Noninvasive in-vehicle alcohol detection with wavelength-modulated differential photothermal radiometry

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Abstract This study describes the potential of wavelength-modulated differential photothermal radiometry (WM-DPTR) for non-invasive in-vehicle alcohol detection which can be of great importance in reducing alcohol-impaired driving. Ethanol content in the range of concern, 0-100 blood alcohol concentration (BAC) in water phantoms and blood serum diffused in human skin in vitro were measured with high sensitivity. The results show that the WM-DPTR system can be optimized for alcohol detection with the combination of two sensitivity-tuning parameters, amplitude ratio R and phase shift ΔP. WM-DPTR has demonstrated the potential to be developed into a portable alcohol ignition interlock biosensor that could be fitted as a universal accessory in vehicles.

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1. Introduction

Alcohol-impaired driving is a major road safety issue of international interest and concern. A statistical projection of traffic fatalities shows that an estimated 34,080 people died in motor vehicle traffic crashes in 2012 in the US [1]. In Canada, the majority of police reported 90,277 impaired driving incidents in 2011were alcohol-related [2]. To reduce alcohol-impaired driving, one strategy is to increase the application of alcohol ignition interlocks on the vehicles of Driving While Intoxicated (DWI) offenders, which requires drivers to provide breath samples before starting their vehicles. If a positive Breath Alcohol Concentration (BrAC) is registered, the vehicle cannot be started. Studies indicate that it can reduce recidivism by about two-thirds [3]. However, many alcohol-impaired drivers have not been convicted of DWI. An effort is underway to develop advanced technologies that could be fitted in vehicles of all drivers to measure driver blood alcohol concentration (BAC) non-invasively, accurately (0.0003% systematic error (SE) and standard deviation (SD) at BAC legal limit level of 80 mg/dl) and rapidly (within 320 ms) [4,5]. There are four potential technology approaches [6]: (1) Tissue Spectrometry: BAC is estimated by measuring alcohol content of interstitial fluid (ISF) in the dermis by means of a Near Infrared (NIR) diffusely reflected beam from skin. This touch-based system requires skin contact. (2) Distant Spectrometry: Alcohol content of tissue or liquid in vapor is assessed through the IR absorption/reflection spectrum or using laser light. (3) Electrochemical/Chemical-reaction-based devices such as transdermal (perspiration) and breathalyzer-based systems: Alcohol in the presence of a reactant chemical will produce colorimetric changes measured by spectral analysis or a semiconductor sensor. (4) Behavior: It detects impaired driving through objective behavioral measures including ocular, gaze, eye movement, and driving performance measures. Current ignition interlock devices (IID) or breath alcohol ignition interlock devices (BAIID) are mostly based on fuel cell technology, which requires very frequent maintenance and calibration service. In addition, the interlock requires drivers to deliver a delicate breath into a device before starting a vehicle, and random breath samples must be provided for test during driving. To be acceptable for use among all drivers, in-vehicle alcohol detection technologies must be seamless with the driving task; they must be non-invasive, reliable, durable, and require little or no maintenance [4]. In order to develop an advanced device to prevent alcohol-impaired drivers from driving, the Driver Alcohol Detection System for Safety (DADSS) has chosen two technologies, TruTouch and Autoliv, for prototype development. TruTouch is a tissue spectrometry technology working on the principle of NIR reflection spectroscopy in the 1.25 µm – 2.5 µm range. Its measurement is performed by transmitting light into the skin in contact with an optical touch pad, and collecting a portion of the light diffusely reflected back by the skin. The collected light is analyzed to determine the tissue alcohol concentration. The challenge with TruTouch is weak ethanol absorption (overtones and combinations of the MIR fundamental band), and confounding absorptions from other skin tissue components to which NIR tissue spectroscopy is usually sensitive. This method generally requires an appropriately chosen tissue location and a well-cleaned skin surface in order not to further compromise the signal to noise ratio of the very low intensity diffuse reflection. Autoliv is a distant spectrometry, breath-based technology. It "sniffs" the air in the car with a mid-infrared (MIR) transmission spectroscopy sensor, working at the ethyl alcohol fundamental absorption band around 9.6 µm, see Fig. 1. Autoliv measures alcohol and CO_2 concentrations simultaneously, and takes samples of human breath diluted (slightly) in air with collection cups. Assuming the dilution factors of CO_2 and alcohol are the same, the device calculates alcohol concentration present in human breath. In practical application, the way of collecting diluted breath samples may impose a large variability of the measured BrAC value. In addition, breath CO_2 concentration varies from person to person and possibly with time as well. This would complicate the calibration procedure and introduce undesired device variability and false readings.
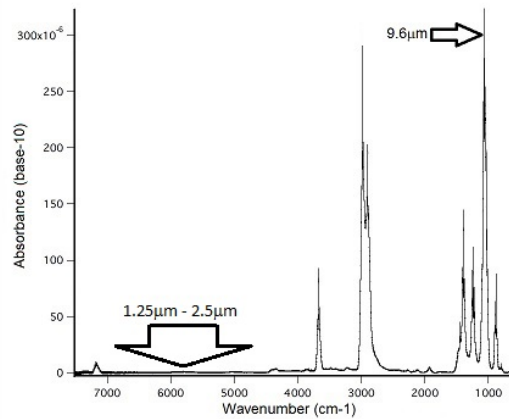
Fig. 1. Infrared ethanol alcohol absorption spectrum (1.4μm to 10 μm).

In comparison, Wavelength-Modulated Differential Photothermal Radiometry (WM-DPTR) is presented as a very promising technology for in-vehicle ignition interlock, capable of overcoming the above-mentioned difficulties. WM-DPTR is a novel mid-infrared thermophotonic, patented [7], ultrasensitive, noninvasive and non-contacting technique for measuring minute absorptions of low-concentration solutes in strongly absorbing fluids like water and blood [8–10]. Working in the MIR range, WM-DPTR eliminates the hurdle of weak absorption and confounding interferences. The difficulty associated with the well-known MIR shallow optical penetration depth due to tissue water content is compensated for by the unique photothermal property of WM-DPTR, that the laser-generated thermal wave can propagate deeper than the irradiating photons depending on modulation frequency and sample, the combined (amplified) optical and thermal property changes of the interstitial fluid (ISF).

2. Methods and materials

2.1 WM-DPTR mechanism and system

The WM-DPTR method involves out-of-phase modulated laser-beam excitation at two discrete wavelengths near the peak (~9.6 μm, laser A) and the baseline (~10.4 μm, laser B) of the ethanol absorption band, Fig. 2. Working at the fundamental MIR band, WM-DPTR takes advantage of the strongest ethanol absorption at 9.6 μm and eliminates the baseline variation through real-time differential measurements at the peak (9.6 μm) and the adjacent valley/baseline (10.4 μm), Fig. 2. This approach allows WM-DPTR to measure blood alcohol concentration (BAC) as mirrored in the ISF in the epidermis [10]. Another crucial property of WM-DPTR is sensitivity tunability: ethanol measurement sensitivity can be optimized for high measurement accuracy and precision, as required by DADSS [5], through careful selection of amplitude ratio R and phase shift ΔP of signals induced by the two laser beams. The detailed differential signal generation is discussed in Ref. 7.
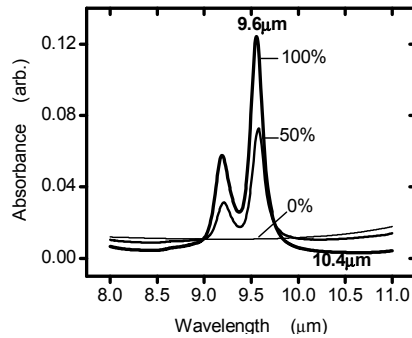
Fig. 2. MIR ethyl alcohol fundamental absorption band.

Figure 3 shows a diagram of the developed experimental WM-DPTR system. The system consists of two quantum cascade lasers (QCL, 1101-95/104-CW-100-AC, Pranalytica, CA) emitting at 9.6 µm (laser A) and 10.4 µm (laser B), a HgCdZnTe detector (MCZT, PVI-4TE-5, Vigo System, Poland) sensitive in the 2-5 µm spectral bandwidth, a function generator (33220A, Agilent Technologies, CA) producing two phase-locked square waves to modulate the laser beams, and a lock-in amplifier (SR850, Stanford Research Systems, CA). When the two out-of-phase square-wave-modulated laser beams irradiate the sample, a differential PTR signal is generated. The signal is collected by a pair of parabolic mirrors and focused onto the MCZT detector, the output of which is then sent to the lock-in amplifier for demodulation. The intensity ratio of the two lasers is controlled by a motorized variable circular neutral density filter (Reynard Corp, CA) placed in front of laser B and the phase shift between the two laser beams is sensitively controlled by the phase-locked function generator. Both laser output powers are *ca.* 34 mW with beam sizes ~2.5 mm. To simulate alcohol detection in the ISF of the epidermis layer, the laser modulation frequency which controls the WM-DPTR probing depth was set at 90 Hz in order to generate a probe depth < 40 µm in the epidermis layer (~60% water) below the 10-µm-thick stratum corneum (~10% water). The reference used in the following measurements is a sample with 0 BAC. The tuning of the amplitude ratio R and phase shift ΔP is achieved by adjusting the laser intensity ratio I_A/I_B and the optical phase shift of the two lasers.
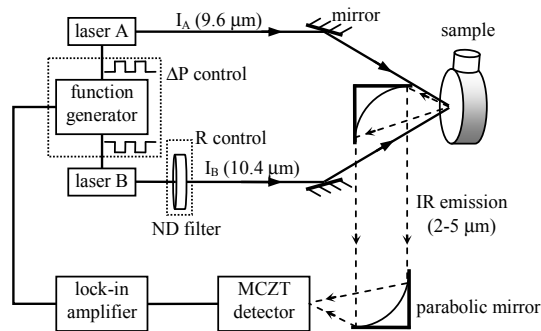

Fig. 3. Current WM-DPTR system configuration. The variable circular neutral density (ND) filter controls the amplitude ratio R. The function generator controls the phase shift ΔP.

2.2 Materials

Two types of phantoms were used in the measurements: (1) ethanol + water solutions and (2) ethanol + human blood serum solutions diffused into human skin. The sample solution was contained in a cylindrical cuvette with either a ZnSe window or human skin on one side facing the laser beams and a glass window on the other side, as indicated in Fig. 4. The

ethanol + water (serum) solutions were obtained by mixing anhydrous ethyl alcohol (GreenField Ethanol Inc. ON, Canada) with deionized water or base human serum (catalog number 1016011, American Biological Technologies Inc. TX) with ethanol concentrations 0 - ~100 BAC (1 BAC = 1 mg/dl). Accurate ethanol concentrations of the mixtures were determined independently using a biochemistry analyzer (YSI 2700S, Life Sciences, OH). The ethanol detection range was chosen to be close to, and include, the legal upper blood alcohol limit (50 BAC in Europe and 80 BAC in US and Canada). Even though driving with excess of blood alcohol above 80 BAC is a criminal offence, an individual's vision (brightness, color, depth, and motion perception) may already be affected and the brain's ability to perform simple motor functions is diminished with a 50 BAC, resulting in degraded driving performance and a significant increase in collision probability. The simplified water solutions were measured first to prove the principle of the WM-DPTR ethanol detection mechanism. For some measurements the concentration range was extended to 1000 BAC. Human serum was chosen because it is a good alternative to ISF [11]. For closest simulation of actual field measurement conditions, serum ethanol concentrations were measured through human skin in vitro. With the approval of the Research Ethics Office of the University of Toronto, human abdomen skin samples were obtained from abdominal plastic surgery ("tummy tuck" procedure), courtesy of TMB Cosmetic Plastic Surgery, Toronto Office. 'Dry' fixed skin (epidermis + dermis) of ~1 mm thickness was cut into a circular shape of ~25 mm diameter and glued onto the sample holder with the stratum corneum layer facing the laser beam.
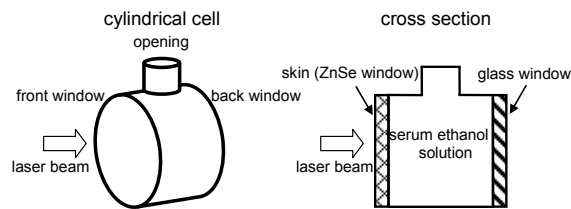

Fig. 4. schematic diagram of sample holder

3. Results and discussion

3.1 Ethanol in water measurements

In order to understand the advantages of the differential WM-DPTR method and compare it with single wavelength PTR measurements, ethanol + water solution measurements with single laser A and laser B were first performed in the range from 0 to 1000 BAC. The wide ethanol concentration range was chosen to have observable signal changes. Figure 5(a) presents the amplitude of single-PTR signals. The data points are the averages of five sequential measurements and the error bars were obtained from the standard deviation of the five measurements. It can be seen that PTR measurements with laser A are more sensitive that those with laser B as expected from the higher absorption coefficient at 9.6 µm. When ethanol concentration changes from 0 to 1000 BAC, the amplitude A_A from laser A, increases from 198 µV to 202.6 µV (3.6% increase), while the amplitude A_B from laser B, increases from 198 µV to 200.8 µV (only 1.4% increase). Figure 5(b) displays the phase of the single PTR signals. For 1000 BAC ethanol concentration rise, the phase P_A from laser A, increases 0.58°, from 90° to 90.58°, whereas the phase P_B from laser B, decreases 0.18°, from 90° to 89.82°.
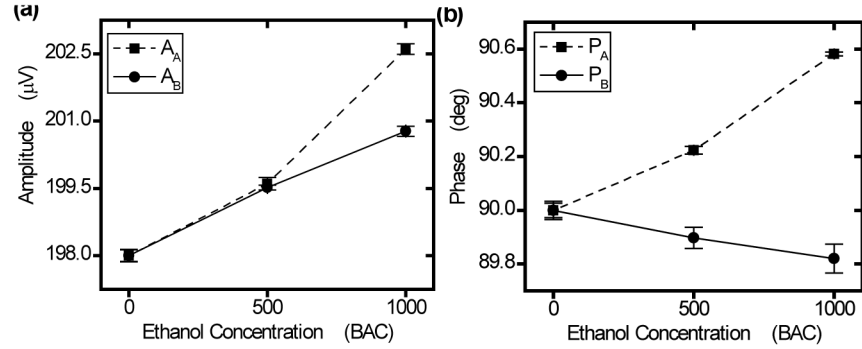
Fig. 5. Single PTR (with laser A and laser B) measurements of ethanol + water solutions at 90 Hz. (a) amplitude A_A (A_B); (b) phase P_A (P_B).

Generally speaking, the PTR signal is induced by both optical and thermal property changes in a sample. From Fig. 2 we can see that compared with the 9.6 μm wavelength (laser A), optical absorption only experiences small variations at 10.4 μm (laser B) when ethanol concentration increases. Thus, we can assume that the changes in A_B and P_B are solely due to sample thermal property (thermal diffusivity and thermal conductivity) changes with ethanol concentration, whereas the changes in A_A and P_A are the result of combined optical and thermal property changes. Taking both Laser A and Laser B changes combined in the PTR system, a 1000-fold BAC ethanol concentration increase in the sample generates asymmetrically a net phase shift P_A-P_B = 0.76° and amplitude ratio increase A_A/A_B = 1.009. These are the driving force of WM-DPTR signal amplification [3], however, they are too small as single-ended PTR responses for practical applications as an in-field alcohol biosensor.

Demonstrated in Fig. 6 are the WM-DPTR measurements of ethanol + water solutions in the range of 0 - ~100 BAC. It is shown that the ethanol concentrations are well resolved both with amplitude, Fig. 6(a), and phase, Fig. 6(b). For ~100 BAC ethanol concentration change, there is ~18% increase in amplitude and ~20° decrease in phase with R and ΔP combinations (1.02, 180.32°) and (0.99, 180.32°), respectively. Compared with the single-ended PTR signals as shown in Fig. 5, WM-DPTR signals are greatly amplified and make sensitive ethanol measurements below 100 BAC possible. It can be seen that the right selection of R and ΔP also renders amplitude and phase complementary: amplitude is more sensitive in the low concentration range and phase is more sensitive in the high concentration range. These trends are consistent with earlier results we obtained with blood glucose monitoring using WM-DPTR [8–10].
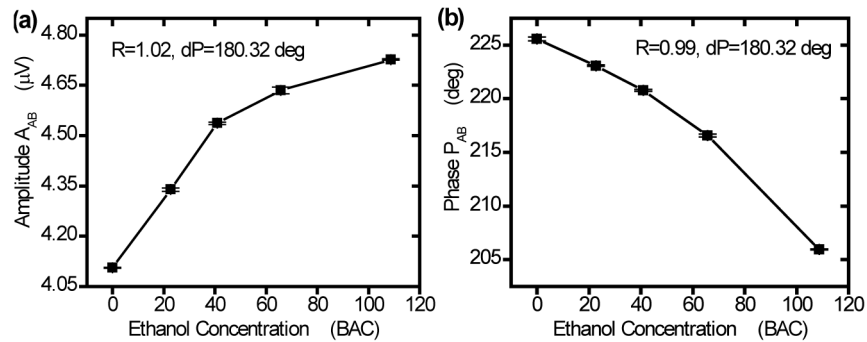

Fig. 6. WM-DPTR signals of ethanol + water solutions measured with various amplitude ratio R and phase shift ΔP combinations. (a) amplitude; (b) phase.

3.2 Ethanol in human skin measurements

In order to simulate more closely the ultimate goal of in vivo WM-DPTR measurements, further in vitro measurements were performed on ethanol in human blood serum diffused into a human skin sample. 25-minute waiting time was applied before the measurements started for each load of new solution so that the ethanol concentration in the skin would reach equilibrium through diffusion with that in the sample holder. Various R and ΔP combinations were used in the measurements to explore the ethanol measurement sensitivity and flexibility of WM-DPTR.

Figure 7 shows the ethanol measurement results with maximized sensitivity in the 0 – 100 BAC concentration range. For the given R and ΔP combinations (0.99, 180.53°) and (0.96, 180.53°), the amplitude, Fig. 7(a), drops 83% and the phase, Fig. 7(b), rises by 150° across the full ethanol concentration range. The amplitude and phase exhibit complementary sensitivity and nearly linear trends. The above-mentioned results underscore the sensitivity tunability property of WM-DPTR. Figure 8 demonstrates another feature of WM-DPTR, "diagnostic phase transition". With the shown R and ΔP combination (0.97, 180.53°), the phase only becomes sensitive to ethanol concentration when approaching the legal threshold and then plunges (~100°) when the legal limit is passed. This boundary sensitivity enhancement might be useful for quick in-vehicle or roadside Pass or Fail alcohol tests.
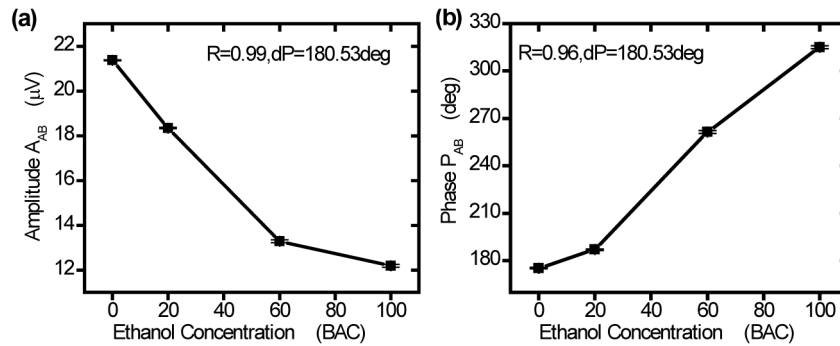

Fig. 7. WM-DPTR signals of ethanol + serum solutions diffused in human skin measured with different amplitude ratio R and phase shift ΔP combinations. (a) amplitude; (b) phase.
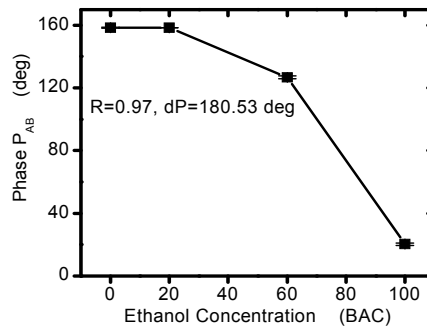

Fig. 8. WM-DPTR phase signal of ethanol + serum solutions diffused in human skin.

Single-PTR comparison measurements were also performed with laser A in the same ethanol concentration range and the results are shown in Fig. 9. It is further confirmed that the ethanol concentration is not resolvable with the conventional single-ended PTR method.
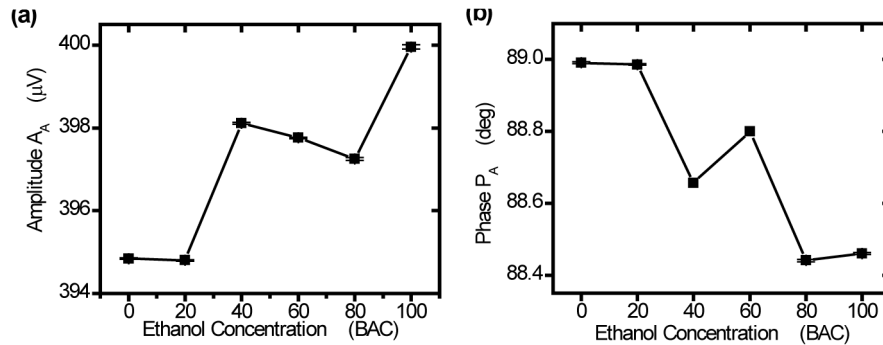
Fig. 9. Single PTR with laser A measurement of ethanol + serum solutions diffused in the human skin. (a) amplitude; (b) phase.

3.3 Confounding contributions from glucose

The glucose fundamental absorption peak is coincident with that of ethanol ~9.6μm. The metabolism-induced blood glucose variation (especially for people with diabetes) will inevitably affect the accuracy of BAC measurements for single-ended tissue spectroscopy. However WM-DPTR can minimize the glucose effect through its unique sensitivity-tuning capability. Because of the difference in thermal properties, ethanol can be differentiated from glucose by ethanol-optimized R and ΔP.

4. Conclusions

Preliminary ethanol phantom (ethanol-water, ethanol-human blood serum diffused from human skin sample) measurements in the countermeasures relevant range (0 – 100 BAC) with our current WM-DPTR system have demonstrated the potential and feasibility of WM-DPTR for non-invasive in-vehicle ignition interlock. The ethanol measurements with WM-DPTR have exhibited high sensitivity and high resolution which can be maximized upon combination of the complementary high sensitivities of amplitude and phase in the low (amplitude) and high (phase) concentration range. The unique sensitivity tunability feature of WM-DPTR through adjusting a pair of parameters, amplitude ratio R and phase shift ΔP, makes WM-DPTR versatile for a variety of applications. It was shown that single-ended photothermal measurements (A_A/P_A, A_B/P_B, measured only with either laser A or laser B) under the same conditions cannot resolve ethanol concentration differences in 0 - 100 BAC range. Future work will include reproducibility and repeatability studies of the measurements, detection range linearity tuning, in-vivo monitoring and system calibration for alcohol concentration determination.

Acknowledgments