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Application of modified difference absorption method to stand-off detection of alcohol in simulated car cabins

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Abstract. Some aspects of stand-off detection of alcohol in simulated car cabins are described. The proposed method is the well-known “difference absorption” method applied to the differential absorption lidar system, modified by taking advantage of a third laser beam. The modification was motivated by the familiar physical phenomena such as dispersion and different absorption coefficients in window panes for applied laser wavelengths. The mathematical expressions for the method were derived and confirmed by experiments. The presented investigations indicate that the method can be successfully applied to stand-off detection of ethyl alcohol in moving cars. © The Authors. Published by SPIE under a Creative Commons Attribution 3.0 Unported License. Distribution or reproduction of this work in whole or in part requires full attribution of the original publication, including its DOI. [DOI: [10.1117/1.JRS.7.073529](https://doi.org/10.1117/1.JRS.7.073529)]

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1 Introduction

There are many papers dealing with stand-off detection of different chemical and biological compounds with the use of a monochromatic laser beam at a wavelength fitting into the absorption spectra of these materials.¹⁻⁶ At the same time, many new developments in lasers that can be used in this application have been achieved in recent years.⁷⁻¹² Thus many devices based on this technology were presented in laboratories as well as applied to civil industry, environmental protection, and military. The differential absorption lidars (DIAL) are the most often used systems to monitor the atmosphere and detect contaminants in the form of gas, aerosol, fume, or dust. They make use of the well-known physical phenomenon of “difference absorption” using two laser beams at different wavelengths. Among many advantages of these systems are the ability to analyze the sample without the necessity to collect it, real-time investigation as well as integration of electro-optical systems and full automation of the measurement. Moreover the time needed to take measurements is equivalent to the speed of light; thus such lidars can be successfully used to face alcohol abuse among drivers.

Ethyl alcohol is a chemical compound that is characterized by high volatility, reflection of which is its high concentration on exhalation of people who have consumed it. Thus the car cabin where the drunken people are located is expected to be filled with air having some concentration of alcohol. In such situations, the simple DIAL system seems to work properly enough to detect the concentration of alcohol in the cabin, but it is not so. In order to assure the reliable results of measurements, the physical phenomena appearing in the side window panes of the car have to be taken into account. In this connection, a new method of detection was proposed and described in this article as well as in three patents and a study.¹³⁻¹⁵

2 Spectral Issues

To effectively detect chemicals in cars using the difference absorption method, it is essential to know not only its transmission spectra but also the transmission spectra of accompanying

substances as well as transmission spectra of the side window panes of cars. In this case the accompanying substances that have to be taken into consideration are carbon dioxide (CO_2) and water vapor (H_2O). In Fig. 1, the measured transmission spectra of vapor of ethanol (rectified spirit), CO_2 , vapor of H_2O , and window pane are presented. To measure the spectra, the first three chemical compounds were placed inside a 12-cm-long glass pipe terminated by quartz windows. The windowpane used for experiments was made of flint glass by Pilkington company and is the most often used window pane in cars.

Ethyl alcohol is characterized by a quite wide absorption band around the $3.39 \mu\text{m}$ wavelength, which is mainly caused by stretching vibrations of OH, CH, and CO bonds being included in $\text{C}_2\text{H}_5\text{OH}$ molecule.¹⁶ This situation is very useful for the choice of the wavelength absorbed by the alcohol, but at the same time it implies the necessity of choosing the reference wavelength, which lies relatively far from the previous one. The absorption band around $3 \mu\text{m}$ wavelength is caused by water that occurs in rectified spirit.

Keeping in mind the measured transmission spectra and availability of lasers, it seems that the He-Ne laser generating at $3.39 \mu\text{m}$ wavelength, strongly absorbed by the alcohol, is the best option. For reference beam, a laser diode generation at $1.5 \mu\text{m}$ wavelength was chosen. Using only two wavelengths is not enough to detect alcohol because of the window pane, which has different transmission for these wavelengths as well as different thicknesses in different cars. If the thickness was the same for all cars or the transmission was the same for both wavelengths, there would not be any problem with the detection. However, the differences are quite big and using the third wavelength, which will enable measurement of the thickness of the windowpane, becomes necessary. The third wavelength should have a transmission different from that of $1.5 \mu\text{m}$ wavelength and lie outside the absorption bands of alcohol, CO_2 , and H_2O . Laser diode generation at $1.3 \mu\text{m}$ wavelength seems to be a reasonable solution. The chosen wavelengths are indicated in Fig. 1 by straight vertical lines.

Very important phenomena that should be taken into account are dusting and misting over window panes. To face these problems, transmission spectra of clean window panes and those covered with water vapor, road dust, and graphite dust were measured and presented in Fig. 2.

To calculate the transmission of the layers of dust and water vapor, the transmissions of the covered window panes were divided by the transmission of the clean window pane at selected wavelengths. The results of the calculations are presented in Table 1, where T_1 is the transmission of the clean window pane, T_2 is the transmission of the window pane covered with water vapor, T_3 is the transmission of the window pane covered with road dust, T_4 is the transmission of the window pane covered with graphite dust, T_w is the transmission of water vapor, T_{rd} is the transmission of road dust, T_{gd} is the transmission of graphite dust, and λ is the wavelength.

Looking at the results of calculations it seems that the appearance of dust does not have any influence on the results of measurement of the concentration of alcohol, because its transmission

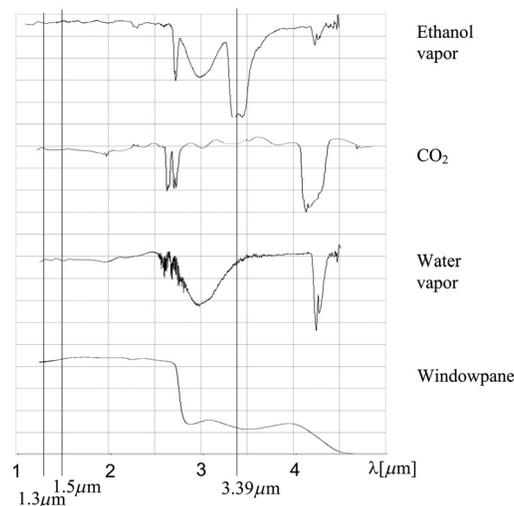


Fig. 1 Transmission spectra of chemicals that can appear on the way of the laser beams.

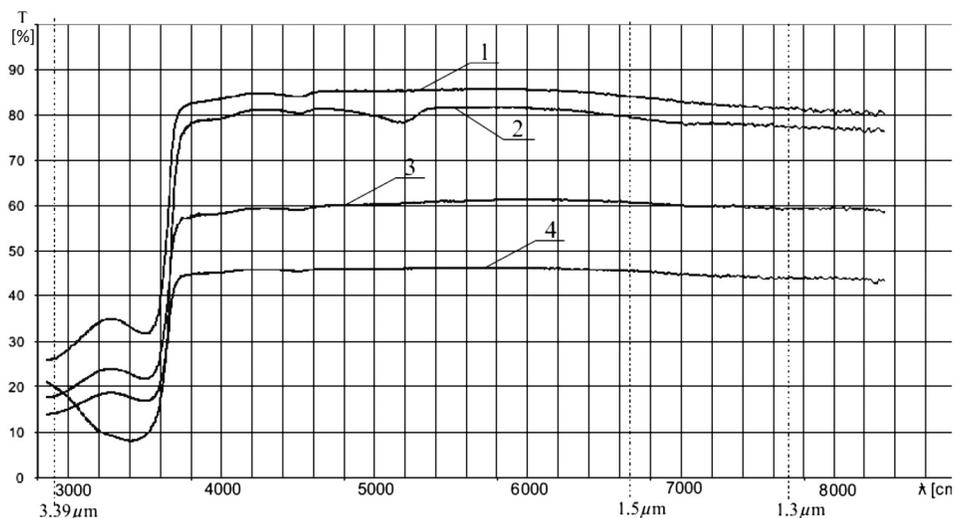


Fig. 2 Transmission spectra of clean windowpane (1), windowpane covered with water vapor (2), windowpane covered with road dust (3), windowpane covered with graphite dust (4).

Table 1 Transmissions of the layers of dust and water vapor.

λ (μm)	T_1 (%)	T_2 (%)	T_w	T_3 (%)	T_{rd}	T_4 (%)	T_{gd}
1.3	81.3178	77.3306	0.951	43.99877	0.541	59.22023	0.728
1.5	84.01771	79.41549	0.945	45.46778	0.541	60.62766	0.722
3.39	26.97764	19.15639	0.710	14.44367	0.535	18.40899	0.682

at the investigated wavelengths is almost the same. Water vapor has lower transmission at 3.39 μm and then at 1.3 and 1.5 μm wavelengths, which is not desired; however, the difference is so small that it should not interfere with the detection of alcohol.

3 Optical Parameters of Windowpane

The window pane used for the experiments was the same as in the previous investigations. The transmission spectra of the window pane before and after thermal processing (tempering and bending) with thickness of 3.15 mm were measured and are presented in Fig. 3.

In Table 2 the transmissions $T(\lambda)$ of the window panes at the investigated wavelengths λ are presented. As can be seen from the table, the tempering does not significantly change the transmission of the window pane at any of the investigated wavelengths. Thus for the next experiments, a window pane that was not tempered was used since it is easier to machine (cutting, grinding, polishing).

Because the optical parameters of the float glass used for the production of the window panes are not available from the producer in the investigated spectral range, they had to be measured. The parameters that are indispensable for accurate detection of alcohol are absorption coefficient and refractive index.

In order to calculate the absorption coefficient of the window pane, the transmission spectra of two samples of float glass with thickness of $d_1 = 1.5$ mm and $d_2 = 3$ mm were measured. The transmission spectra are presented in Fig. 4, while in Table 3 the transmissions $T(\lambda)$ at the investigated wavelengths λ are shown.

Assuming that there is no scattering on the surface of the sample and inside it, the intensity of the radiation going through it can be written as

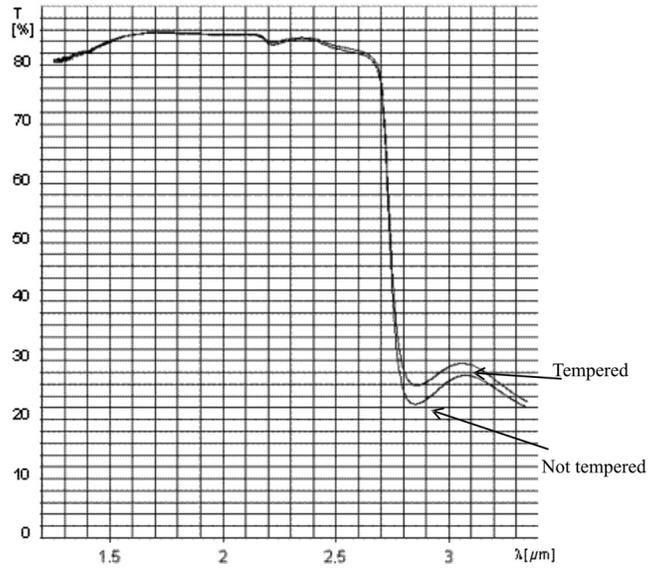


Fig. 3 Transmission spectra of windowpane before and after thermal processing.

Table 2 Transmissions of windowpanes at the investigated wavelengths.

Type of windowpane	$T (1.3 \mu\text{m})$	$T (1.5 \mu\text{m})$	$T (3.39 \mu\text{m})$
Tempered	0.8092	0.8391	0.2229
Not tempered	0.8136	0.8407	0.2138

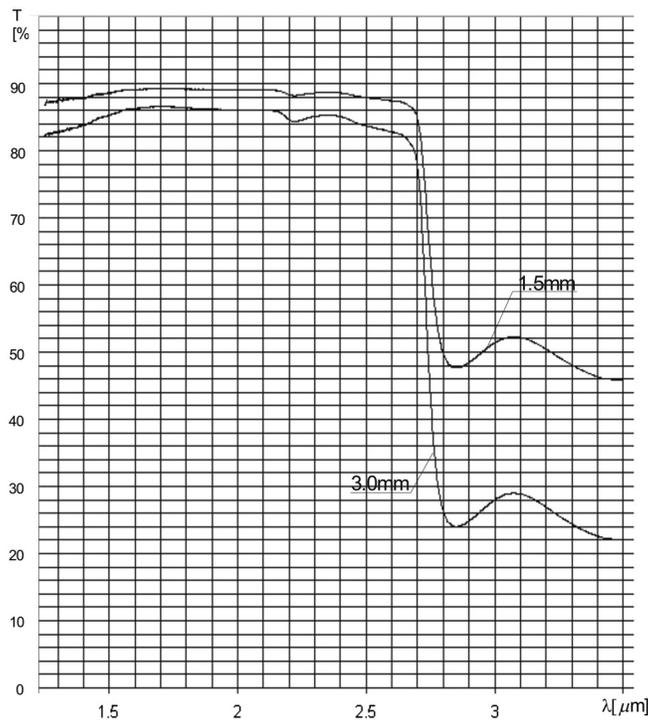


Fig. 4 Transmission spectra of two samples of float glass with thickness of 3 and 1.5 mm.

Table 3 Transmissions $T(\lambda)$ at the investigated wavelengths λ .

Thickness of the sample	$T(1.3 \mu\text{m})$	$T(1.5 \mu\text{m})$	$T(3.39 \mu\text{m})$
1.50	0.87514	0.88497	0.46484
3.00	0.82892	0.85193	0.22747

$$I = I_0(1 - r)e^{-\kappa d}(1 - r) + I_1 r e^{-\kappa d} r e^{-\kappa d}(1 - r) + \dots, \tag{1}$$

where I_0 is the intensity of the incident radiation,

$$I_1 = I_0(1 - r)e^{-\kappa d}, \tag{2}$$

r is the reflection coefficient, κ is the absorption coefficient, and d is the thickness of the sample.

In the investigated situation,

$$r^2 e^{-\kappa d} \ll r, \tag{3}$$

and the transmission of the sample can be described as

$$T = \frac{I}{I_0} = (1 - r)^2 e^{-\kappa d}. \tag{4}$$

Introducing the following notation for radiation at a specified wavelength λ ,

$$F(\lambda) \equiv F_\lambda,$$

Eq. (4) can be written as

$$T(d, \lambda) = T_{0\lambda} e^{-\kappa_\lambda d}, \tag{5}$$

where

$$T_{0\lambda} \equiv [1 - r(\lambda)]^2. \tag{6}$$

For the samples with thickness of $d_1 = 1.5$ mm and $d_2 = 3$ mm,

$$T_{d2\lambda} = T_{0\lambda} e^{-\kappa_\lambda 3}, \tag{7}$$

$$T_{d1\lambda} = T_{0\lambda} e^{-\kappa_\lambda 1.5}. \tag{8}$$

After dividing Eq. (7) by Eq. (8),

$$\frac{T_{d2\lambda}}{T_{d1\lambda}} = e^{-\kappa_\lambda 1.5}.$$

Thus

$$\kappa_\lambda = \frac{1}{1.5} \ln\left(\frac{T_{d1\lambda}}{T_{d2\lambda}}\right). \tag{9}$$

Taking into account the transmissions from Table 3, the absorption coefficients for the investigated wavelengths are

$$\kappa_{1.3} = 0.0384 \text{ mm}^{-1}, \quad \kappa_{1.5} = 0.0266 \text{ mm}^{-1}, \quad \kappa_{3.39} = 0.479 \text{ mm}^{-1}. \tag{10}$$

Squaring both sides of Eq. (8),

$$(T_{d1\lambda})^2 = (T_{0\lambda})^2 e^{-\kappa_\lambda^3}. \tag{11}$$

Dividing Eq. (11) by Eq. (7),

$$\frac{(T_{d1\lambda})^2}{T_{d2\lambda}} = T_{0\lambda}. \tag{12}$$

Thus for three investigated wavelengths,

$$T_{01.3} = 0.92394, \quad T_{01.5} = 0.91929, \quad T_{03.39} = 0.94991. \tag{13}$$

The refractive index of the window pane was determined by the use of the experimental setup shown in Fig. 5. During the experiment the laser beams were passing through the wedge made of flint glass with the vertex angle of $\Phi = 9^\circ 40'$ and the angular deviation δ was measured by a detector. The experiment was repeated for three investigated wavelengths.

The measured angular deviations were equal to

$$\delta_{1.3} = 5^\circ 2', \quad \delta_{1.5} = 4^\circ 58', \quad \delta_{3.39} = 4^\circ 37'.$$

Using the expression

$$n_\lambda = \frac{\sin(\varphi + \delta)}{\sin \varphi}, \tag{14}$$

the refractive indexes can be calculated to be equal to

$$n_{1.3} = 1.51, \quad n_{1.5} = 1.50, \quad n_{3.39} = 1.47. \tag{15}$$

The calculated absorption coefficients, refractive indexes, and transmissions $T_0(\lambda)$ are presented in Table 4.

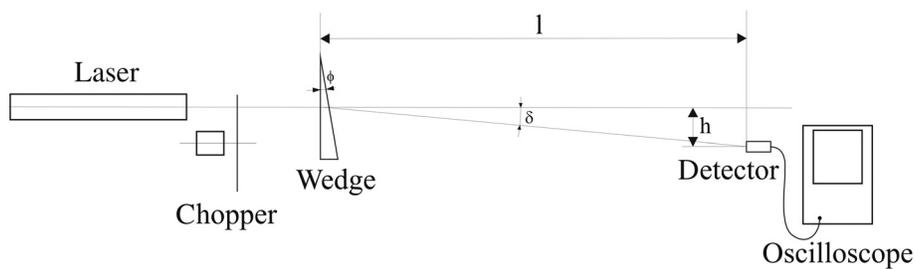


Fig. 5 Experimental setup for determining the refractive index of the windowpane.

Table 4 Calculated absorption coefficients, refractive indexes, and transmissions $T_0(\lambda)$.

λ (μm)	1.3	1.5	3.39
κ_λ (mm^{-1})	0.0384	0.0266	0.4785
n_λ	1.51	1.50	1.47
$T_{0\lambda}$	0.9239	0.9193	0.9499

4 Derivation of the Modified Difference Absorption Method

The commercially available lasers are characterized by relatively high power instability. Moreover, the strong absorption of the 3.39 μm wavelength by a window pane implies the necessity to use detectors with very high sensitivity, which, in the case of lack of window pane, causes saturation of the detectors. These obstacles can be solved by the application of comparative method using two detectors for measuring the optical power before and behind the glass pipe simulating a car and additionally using the reference glass pipe with known optical parameters. The method is schematically presented in Fig. 6.

In the absence of any glass pipe, the laser beam is divided by the beam splitter into beam I_{01} incident on detector 1 and beam I_{02} incident on detector 2. Because of the instability of the laser, its power can change in time by the index k . Thus, after inserting the investigated glass pipe in the laser beam, the intensity can be described as

$$I \rightarrow Ik.$$

As a result of this, the transmission of the glass pipe can be expressed as

$$T_k = \frac{I_2}{kI_{02}}. \tag{16}$$

Assuming that there is no change of polarization and power distribution in the laser beam, the k index can be described as

$$k = \frac{I_1}{I_{01}}.$$

Inserting this expression into Eq. (16),

$$T_k = \frac{I_2 I_{01}}{I_{02} I_1} = \frac{I_{01} I_2}{I_{02} I_1}. \tag{17}$$

Introducing notations

$$\frac{I_2}{I_1} \equiv i, \quad \frac{I_{02}}{I_{01}} \equiv i_0, \tag{18}$$

Eq. (17) can be written as

$$T_k = \frac{i}{i_0}. \tag{19}$$

Inserting the reference glass pipe with known transmission T_{wk} in the laser beam and measuring the intensities I_{2w} and I_{1w} and at the same time introducing the expression $i_w \equiv I_{2w}/I_{1w}$, the equation for the transmission T_{wk} can be written as

$$T_{wk} = \frac{i_w}{i_0}.$$

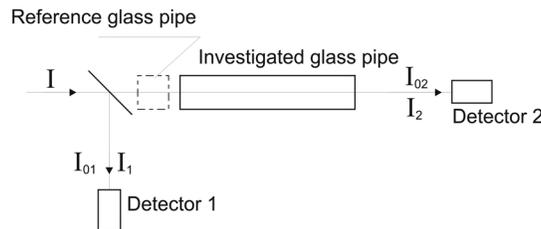


Fig. 6 Diagram of comparative method.

Thus,

$$i_0 = \frac{i_w}{T_{wk}}.$$

After inserting this into Eq. (19), the transmission of the investigated glass pipe is

$$T_k = T_{wk} \frac{i}{i_w}. \tag{20}$$

Assuming that the reference glass pipe is terminated by glass windows with thickness of d and is empty, its transmission can be described as

$$T_{wk} = (T_0)^2 e^{-2\kappa d}. \tag{21}$$

On the other hand, the transmission of the investigated glass pipe filled with alcohol vapor with transmission of T_a terminated by the same windows as in the case of the reference glass pipe, covered with some material with transmission of T_p , can be expressed as

$$T_k = (T_0)^2 T_a T_p e^{-2\kappa l}, \tag{22}$$

where l is the distance inside the window that the laser beam has to pass through.

After inserting Eqs. (21) and (22) into Eq. (20),

$$T_a T_p = \frac{i}{i_w} e^{2\kappa(l-d)}. \tag{23}$$

For a wavelength λ , Eq. (23) can be written as

$$T_a(\lambda) T_p = \frac{i_\lambda}{i_{w\lambda}} e^{2\kappa_\lambda[l(\lambda)-d]}. \tag{24}$$

If the normal to the window pane with thickness of d is not parallel to the laser beam, the distance that the laser light has to pass through inside the window can be described as

$$l(\lambda) = \frac{d}{\sqrt{1 - \frac{\sin^2 \alpha}{n_\lambda^2}}}, \tag{25}$$

where α is the angle of incidence of the laser beam onto the window pane.

Taking into account the calculated refractive indexes from Table 4, the relative difference of l for extreme wavelengths (1.3 and 3.39 μm) at $\alpha = 20$ deg can be expressed as

$$\delta \equiv \frac{l(1.3) - l(3.39)}{l(1.3)} < \frac{\frac{d}{\sqrt{1 - \frac{\sin^2 20^\circ}{1.51^2}}} - \frac{d}{\sqrt{1 - \frac{\sin^2 20^\circ}{1.47^2}}}}{\frac{d}{\sqrt{1 - \frac{\sin^2 20^\circ}{1.51^2}}}} = 0.0015. \tag{26}$$

In this case, it can be assumed that for $\alpha < 20$ deg

$$l(1.3) \approx l(1.5) \approx l(3.39) = l. \tag{27}$$

Thus Eq. (24) can take the form of

$$T_a(\lambda) \cdot T_p = \frac{i_\lambda}{i_{w\lambda}} e^{2\kappa_\lambda \Delta}, \tag{28}$$

where $\Delta = l - d$.

Assuming that the difference between l and d is very small (at $\alpha = 20$ deg, $2\kappa_{3.39}\Delta = 0.08$), it can be written that

$$e^{2\kappa_l\Delta} \approx 1 + 2\kappa_l\Delta. \tag{29}$$

In this case, Eq. (28) for the investigated wavelengths takes the form of

$$T_p T_a = \frac{i_{3.39}}{i_{w3.39}} (1 + 2\kappa_{3.39}\Delta), \tag{30}$$

$$T_p = \frac{i_{1.5}}{i_{w1.5}} (1 + 2\kappa_{1.5}\Delta), \tag{31}$$

$$T_p = \frac{i_{1.3}}{i_{w1.3}} (1 + 2\kappa_{1.3}\Delta). \tag{32}$$

After solving the above equations,

$$\Delta = \frac{1}{2\kappa_{1.3}} \frac{1 - \frac{i_{w1.3} i_{1.5}}{i_{w1.5} i_{1.3}}}{\frac{\kappa_{1.5} i_{w1.3} i_{1.5}}{\kappa_{1.3} i_{w1.5} i_{1.3}} - 1}, \tag{33}$$

$$T_p = \frac{(\kappa_{1.5} - \kappa_{1.3})}{\kappa_{1.3}} \frac{\frac{i_{1.5}}{i_{w1.5}}}{\frac{\kappa_{1.5} i_{w1.3} i_{1.5}}{\kappa_{1.3} i_{w1.5} i_{1.3}} - 1}, \tag{34}$$

$$T_a = \frac{\kappa_{1.5} - \kappa_{3.39}}{\kappa_{1.5} - \kappa_{1.3}} \frac{i_{3.39}}{i_{w3.39}} \frac{i_{w1.3}}{i_{1.3}} - \frac{\kappa_{1.3} - \kappa_{3.39}}{\kappa_{1.5} - \kappa_{1.3}} \frac{i_{3.39}}{i_{w3.39}} \frac{i_{w1.5}}{i_{1.5}}. \tag{35}$$

Inserting the notation

$$a_\lambda \equiv \frac{i_\lambda}{i_{w\lambda}}, \tag{36}$$

Eqs (33), (34), and (35) can be expressed as

$$\Delta = \frac{1}{2\kappa_{1.5}a_{1.5} - \kappa_{1.3}a_{1.3}} (a_{1.3} - a_{1.5}), \tag{37}$$

$$T_p = a_{1.3}a_{1.5} \frac{\kappa_{1.5} - \kappa_{1.3}}{\kappa_{1.5}a_{1.5} - \kappa_{1.3}a_{1.3}}, \tag{38}$$

$$T_a = \frac{\kappa_{1.5} - \kappa_{3.39}}{\kappa_{1.5} - \kappa_{1.3}} \frac{a_{3.39}}{a_{1.3}} - \frac{\kappa_{1.3} - \kappa_{3.39}}{\kappa_{1.5} - \kappa_{1.3}} \frac{a_{3.39}}{a_{1.5}}. \tag{39}$$

Inserting the calculated absorption coefficients into the above equations,

$$\Delta = \frac{1}{2 \cdot 0.0266a_{1.5} - 0.0384a_{1.3}} (a_{1.3} - a_{1.5}), \tag{40}$$

$$T_p = a_{1.3}a_{1.5} \frac{0.0118}{0.0384a_{1.3} - 0.0266a_{1.5}}, \tag{41}$$

$$T_a = 38.297 \frac{a_{3.39}}{a_{1.3}} - 37.297 \frac{a_{3.39}}{a_{1.5}}. \tag{42}$$

5 Experiment

The investigations of the detection of alcohol in the glass pipe simulating a car cabin were carried out in the setup presented in Fig. 7. Four lasers generating at 3.39, 1.5, 1.3, and 0.6 μm wavelengths were used. Their beams were combined into one single beam by glass plates with appropriate thin-layer coatings. The laser generating wavelength at 0.6 μm was used as a pointer to make the adjustment of the system easier. All lasers worked in continuous regime, so the chopper for modulation was used. Small displacement of different laser beams at the surface of the chopper enabled separation of the beams in time domain so as to have been detectable by only two detectors. Two glass pipes with lengths of 140 cm and diameter of 5 cm terminated by the flint glass with thickness of 3.15 mm were used. One of them was the reference glass pipe with clean windows that were perpendicular to the laser beam. The second one was the investigated glass pipe filled with different concentrations of alcohol vapor with windows at angles of 15 deg to the laser beam, covered with graphite dust.

Two PbSe detectors were used and the signals were registered by an oscilloscope. In Fig. 8 the picture of the screen of the oscilloscope is presented. The experimental results and calculations are shown in Table 5.

Inserting the calculated a_λ in Eq. (42), the transmission of the alcohol can be expressed as

$$T_a = 1.4784a_{3.39}. \quad (43)$$

To prepare the required concentration of the alcohol vapor inside the investigated glass pipe, the appropriate volume of saturated vapor of alcohol was injected into it with the use of a syringe. On the basis of physical tables, the pressure of saturated alcohol vapor was assumed to be 60 hPa at the temperature of 20°C. Thus the concentration of the alcohol vapor in mole fraction can be described as

$$C = \frac{60 \text{ hPa}}{1013.2 \text{ hPa}} = 0.059. \quad (44)$$

Changing the units into particle per million (ppm), the concentration can be expressed as

$$C = 0.059 \times 10^6 = 59,000 \text{ ppm}$$

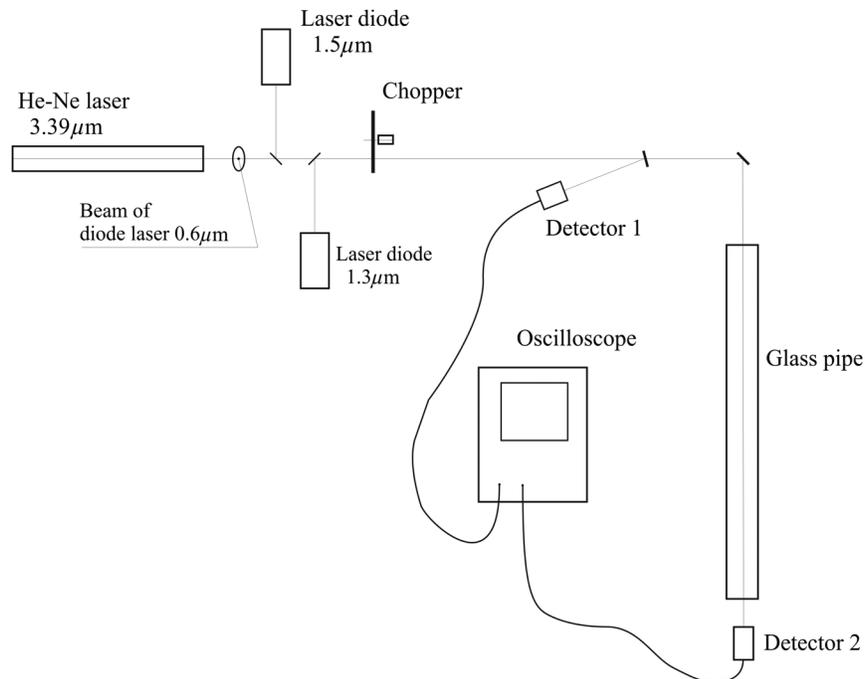


Fig. 7 Experimental setup for detection of alcohol in glass pipe.

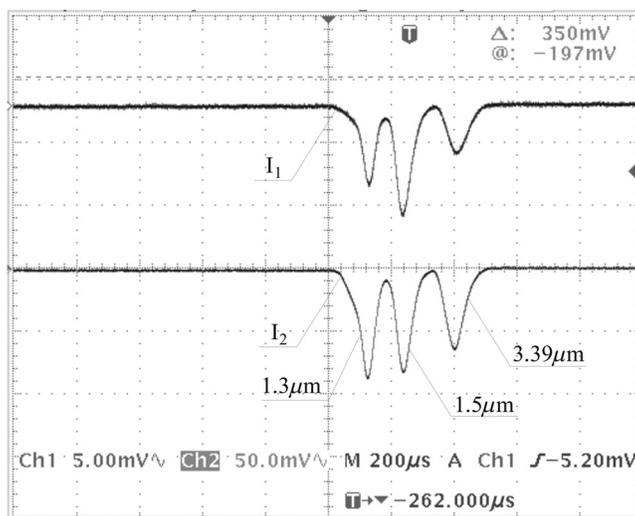


Fig. 8 Picture of the screen of the oscilloscope.

Table 5 Experimental results and calculations.

λ (μm)	$\bar{I}_{1w}(\lambda)$ (mV)	$\bar{I}_{2w}(\lambda)$ (mV)	$i_{w\lambda}$	$\bar{I}_1(\lambda)$ (mV)	$\bar{I}_2(\lambda)$ (mV)	i_λ	a_λ
1.3	1.3926	4.6149	3.3139	1.4134	3.3181	2.3476	0.7084
1.5	1.3451	4.8012	3.5694	1.3637	3.4526	2.5318	0.7093
3.39	1.4948	3.2763	2.1918	1.4536	2.1549	1.4825	0.6764

Multiplying C by the ratio of the volume of the injected alcohol vapor and the volume of the glass pipe (2749 cm^3), the concentration of the alcohol vapor in the investigated glass pipe was determined. The results of the investigations for different concentrations of alcohol vapor are shown in Table 6. In Fig. 9 the transmission of alcohol vapor as a function of concentration is presented.

In the presented experiment the accuracy of measurement of the signals from the detectors appears to be very crucial in order to assure reliable results. Applying the uncertainty theory and

Table 6 Results of investigations of detection of alcohol vapor at different concentrations.

V_a (cm^3)	C (ppm)	$\bar{I}_{1,3.39}$ (mV)	$\bar{I}_{2,3.39}$ (mV)	$i_{3.39}$	$a_{3.39}$	T_a
0	0	1.4536	2.1549	1.4825	0.6764	1
10	211	1.4231	1.8778	1.3195	0.6020	0.89
20	422	1.4028	1.6431	1.1713	0.5344	0.79
30	633	1.4357	1.5111	1.0525	0.4802	0.71
40	844	1.4412	1.4101	0.9784	0.4464	0.66
50	1055	1.4508	1.2474	0.8598	0.3923	0.58
60	1266	1.4621	0.9970	0.6819	0.3111	0.46
70	1477	1.4413	0.8548	0.5931	0.2706	0.40
80	1688	1.4217	0.7799	0.5486	0.2503	0.37
90	1899	1.4315	0.7217	0.5041	0.2300	0.34
100	2110	1.4371	0.6819	0.4745	0.2165	0.32

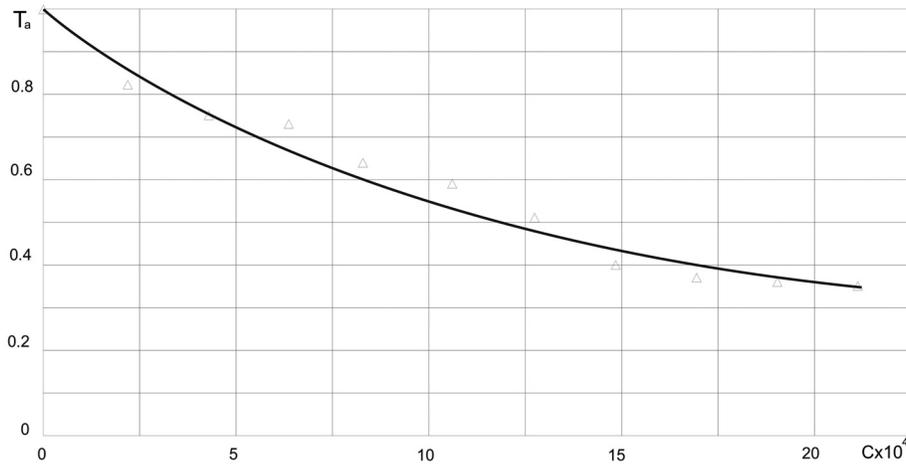


Fig. 9 Transmission of alcohol vapor as a function of concentration.

assuming that a man having a concentration of alcohol of 0.2‰ in his blood breathes out 0.1 mg/dcm³ of it, which corresponds with the alcohol concentration of 50 ppm,^{17,18} the accuracy of the detection of relative value of the laser radiation should be at least $\Delta i < 3 \times 10^{-4}$.

6 Conclusion

The results of the experiments prove that the proposed modified difference absorption method of detection of alcohol is reliable. Even if there is an angular tilting of the car windows with respect to the laser beam and car windows are dirty, the proposed solution can work properly. Thus the development of a trustworthy device seems to be feasible.

Acknowledgments

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