by the following reflection coefficients, for each polarization (parallel and perpendicular):

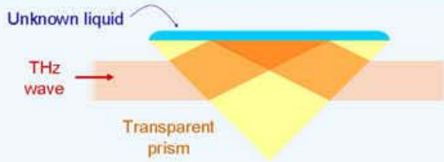
$$r_{\parallel} = \frac{n_1 \cos\theta_0 - n_0 \cos\theta_1}{n_1 \cos\theta_0 + n_0 \cos\theta_1} \qquad r_{\perp} = \frac{n_0 \cos\theta_0 - n_1 \cos\theta_1}{n_0 \cos\theta_0 + n_1 \cos\theta_1}$$

where

- the n's are the complex refraction indices,
- the θ's are the complex angles inside each medium, and
- subscripts 0 and 1 represent the prism and the sample, respectively.

The refraction indices and the angles are linked through the Snell's law, in complex quantities:

$$n_0 \sin\theta_0 = n_1 \sin\theta_1$$



The refractive index of the sample can be measured by analyzing the polarization state of the output beam. We propose using an **ellipsometric technique**:

1. The input beam is polarized at 45°, halfway between the parallel and the perpendicular polarization.

2. The output beam is passed through a second polarizer, whose orientation can be adjusted.

3. By rotating the second polarizer, a sine-shaped intensity signal is recorded:

$$I = I_0 (1 + a \cdot \cos 2\alpha + b \cdot \sin 2\alpha)$$

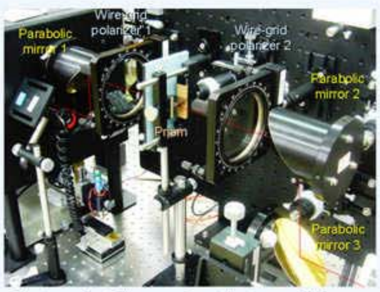
4. The ellipsometric parameters Ψ and Δ are calculated using the relations:

$$\cos 2\Psi = -a \qquad \cos\Delta = \frac{b}{\sqrt{1-a^2}}$$

5. Finally, the sample refraction index is calculated:

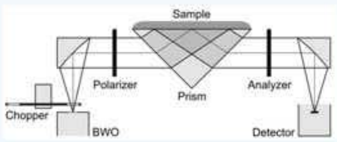
$$n_1 = n_0 \frac{\sin\theta_0}{\sqrt{1 + \left(\frac{1-\rho}{1+\rho}\right)^2 \tan^2\theta_0}} \qquad \text{where } \rho = \tan\Psi \cdot e^{i\Delta}$$

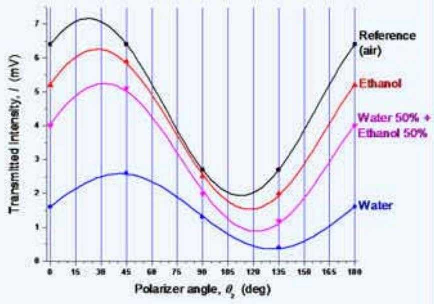
Experiments



Photograph of the experimental setup, with the main components and the optical axis identified.

The BWO source can output about 1 mW of power in the 400–700 GHz frequency range.





Preliminary results on three liquids: water, ethanol, and a 50% (vol.) ethanol solution in water. A measurement with air as sample is given for reference. The liquids can be clearly distinguished.

Future plans

- At present the liquids can be distinguished (hence identified), but without determining the exact complex refraction index. We plan to develop the formulas so that we can use published data about liquids without actually measuring them.
- We plan on using a 50 or 60 mm prism for an improved signal-to-noise ratio. Our present prism is only 20 mm, whereas the BWO collimated beam is about 60 mm in diameter.
- Slight prism misalignments introduce repeatability errors in the data. We are looking for a way to reduce those errors.

Acknowledgment

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Abstract for the 34th International Conference of Infrared, Millimeter, and Terahertz Waves:

Ellipsometry in the Terahertz Range for Liquid Identification

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***Abstract*—We present a method to determine the optical constants of a liquid at a given frequency in the terahertz range, by using a backward-wave oscillator as a continuous-wave source and an ellipsometric measurement setup consisting of two wire-grid polarizers. The liquid sample is placed on the total internal reflection surface of a silicon prism, thus changing the polarization state of the wave. By rotating the analyzer, useful information can be gathered about the optical properties of the liquid. We expect that this will ultimately allow the liquid identification or analysis.**

I. Introduction

RECENT years have seen a rapid progress in the field of terahertz (THz) radiation physics and applications, mostly due to the growing availability of sources, detectors and components for this frequency range. Many of the applications respond to the constant need for new technologies in homeland security and other related fields. This report belongs to the same category, in that it describes the preliminary study of a technique that could be applied for identifying liquids.

One of our previously reported results[1] consists in using a monolithic Fabry–Pérot resonator, in the form of a silicon prism, to investigate the loss produced by a liquid in contact with the total internal reflection surface of the prism. The measurements showed that it is possible to obtain valuable information on the optical properties of the liquid, which in turn can be used, for example, to determine the unknown concentration of a water–ethanol mixture. The present report uses a similarly-shaped prism, but the method used to analyze the interaction between the liquid and the electromagnetic radiation no longer relies on resonance, but are sensitive to changes in the polarization status of the incoming wave, evaluated using ellipsometric measurements. Jepsen *et al.* have recently reported yet another method, based on pulsed sources and time-domain spectroscopy[2].

II. Experiments and Results

The experimental setup, shown in Figure 1, consists of a backward-wave oscillator (BWO) as a continuous-wave source, with an output power in the order of 1 mW output at around 500 GHz. The beam is collimated, passed through a wire-grid polarizer to orient the polarization plane at an angle of 45° with both principal directions, and then enters the high-resistivity silicon prism. When no sample is placed on the top surface of the prism, a total internal reflection takes place, whereas if the beam encounters a liquid, both parts of the liquid's complex refractive index have an impact on the polarization state, which becomes elliptical. To measure this ellipticity, the wave is passed through a second polarizer (the analyzer), whose orientation is set by a computer-controlled rotation stage.

The power reaching the room-temperature pyroelectric sensor is measured at various angles of the analyzer. It is found that every liquid affects the polarization state in a different way. This is what provides the ability to identify an unknown liquid, except if two liquids have a very similar value of the complex refractive index at all frequencies available from the BWO, approximately 400–700 GHz in our case.

Figure 2 shows a few examples of such a measurement on three liquids, with air taken as reference. The measured data are fitted with sine curves, which carry in their parameters information on the complex refractive index of the respective samples.

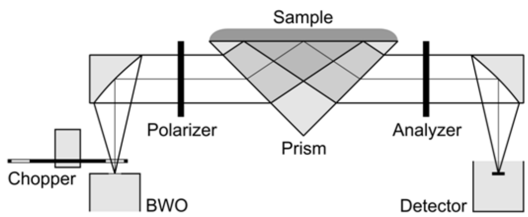

Fig. 1. Schematic of the optical setup (not to scale).

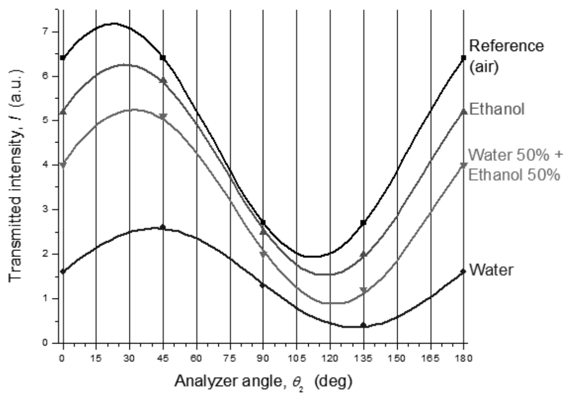

Fig. 2. Experimental data obtained on water, ethanol and their 50% v/v mixture. A measurement with air as sample is given for reference.

References

[1] A. Dobroiu, R. Beigang, C. Otani, and K. Kawase, "Monolithic Fabry-Perot resonator for the measurement of optical constants in the terahertz range," *Applied Physics Letters* **86**, p. 231107, 2005

[2] P. U. Jepsen, U. Møller, and H. Merbold, "Investigation of aqueous alcohol and sugar solutions with reflection terahertz time-domain spectroscopy," *Optics Express* **15**, p. 14717, 2007