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Homeland security enforcement using novel terahertz technology

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Abstract: The purpose of this research was to investigate the application an ellipsometric technique in the determination of the optical properties of liquids in the terahertz frequency range. The main results obtained in this period were as follows. 1. The reasons for the previous problems with repeatability were identified and the problems were largely solved. They consist in the occurrence of a Fabry-Pérot effect within the optical setup, which makes the signals sensitive to very slight misalignments. 2. Other phenomena that could affect the results by changing the polarization state of the radiation were found, explained, and canceled. They relate partly to the same Fabry-Pérot effects and partly to the changes in the polarization state due to the passage of a focused beam through circular apertures whose diameter is comparable to the wavelength. 3. Advances were made in the signal and data processing. Instead of complex analytical calculations we chose to realize a computer program that simulates the beam propagation and interaction with the various elements of the setup. This allows a fitting procedure to find the values of the optical properties of the sample liquid by adjusting a set of simulation parameters directly related to physical parameters in the actual setup. 4. Investigations at other terahertz frequencies showed that such measurements have the potential to bring in more data which would allow an even more reliable identification of the sample liquid, but that for such measurements to contribute significantly it is necessary to use terahertz frequencies more widely spaced than available from the BWO source that we used in our experiments. 5. By using a larger silicon prism, the errors were greatly reduced as the beam was used more efficiently.

Introduction: Terahertz radiation is the electromagnetic waves with a frequency around 1 THz = 10^{12} Hz. In the electromagnetic spectrum, this range is found between high frequency millimeter waves and long wavelength infrared radiation. Until about 15 years ago the terahertz frequency range was very little studied and material properties in this range were largely unknown, because there were not many affordable sources and detectors to work at these frequencies. As soon as these became available, the research in this field exploded, so that now there are hundreds of scientific papers published every year just concerning this frequency range and many practical applications have appeared which prove that it has a very good potential to help solve real-world problems: post-office screening of envelopes to detect illicit drugs, airport checks for detection of weapons and explosives hidden under passenger clothes, diagnosis of cancer, burns and other medical conditions, art conservation and diagnostics, agricultural automation, industrial investigation, etc. The main qualities that recommend this frequency range are the fact that many otherwise opaque materials are transparent to terahertz waves, the fact that many organic materials have specific fingerprint spectra allowing their identification, and the fact that, unlike for instance X rays, terahertz waves have no detectable impact on the living tissue, which makes them safe to use where operator health is a concern.

The ultimate goal of our research is to develop a technique for identifying liquids. This can have numerous applications, from homeland security (for instance checks in airports) to pharmaceutical and chemical analysis.

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14. ABSTRACT
This is a report of research on the application of an ellipsometric technique to determine the optical properties of liquids in the terahertz frequency range.

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The principle of liquid identifying is to allow the interaction between a terahertz beam and the unknown liquid to interact, which gives information about the absorption coefficient and refractive index of the liquid. These two optical properties are different from one liquid to another.

If, by an unlikely coincidence, two liquids have the same pair of optical properties at one frequency, then they can most likely be discriminated at another frequency. We deemed it to be practically impossible to create, even intentionally, a mixture of liquids that imitate the optical properties of a given liquid at all terahertz frequencies. As a consequence we believe that a machine employing terahertz radiation to identify liquids cannot be fooled.

This means that, by using a database of liquid optical properties, measuring the absorption coefficient and the refractive index of an unknown liquid will allow the reliable identification of the unknown liquid.

Experiment: The experimental setup consists of several basic elements: a terahertz source and an appropriate detector, a silicon prism which allows the interaction between the radiation and the liquid in a controlled way, and a pair of polarizers, as shown in Figure 1.

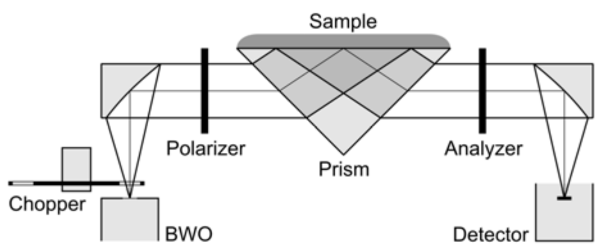


Figure 1. Schematic of the experimental setup, showing only the main components. The optical chopper placed in front of the BWO source is non-essential; it is used to achieve a low-noise detection.

The first polarizer is fixed so that it makes 45° with both axes, while the second polarizer (labeled "analyzer") is mounted on a computer-controlled motorized rotation stage and is used to analyze the polarization properties of the radiation after it interacted with the liquid sample. The same computer that controls the rotation stage reads and processes the signal from the detector synchronously with the analyzer rotation.

The prism used in the setup is made of high-resistivity silicon, which is the most transparent material available for the terahertz range. Compared to the prism we used in previous measurements, this one is large (the small sides are about 55 mm) and allows an efficient use of the available radiation, with minimum losses. The upper surface of the prism has an edge rail made of silicone rubber that prevents the liquid from spilling over the edge. A correct interaction between the terahertz radiation and the liquid requires a liquid thickness that is considerably larger than the so-called penetration depth of the liquid. This penetration depth differs from liquid to liquid, but is generally in the order of tenths of a millimeter; consequently we chose a liquid thickness of about 3 mm.

We made experiments on several liquids: water, alcohol, various water-alcohol mixtures, acetone, milk, etc. The typical curves that were obtained are shown in Figure 2.

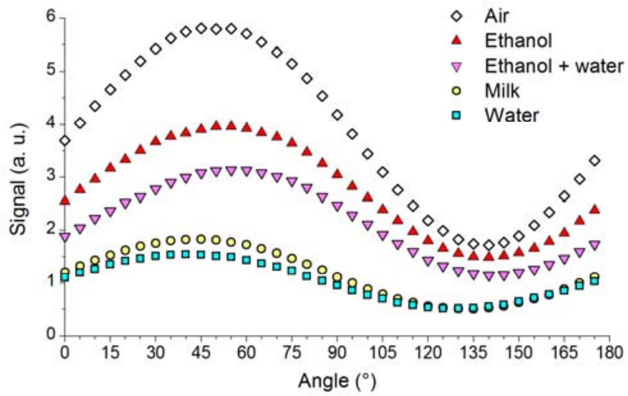


Figure 2. Examples of signals obtained on various liquids.
The water-ethanol mixture is 50%-50% by mass.

The graph shows that at least for these sample liquids there is no overlapping and the liquids can be easily identified even without further processing of the data.

For easy understanding, a 2D map is shown in Figure 3, where each liquid is represented as a dot.

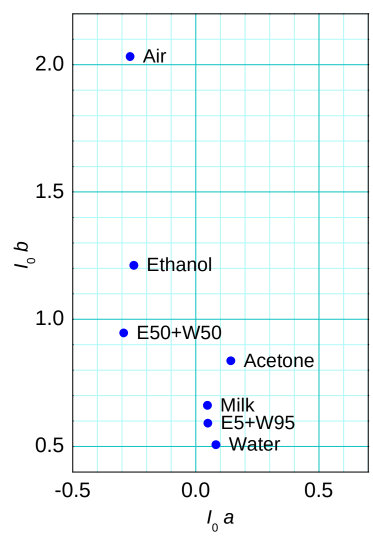


Figure 3. A 2D representation of the data. Air data is also given for reference. The liquid labeled E5+W95 is a mixture of 5% ethanol in water by mass (a concentration similar to that of beer).

The optical setup contains elements used for alignment, such as a visible light source that creates a beam exactly overlapping the terahertz beam, a removable small aperture used for checking if the prism has moved. Occasionally we used a partial absorber (attenuator) to cancel the Fabry-Pérot effects caused by repeated reflection between the source and the detector.

Data processing: The experimental data were processed using a computer program that we made for this purpose. In essence, the program simulates the experimental setup, allowing for the adjustment of many parameters such as the polarizer angle, an offset of the analyzer angle, tilt angle for the silicon prism, errors of the prism angles, the refractive index of the prism and that of the liquid, etc.

By repeatedly adjusting these parameters the program allows the fitting of the measured signal with the simulated signal. This gives the two optical parameters of the unknown liquid as an output of the program.

In principle the same result should be obtained analytically, by solving the equations governing the setup. However, the complexity of the setup and the fact that the radiation suffers a total of three Fresnel interactions (two refractions and one reflection) makes such analytical calculation extremely difficult and any additional parameter complicates the formula beyond usability. We decided that the simulation option was the best available.

Results: The first result is the experimental confirmation that two liquids give two different output signals, which is the most important point in any identification process. By further data processing we were able to obtain the optical parameters of the liquids. This allows the use of data measured and published by others, so that we don't have to measure everything by ourselves in order to make the database.

Besides this main result, we believe that several other observations made during the experiments are interesting both as theoretically and practically. An important observation was the fact that Fabry-Pérot resonances inside the optical path have a strong negative effect on the measurements and must be canceled. They are particularly destructive in ellipsometry measurements because the quality factor of the resonances changes with the rotation of the analyzer; this has an impact on the perceived linearity of the detector and eventually on the measured data. The effect is so strong that the procedure we used to fit the ellipsometric data with a sine wave can no longer work, as it gives negative values for part of the fitted data, which is impossible (any radiation intensity must be non-negative).

In order to cancel the resonances we had two options. One is to use an attenuator somewhere along the optical path. The absorption must be strong enough to sufficiently extinguish the beam coming back after a reflection, but weak enough to let some radiation pass. The other option is to modulate the frequency of the terahertz source so that the effects of many frequencies average in a way that the effects of the Fabry-Pérot resonance are canceled. By far the best option is the latter one, because it ensures a good use of all the terahertz power available, but the first option must be retained for the case where in a practical implementation a source is used that does not allow frequency modulation.

Another useful observation was the fact that the small aperture we used for checking the prism alignment had an effect on the polarization state of the beam, in that it reduced the polarization contrast of a linearly polarized beam. We still have to investigate what the theoretical reason for that observation may be, but from a practical point of view we found that it is important to remove the aperture before the data is measured. Another implication of this is that we cannot use a detector whose entrance pupil is so small as to affect the polarization.

Discussion: The research presented here shows that in principle it is possible to identify liquids. Before this principle can be implemented in a practical application, however, it is necessary to solve a few problems. Among them, probably the most challenging one is to achieve measurements while the liquid is still in its container. This means that the effect of the container on the signal must be either known or somehow canceled. One possible solution for this, that we haven't tried out yet, is to use a pulsed terahertz beam, which would allow a gated measurement, that is, the data would be collected only from the interaction of the beam with the liquid, not with the container.

However, the research can be used in its current stage for other applications such as the measurement of concentration of mixtures. For this purpose a series of mixtures with known concentrations are measured, then the place the mixture with the unknown concentration is found among the others by interpolation. As a rough approximation it is possible to measure just the two pure liquids and assume that the optical properties of the mixture are linearly distributed between the extremes, but from our experience with mixtures of water and ethanol the error such an assumption leads to are too large for any practical purpose, so at least a few intermediate concentrations must be checked.

List of Publications: For now only the work below was published on this subject, presented at the annual conference on terahertz physics. We plan on publishing a full journal paper soon.

- Adrian Dobroiu and Chiko Otani, “Measurement of the complex refractive index of liquids in the terahertz range using ellipsometry,” 35th International Conference on Infrared, Millimeter and Terahertz Waves (IRMMW-THz 2010) Sept. 5-10, 2010, Rome, Italy, (paper Tu-P.51)

A copy of the poster presented at the conference is attached on the next page.

Measurement of the complex refraction index of liquids in the terahertz range using ellipsometry

Adrian Dobroiu, Chiko Otani

RIKEN – Terahertz Sensing and Imaging Laboratory, Sendai, Japan

IRMMW-THz 2010
Rome, Italy

Purpose

- Liquid identification
- Liquid analysis
- *Solution concentration measurement*

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)

Principle

A terahertz wave is sent through a transparent silicon prism. Without the liquid sample, a total reflection occurs on the upper surface. When a liquid 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, which allows measuring the the complex refraction index of the sample.

The internal reflection is governed by two Fresnel reflection coefficients, one for each polarization

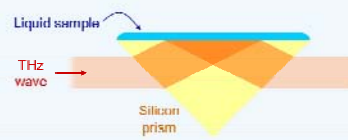
$$r_{\parallel} = \frac{n_1 \cos\theta_0 - n_0 \cos\theta_1}{n_1 \cos\theta_0 + n_0 \cos\theta_1} \qquad r_{\perp} = \frac{n_0 \cos\theta_0 - n_1 \cos\theta_1}{n_0 \cos\theta_0 + n_1 \cos\theta_1}$$

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:

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



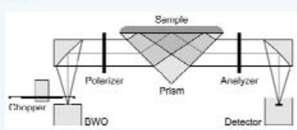
The refractive index of the sample can be measured by analyzing the polarization state of the output beam. We propose using an **ellipsometric technique**:

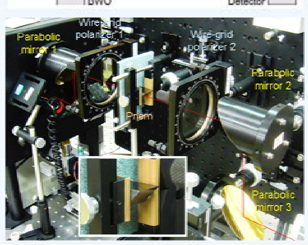
1. The input beam is polarized at 45°, halfway between the parallel and the perpendicular polarization.
2. The output beam is passed through a second polarizer, whose orientation can be adjusted.
3. By rotating the second polarizer, a sine-shaped intensity signal is recorded.

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

4. Then the complex refraction index of the liquid is calculated.

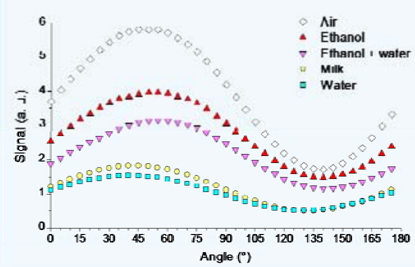
Setup



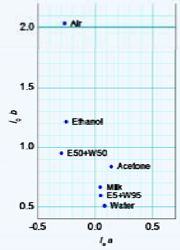


The BWO source can be tuned: 400–700 GHz.

Results



Sine signals with various liquids as sample.
It is obvious that liquids can be identified.



2D mapping of a few samples

Present challenges

- We still need to go beyond simple *identification and actually calculate the* complex refraction index. This will allow using published data for liquids.
- Fabry–Pérot effects occur in the system.
- Repeatability errors still affect the results. *We have some ideas to reduce them.*
- The polarization state is affected by a number of factors, including the passage through a small aperture used for alignment.

Acknowledgment

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