Determining Ethanol Content of Liquid Solutions Using Laser Feedback Interferometry with a Terahertz Quantum Cascade Laser

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Abstract—Over the last decade, terahertz (THz) time-domain spectroscopy has been investigated as a technique for assaying the ethanol content of liquid solutions—indeed, operating at THz frequencies addresses some of the challenges that traditional optical refraction measurements face, such as delineation between sugar–ethanol content, florescence, and problems arising from carbonation or other dissolved gasses. In this article, we propose an alternative system and method for assaying ethanol content of liquid solutions at THz frequencies, which employs a laser feedback interferometer built around a 2.6-THz quantum cascade laser. The system is tested against a series of controlled water–ethanol solutions, as well as a series of commercially available beverages. The accuracy of the estimated ethanol content compares favorably to THz time-domain spectroscopy techniques.

Index Terms—Electromagnetic wave sensors, laser feedback interferometry, terahertz (THz) sensing.

I. INTRODUCTION

Measurement of ethanol content of various liquid solutions (including alcoholic beverages) is of significant interest, notably from a process-control viewpoint during manufacture, as well as to ensure final products meet regulatory guidelines. Traditional optical refraction-based measurements of alcohol (ethanol) content are affected by the difficulty in distinguishing changes in signal arising from alcohol content and sugar content [1]; typically, an additional density measurement (using a hydrometer) is also required to assay the final alcohol content. More sophisticated optical techniques exist, including those operating at near- and midinfrared wavelengths and those utilizing Raman spectroscopy [2]–[4], as well as recent technological developments that could be applied to the determination of ethanol content in liquid solutions, notably the use of nanoparticles to create localized surface plasmon resonance detectors [5], [6]. However, even these suffer from interference due to fluorescence, challenges arising from carbonation (requiring liquids to be degassed prior to measurement), and difficulty separating sugar and ethanol contributions from observed spectral signatures [2].

Terahertz (THz) time-domain spectroscopy (TDS) has been utilized in the literature to measure various normal alcohols (such as methanol, ethanol, n-propanol) [7]–[9], alcohol–water mixtures [10]–[14], alcohol–fuel mixtures [15], as well as determining ethanol content in commercially available alcoholic beverages [10], [16]. There is also a potential for the use of THz metamaterials to facilitate the detection of ethanol content in liquid solutions in a similar way to nanoparticles [17]. However, at the lower end of the THz spectrum (<2 THz), the separation of signal contributions sugar and ethanol remains a challenge and can affect the accuracy of measured ethanol content [10]. Fortunately, since the contributions to the signal from sugar in solution decrease faster with increasing THz frequency than those attributed to ethanol (see [10]), using a higher frequency THz source, such as a THz quantum cascade laser (QCL), would mitigate this effect.

In this article, we propose a simple system for assaying ethanol–water mixtures based on a THz QCL utilizing laser feedback interferometry (LFI). Laser feedback interferometry is a useful tool for laser sensing and utilizing the so-called self-mixing effect [18], [19]. Due to the possibility of extracting the LFI signal from across the laser terminals without the need for an external or separate detector, LFI is particularly applicable in the THz frequency range, which suffers from a lack of convenient, fast, and sensitive detector technologies [20], [21]. Previously, we have developed a method for material characterization [22], [23] utilizing this effect. However, several complicated and time-consuming calibration, sample preparation, and signal-processing techniques were required to produce accurate results. The method proposed here is substantially simpler, and once calibrated, the system only requires a small sample of the liquid for characterization. This approach could be adapted to investigate other fluid mixtures at THz frequencies.

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II. EXPERIMENTAL SETUP AND MODELING

The experimental setup (see Fig. 1) used a 2.59-THz QCL mounted in a free-flow cryostat operating at a temperature of 15 K. The THz QCL consisted of a 11.6–μm-thick GaAs/AlGaAs bound-to-continuum active region [24] that was processed into a semi-insulating surface-plasmon ridge waveguide with dimensions 1.78 mm × 140 μm. The laser was biased by a laser driver (Thorlabs LDC500C) with a dc current of 0.42 A, superimposed with a 1-kHz sawtooth-wave modulation with an amplitude of 75 mA (producing a 900-MHz frequency sweep). The signal was monitored by measuring the voltage across the THz QCL’s terminals after passing through an ac-coupled 20× amplifier (SRS SR560). This amplified signal was then digitized using a 16-b PC-DAQ with a 1 MS/s acquisition rate (National Instruments NI PCI-6251). Two 2-in parabolic mirrors (f = 4 in) were used to collect, collimate, and focus the emitted beam onto a target area. The signal was processed by negatizing the current-induced power ramp leaving only the interferometric self-mixing fringes (which we refer to as the self-mixing signal) [25].

Two 4.5-ml (10 × 10 × 45 mm³) cuvettes were used to hold the fluids under test. The two identical cuvettes were modified so that they shared a common 3-mm thick z-cut quartz window and were mounted on a three-axis (x-y-z) motorized stage. One cuvette was filled with deionized water as a reference material, with the other containing the liquid being assayed. Measurements of both the water reference and the liquid under test were taken in rapid succession, thus ultimately mitigating against small changes in the external cavity path (such as atmospheric changes like humidity, temperature, and minor mechanical changes).

The triple sec, port, and beer samples contain significantly more sugar (in solution) than the other beverages under test. Despite the low sensitivity of THz radiation to sugar content, its impact is still

![Fig. 1. Experimental setup of the system. (Inset) Compound nature of the target comprised of the cuvettes R (reference) and T (liquid under test).](image)

![Fig. 2. Self-mixing signals corresponding to test water–ethanol solutions with ABW of 0% (green line), 25% (purple line), 45% (yellow line), 75% (orange line), and 100% (blue line).](image)
evident here. Indeed, it is clear from Fig. 4 and Table I that the estimated ABW is slightly larger than that stated by the manufacturer on the label for these beverages. Nevertheless, due to the relatively high operating frequency of the THz QCL used here (2.6 THz), this impact is much reduced when compared with THz TDS measurements, which operate over a lower frequency range [10]. For the remaining beverages under test, estimated ABW aligns well with the expected linear relationship—deviations are on the same order as for THz TDS measurements.

For all these measurements, only a small volume of liquid was used for testing (~1 mL). However, the minimum volume of liquid required is much less. This is a consequence of the high attenuation of the THz signal for both ethanol and water at the measurement frequency (2.6 THz)—the longitudinal depth of either liquid required to act as a half-space for THz waves is < 500 μm. As such, with only minor changes to the geometry of the external optical components, the same system could be implemented using a quartz window on which a single drop of liquid under test is placed for measurement.

IV. CONCLUSION

In this article, we proposed and demonstrated a system and a method of assaying the ethanol content of liquid solutions. The system is built around a THz QCL operating at 2.6 THz as a laser feedback interferometer, employs a reference to mitigate environmental effects during measurement, and only requires a small volume of liquid for testing. The method compares well with existing THz TDS ethanol-assaying techniques.

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