Double-modulated DTDS-THz liquid spectroscopy using a novel spinning wheel technique

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Abstract—**A fast and reliable novel technique for a terahertz liquid spectroscopy based on the double-modulated differential time-domain spectroscopy (Double-modulated DTDS) is presented. This technique is implemented using a robust stainless-steel wheel with a dual-thickness cyclic olefin copolymer (COC) window driven by a high speed brushless 3 phase motor.**

I. Introduction And Background

LIQUID spectroscopy in the terahertz frequency region has always been a topic of interest among researchers. Liquid spectroscopy using terahertz radiation allows analysis of chemical composition and provides a better understanding of the solvation dynamics of various types of liquids[1-4]. Due to the fact that terahertz significantly absorbs water, i.e approximately 200 cm^{-1} at 1 THz[3], various techniques to improve the sensitivity of the measurement have been explored[5-8]. In this paper, we present a novel technique for liquid spectroscopy using a wheel with a dual-thickness window to measure the terahertz liquid properties. This technique allows reduction on the measurement time to half and with a better SNR. The described technique uses well-known double-modulated differential time-domain spectroscopy (double-modulated DTDS)[5,7].

Double modulated DTDS is a technique used to measure the terahertz properties of thin samples with a reduced measurement time between the sample and the reference[9,10]. In the paper presented by Mickan *et al*[5,7], a theory describing the double-modulated DTDS technique to measure dual-thickness liquid is presented. This technique is implemented using an audio speaker as ditherer to produce the dual-thickness liquid. However, due to its mechanical instability, the accuracy of the measurement is not promising. Here, we present an improvement to address the limitations of the dithering technique. This technique is implemented using a robust stainless-steel wheel with a dual-thickness window driven by a brushless 3-phase DC motor spinning at a constant high speed. The prototype of the wheel and the cross-sectional view of the dual-thickness window are as shown in Figure 1. A window material, cyclic olefin copolymer (COC), which has a very low refractive index, is high in transmission, low in absorption and has low hygroscopicity[11] is used. The window is designed to have two cavities with two different thicknesses representing the reference and the sample. As the wheel spins at a constant speed, the radiation hits the window containing the sample to produce the mean and the amplitude signals. The setup configuration for the mean signal extraction and the amplitude signal extraction are as shown in Figure 2. The double-modulated signal is demodulated twice based on two

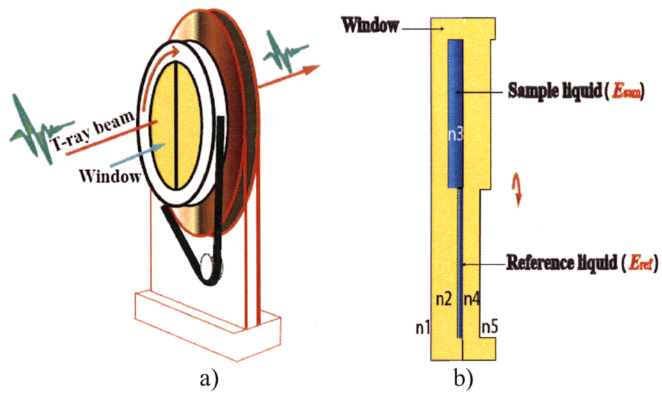

Figure 1: a) The prototype of the robust stainless-steel wheel, b) cross-sectional view of the window with dual-thickness cavity. The refractive index, $n_1 = n_5$ = refractive index of the air = 1, $n_2 = n_4$ = refractive index of the window = 1.52 and n_3 = refractive index of the liquid to be measured.

different frequencies. As the demodulated signal enters the first lock-in amplifier (LIA 1), the LIA 1 demodulates the signal with the chopper frequency. The other frequency components of the signal are filtered.

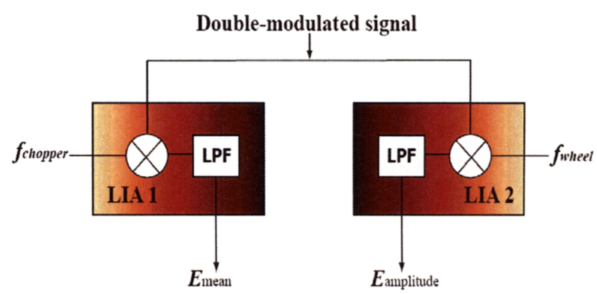

Figure 2: a) The lock-in amplifiers setup configuration for the mean and amplitude signal extraction.

Thus, the output channel of LIA 1 produces the mean signal (E_{mean}). As for the second lock-in amplifier (LIA 2), the incoming signal is demodulated at the wheel's frequency while the other frequency components of the signal are filtered. The amplitude signal ($E_{amplitude}$) is obtained at the output channel of LIA 2. Based on the extracted mean and amplitude signals, the E_{ref} and E_{sam} are obtained according to the following formula:

$$E_{ref}(\omega) = E_{mean}(\omega) + E_{amplitude}(\omega) \quad (1)$$
$$E_{sam}(\omega) = E_{mean}(\omega) - E_{amplitude}(\omega) \quad (2)$$

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Thus, the transmission coefficient, $T(\omega)$ can be obtained by taking the ratio of the E_{sam} and E_{ref}.

$$T(\omega)=\frac{E_{sam}(\omega)}{E_{ref}(\omega)}=\rho e^{i\phi}, \tag{3}$$

Equation (3) simplifies to

$$T(\omega)=\frac{\exp(-i\omega d_{sam}\tilde{n}_3/c)}{\exp(-i\tilde{n}_3\,\omega d_{ref}/c).\exp(-i\tilde{n}_5\,\omega d_{diff}/c)}, \tag{4}$$

and Equation (4) can be further simplified to

$$T(\omega)=\frac{-i\omega d_{diff}}{c}(\tilde{n}_3-1), \tag{5}$$

where $\tilde{n}_3 = n - i\kappa$ and $d_{diff} = d_{sam} - d_{ref}$. Therefore, based on Equation (3) and (4), the magnitude ρ and the phase ϕ can be rearranged as follow:

$$\rho=\exp(-\omega d_{diff}\kappa/c), \tag{6}$$

$$\phi = -\omega d_{diff}(n_3 - 1)/c. \tag{7}$$

Rearranging the magnitude and the phase produces the refractive index, n_3 and the kappa, κ.

II. Results and Discussion

The waveform in Figure 3 shows the relationship between the E_{mean}, $E_{amplitude}$, E_{ref} and E_{sam} at the nth step of the delay stage.

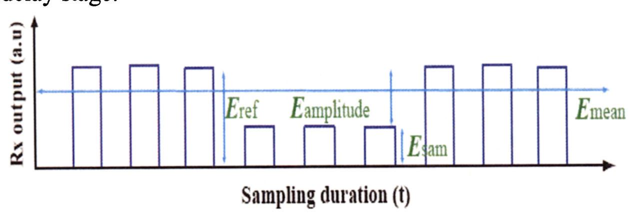

Figure 3: The time-domain waveform at the nth step of the delay stage. As the chopper and the spinning wheel spins, the signal is modulated at two different frequencies. The chopper frequency is set to 400 Hz and the spinning wheel frequency is set to 85 Hz.

The Figure 4 shows the experimental results of the temporal pulses and the spectral amplitudes for the E_{ref} and E_{sam} (calculated based on Equation 1 and 2) obtained through a distilled water case study. The accuracy of the measurement is confirmed by statically measuring the reference ($E_{ref\text{-}static}$) and the sample ($E_{sam\text{-}static}$). The E_{mean} and the $E_{amplitude}$ of the distilled water measurement are included on the plot. According to the Figure 4, a slight difference is noticed in both the time-domain and the frequency domain. This is due to the vibration introduced by the spinning wheel as it spins at a selected speed.

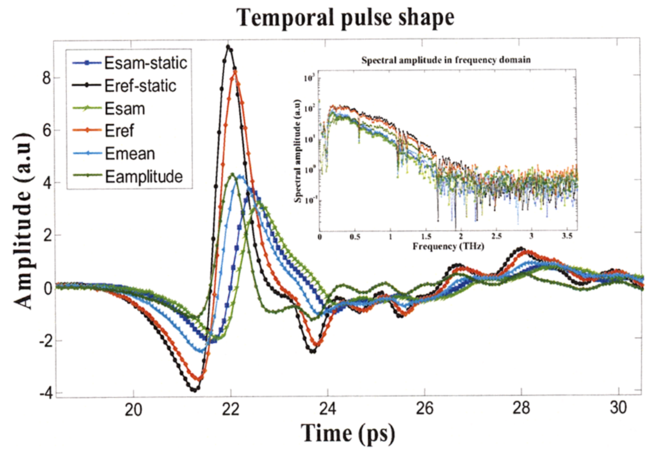

Figure 4: Depicts the temporal pulses for E_{mean}, $E_{amplitude}$, E_{ref} and E_{sam}, and $E_{ref\text{-}static}$ and $E_{sam\text{-}static}$. The insert shows the spectral amplitudes of the temporal pulses respectively.

III. Conclusion

We have presented a fast and reliable novel technique for terahertz liquid spectroscopy based on the double-modulated DTDS. This technique enables reduction in the sample measuring time to half and provides a better SNR.

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