Investigation on improving the noise performance of T-ray liquid spectroscopy via double-modulated differential time-domain spectroscopy

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ABSTRACT

Liquid spectroscopy allows analysis of chemical composition and provides a better understanding of the solvation dynamics of various types of liquids. Although it has been shown that liquid spectroscopy using T-rays is feasible, liquid water absorption is still considered to be one of the most challenging problems facing THz imaging and spectroscopy in biomedical applications. The absorption coefficient for liquid water shows a very high THz absorption, 200 cm^{-1} at 1 THz. This paper describes a promising novel liquid double-modulated differential time-domain spectroscopy (Double-modulated DTDS) technique to extract the optical parameters with a dual-thickness measurement. The described technique improves on the previous work, by replacing the required sample dithering technique with a rotating spinning wheel resulting in an improved noise performance up to two orders of magnitude.

Keywords: liquid DTDS, T-rays, spectroscopy

1. INTRODUCTION

Terahertz (THz) radiation or T-rays lies between the millimetre-wave and infrared band in the electromagnetic spectrum, at approximately 0.1 THz to 10 THz, which corresponds to the wavelength range 3 mm to 30 μm (Figure 1). Recent advances in terahertz generation and detection techniques have stimulated researchers throughout the world to explore and take an interest in its possible applications. Over the last decade, there has been a pronounced interest in THz spectroscopy of liquids. A number of papers have been published to show that liquid spectroscopy is feasible using T-rays.[1–6] Liquid spectroscopy allows analysis of chemical composition and provides a better understanding of the solvation dynamics of various types of liquids.[2–4,6–8] This leads to possible applications such as blood sample screening, urine testing, contaminant detection in liquid, wet protein sample analysis, detection of inflammable liquids, sugar and salt level of content in water, fat or oil content in polar liquids, and many more applications. However, although it has been shown that liquid spectroscopy using T-rays is feasible, the absorption coefficient for liquid water shows a very high THz absorption, 200 cm^{-1} at 1 THz[5] and there are still open questions that need to be addressed to improve the signal-to-noise ratio (SNR) of liquid spectroscopy. The two major sources of uncertainties that affect the SNR are the fluctuations in T-ray pulses caused by the fluctuations in the generated laser pulses and the accuracy of the sample thickness measurement. In this paper, a fast and reliable method of extracting the optical parameters of liquids in the THz range is investigated, using the double-modulated differential time-domain spectroscopy (DTDS) method.

1.1. Dithering technique

Differential time-domain spectroscopy (DTDS) is a technique used to measure the optical properties at THz frequencies with a reduced measurement time between the sample and reference.[1,9–11] Here, DTDS is implemented by using a galvanometric shaker, which is dithered at a constant frequency.[10] The reference and sample liquid are placed on the shaker. As the shaker dithers, modulation between the reference and sample liquid can occur at a maximum frequency rate of up to 66 Hz.[9] Since the T-ray noise follows a $1/f$ trend,[1] it was noticed that

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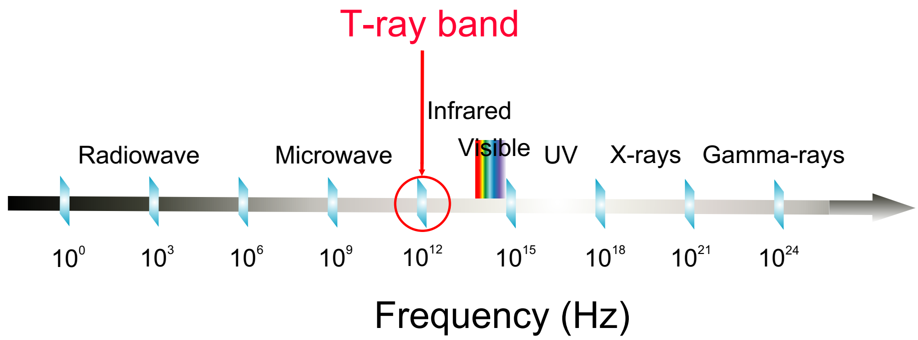

Figure 1: Electromagnetic spectrum

further improvement in the noise performance can be made by increasing the modulating frequency. Thus, this was carried out by introducing the double-modulated DTDS technique whereby the information is encoded on a carrier wave. The modulated signal at a lower frequency via dithering is modulated again by a higher frequency via a chopper.[11] Double-modulated DTDS can be carried out by using a rapid modulation of the liquid thickness in the T-ray beam at higher modulation frequency.[1] Based on this technique, it is observed that by rapidly modulating the samples (swapping between reference and sample materials) and measuring the waveforms in quick succession, the measurement time between the reference and the sample waveforms can be further reduced.[1] In a prototype system, this technique is implemented by using an audio speaker for dithering purposes. A very thin high density polyethylene bag (HDPE) is used as the liquid holder for the measurements.[1] With the assumption that only two thicknesses are present, a square wave is applied to the speaker for the thick and thin sample switching.[1] Based on this experiment, a reasonable result for the noise performance was obtained. However, the objective which was to measure optical parameters with an accuracy better than 10^{-4} was not met. This is basically due to the fact that the speaker introduces uncertainties via its mechanical noise and also partially due to the uncertainties in the system itself. The experiments in this paper address the limitations of the sample dithering technique by introducing the spinning wheel technique at a higher modulating frequencies.

1.2. Novel spinning wheel technique

This paper shows a novel T-ray holder (spinning wheel) modulating at a range of frequencies that will increase the measurement accuracy of the liquid parameter estimation by increasing the SNR. A fast measurement between the reference (thin) and the sample (thick) portion of the same liquid is carried out using the spinning wheel. Reduction in the acquisition time between the waveforms can be observed and hence minimize the effect of noise caused by the laser fluctuations. Compared to the previous method,[1] where the thickness depends on the variation of d of the dithering technique, this technique uses a fixed thicknesses. This removes the uncertainty caused by the error in thickness and thus further improves the SNR. A suitable window material that has a optimal refractive index, low absorption and low hygroscopicity is required to improve the SNR performance.

2. THEORY

The spinning wheel is a mechanism proposed to address the limitations of the existing sample dithering technique. The wheel is used to determine the characteristics of different types of liquids with an expected improvement in noise performance up to two orders of magnitude. The spinning wheel is designed for transmission experiments and therefore requires a window material that has a optimal refractive index (to reduce reflection), low absorption and low hygroscopicity (Figure 2).

The window material used has a cavity with two different thicknesses, reference (thin) and sample (thick). The cavity is used to hold the liquid to be measured. As the wheel spins, rapid thickness modulation will occur. Rapid modulation will result in reduction of the measurement time between samples. Reducing this measurement

Figure 2: A photograph of the prototype spinning wheel with dual-thickness. The thick and the thin sample waveforms can be measured by spinning the wheel at a high modulating frequencies to improve the signal-to-noise ratio.

interval results in better accuracy because it reduces the probability of laser fluctuations within that time frame. In addition, only one delay scan is required to measure both the reference and sample waveforms. Figure 3 shows the cross-sectional view of the spinning wheel.

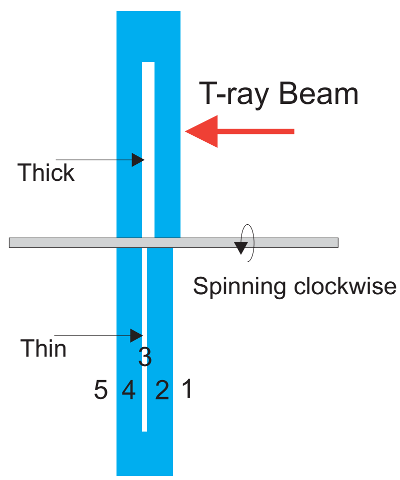

Figure 3: Cross-sectional view of the spinning wheel. The blue layers are the window materials that hold the liquid. The liquid is inserted in the cavity between the two windows. The T-ray beam passes through the wheel as it spins. Two different T-ray waveforms—reference (thin) and sample (thick)—will exit the wheel. These waveforms will be used to determine the liquid characteristics.

Assuming that, the T-ray beam is incident on the wheel, the spectral components at angular frequency ω of the electric field of the terahertz wave transmitted through the sample and reference can be written as:

$$\tilde{S}_{\rm sam}(\omega) = T_{12}(\omega)\ .\ P_2(\omega, d_{\rm mat})\ .\ T_{23}(\omega)\ .\ P_{\rm thick}(\omega, d_{\rm thick})\ .\ T_{34}(\omega)\ .\ P_4(\omega, d_{\rm mat})\ .\ T_{45}(\omega)\ .\ A(\omega)\ , \quad (1)$$
$$\tilde{S}_{\rm ref}(\omega) = T_{12}(\omega)\ .\ P_2(\omega, d_{\rm mat})\ .\ T_{23}\ (\omega)\ .\ P_{\rm thin}(\omega, d_{\rm thin})\ .\ T_{34}\ (\omega)\ .\ P_4(\omega, d_{\rm mat})\ .\ T_{45}(\omega)\ .\ A(\omega)\ , \quad (2)$$

where $\tilde{S}_{\rm sam}$ and $\tilde{S}_{\rm ref}$ are the complex spectral components of the sample and the reference pulses respectively. The P_2 and P_4 refer to the propagation coefficient of the window material and $P_{\rm thick}$ and $P_{\rm thin}$ refer to the propagation coefficient of the cavities filled with liquid. Also $T_{\rm ab}$ is the transmission coefficient of the THz wave from material a to b and $A(\omega)$ accounts for the amplitude of each frequency component.[1] Note that $T_{\rm ab}(\omega)$,

$P_{\mathrm{a}}(\omega)$ are dimensionless, $A(\omega)$ is in arbitrary units and the indices are defined in Figure 3. With the assumption that the thickness change d is large enough as it spins, the FP (ω), Fabry Pérot reflections can be ignored.

The ratio of the transmission spectra is as follows:

$$\tilde{T}(\omega) = \frac{\tilde{S}_{\mathrm{sam}}(\omega)}{\tilde{S}_{\mathrm{ref}}(\omega)} . \tag{3}$$

Thus, the optical parameters can be extracted from the ratio of the transmission spectra using the formulas as follows,

$$n(\omega) = \frac{\phi c_0}{\omega d} + 1 , \tag{4}$$

$$\kappa(\omega) = -\ln(\rho)\frac{c_0}{\omega d} , \tag{5}$$

$$\alpha(\omega) = \frac{2\kappa(\omega)\omega}{c_0} , \tag{6}$$

where n is the refractive index, κ is the extinction coefficient, α is the absorption coefficient, ρ is the magnitude, ϕ is the phase of the transmission spectrum, c_0 is speed of light, $\omega = 2\pi f$ and $d = d_{\mathrm{thick}}$ - d_{thin}, which is the thickness change.

3. THE EXPERIMENTAL METHOD

This section describes the experimental procedure for the liquid DTDS. The THz-DTDS for liquid transmission is shown schematically in Figure 4. This setup is based on the conventional THz-TDS with an addtional chopper and lock-in amplifiers for noise reduction and double modulation purposes. A femtosecond laser with 100 fs pulse width, 800 nm wavelength and 82 MHz repitition rate is used to drive the THz system. The laser pulses are split into two beams, a pump and a probe beam using a beam splitter. The pump beam generates the T-ray pulses when incident on the THz emitter. The pump beam is modulated by a mechanical chopper. The chopper in conjunction with a lock-in amplifier reduces the background noise.[12] A motorised delay stage is required for adjusting the path delay and hence phase between the pump and the probe beams. This delay stage limits the frequency resolution of the T-ray spectrum and the maximum observable T-ray frequency. The terahertz beam is collimated and focused on the spinning wheel by two parabolic mirrors. The transmitted THz beam through the spinning wheel is collimated and focused onto the THz detector by the second pair of parabolic mirrors. The probe beam is coincident with the THz beam for detecting the polarization changes through the Wollaston polarising beam splitter and the balanced photodetectors. A variable frequency f_2 is applied to the spinning wheel using an external function generator via the lock-in amplifier 2 to allow the rapid thickness modulation (Figure 4). Another external function generator is used to provide frequency f_1, to the mechanical chopper via lock-in amplifier 1. The mean signal, y_m and the amplitude signal y_a can be obtained by using the built-in LPF in the lock-in amplifiers.

3.1. Sample preparation procedures

This section provides the technique used in preparing the liquid sample and the process of building the spinning wheel using different types of window materials. The spinning wheel is designed with a diameter of 10 cm with dual-thickness geometry, reference (thin) and sample (thick) approximately 10 μm and 100 μm of thickness respectively. Different types of window materials have been tested for the spinning wheel and the results are as shown in Table 1.

Two different measurements are carried out on the window materials; firstly in a dry condition and secondly in a soaked condition, where the window materials are soaked in water for 48 hours. Based on the measurements, the refractive index and the absorption coefficient of the window materials are analysed and the results are

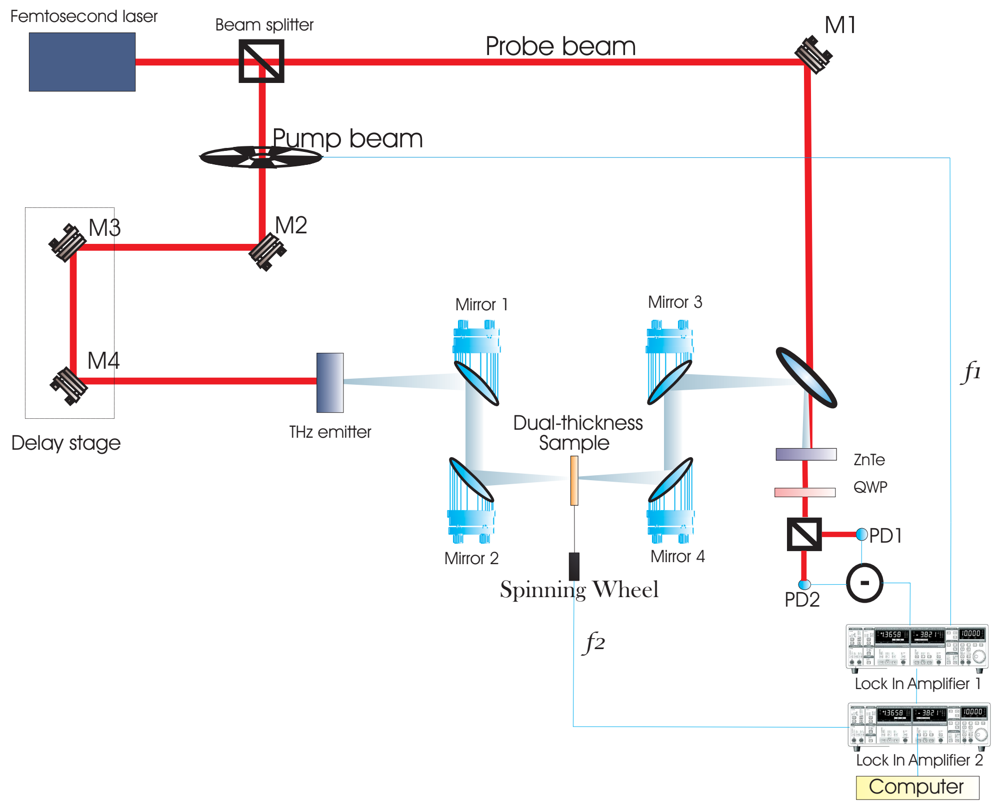

Figure 4: Setup diagram for liquid spectroscopy using double-modulated DTDS. The spinning wheel with dual-thickness is placed at the focal point of the THz beam to measure the differential THz signal.

Window material (0.8 THz)	**Material optical properties**			
	n (dry)	n (soaked)	α cm^{-1}(dry)	α cm^{-1}(soaked)
Acrylic	1.515	1.525	7.5	9
Nylon	1.534	1.537	0.25	0.5
HDPE	1.477	1.483	0.1	0.2
Polycarbornate	1.593	1.597	5	6.2
PVC	1.652	1.71	12	14.9
Teflon	1.443	1.447	0.1	1.5

Table 1: Experimental results of the refractive index (n) and absorption coefficient (α) of different window materials. Two different measurements were taken; dry and soaked condition to observe the absorption.

shown in the above table. Since the requirement of the window materials are to have optimal refractive index, low absorption coefficient and low hygroscopicity, high density polyethylene (HDPE), nylon, and teflon show promising results as compared to the rest of the measured window materials. A preliminary experiment is conducted using the HDPE as window material. The HDPE window material is moulded with a dual-thickness, thin and thick layers with the thicknesses of 10 μm and 100 μm respectively. The liquid water is inserted in the wheel by immersing the window material (HDPE) with vacuumed cavities into water. This ensures that no macroscopic bubbles exist in the water sample. A washer is placed between the two windows to avoid leakage. It is then clamped with a light weight aluminium (Note the aluminium rim in Figure 2). The wheel is then mounted on a DC motor for rotation action. The speed of the motor is controlled by the frequency supplied through the function generator via lock-in amplifier 2.

3.2. Data flow

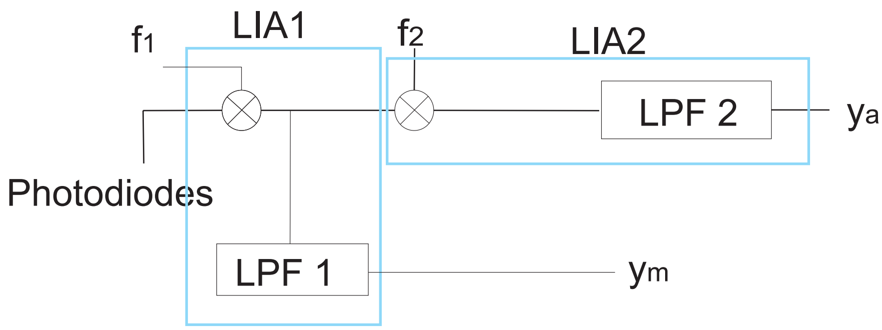

Figure 5: Signal recovery using the lock-in amplifiers. The block diagram shows the extraction method of the mean signal y_{m}, and the amplitude signal y_{a}, using the 2 lock-in amplifiers, which can be used for the optical parameter estimation. The average differential signal from the photodiodes is demodulated in two stages using the lock-in amplifier to obtain y_{m} and y_{a} for further signal processing.

Since the average differential signal from the photodiodes is a combination of noise sources and frequency signals f_1 and f_2, lock-in amplifier (LIA) can be used to recover the desired signals. The modulated differential signal from the photodiodes is demodulated in two stages using lock-in amplifiers. The first demodulation is performed by using the LIA1. The output of LIA1 is mixed with the frequency signal f_2, using the second LIA. The built-in LPF2 is used to recover the amplitude signal y_{a}, by filtering out any undesired signals. The mean signal y_{m}, can be recovered by using the built-in LPF1 in LIA1 (Figure 5). Thus, by recovering the amplitude and the mean signals, the reference signal y_{r}, and the sample signal y_{s} can be obtained by using the following formulas and further signal processing.

$$y_{\mathrm{r}} = y_{\mathrm{m}} - y_{\mathrm{a}} \, , \tag{7}$$

$$y_{\mathrm{r}} = y_{\mathrm{m}} + y_{\mathrm{a}} \, . \tag{8}$$

The fast fourier transform of the y_{r} and y_{s} using the formula given in Equations 1 and 2 gives the spectral components. Hence by taking the ratio of the spectral components using the Equation given in 3, the optical parameters n, κ, and α can be obtained.

4. PROGRESS AND RESULT

Based on the experiments conducted, it is found that the thickness variation in the window material is very high. The resulting errors are in the range of 10 to 50 μm. The main reason for this is basically due to the machine inaccuracies during the moulding process. Due to this error, it is predicted that the result of the liquid measurement using the spinning wheel will be affected. In addition, it is found that HDPE is too soft and flexible to act as a window material. This may result in centrifugal forces as the wheel spins. Further investigations are being carried out on materials such as cycloolefin and silicon wafers, which could be possibly used as a window material for the spinning wheel. Preliminary results that demonstrate the potential of this technique for liquid spectroscopy using the cycloolefin and silicon wafer as a window material will be presented in the near future.

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