

Iodine Molecules Differential Absorption Cross section Lidar Studies

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Lidar monitoring of the gaseous molecules in atmosphere can be used for concentration measurements of the toxic pollutants in air of the urban and industrial regions. Such a concentration study in the multi-components gaseous mixture by lidar technique is a complex problem in atmospheric pollution monitoring. The differential absorption lidar (DA-lidar) is the most preferable for the detection of iodine molecules at very low concentration levels. The iodine molecules absorption cross section in the laboratory lidar studies and lidar equation simulation in this lidar variant corresponds with earlier data.

Key words: iodine molecules, concentration, lidar, differential absorption cross section

1. INTRODUCTION

LIDAR SYSTEMS for gaseous molecule concentration measurements in multi - component gas mixture monitoring have been realized by any spectroscopic variants determined by the studied molecules [1 – 4]. Molecular iodine concentrations measurements in technological gases and atmosphere upper AES or other radiochemical enterprises are of interest because its heavy isotopes ¹³¹I₂ can serve as indicators of air quality [4]. The lidar equation computer simulation has been performed earlier in [2, 5, 7] for all lidars and it has been concluded that differential absorption lidar (DA-lidar) is the most preferable for the iodine molecules detection at low concentration levels. The goal of this paper is experimental studies of molecular iodine DA-lidar differential absorption coefficient versus iodine concentration dependences and DA-lidar equation computer simulation.

2. EXPERIMENTAL SETUP

The laboratory experimental setup is shown in Fig.1. YAG: Nd Q-switching laser (1) beam with pulse duration of 10 ns and pulse energy about 25 mJ at wavelength 532 nm and 40 mJ at 1064 nm of laboratory on-axial lidar with ranging distance up to 8 m was directed into special Brewster windows – ended by a vacuum cuvette (4) with I₂ molecules. The 532 nm wavelength YAG: Nd laser (1) radiation intensity of laboratory lidar passing through cuvette (4), was directed to FD-24K type photodiode (6) with interference filter (7) at this wavelength. The 1064 nm wavelength laser (1) radiation intensity passing through vacuum cuvette (4) was directed by the dichromatic mirror (5) to the same type photodiode (9) with interference filter (8) too. It served as reference beam in the differential measuring scheme. The portions of both wavelengths of the laser (1) beams were turned by two glass plates (2) and (3) to the same type of photodiode (12) and (11) with two interference filters (13) and (14). The I₂ concentration in

cuvette (4) was varied by cuvette appendix heating in the water heater (10) from 25 to 85°C. Four photodiode pulse outputs of the same type were recorded by memory oscilloscope or imported into a special PC – controlled measuring system as the DA coefficient X that can be determined from DA-lidar equation [2, 8] and is equal to $X = \sigma_0 N \Delta R$, where the absorption cross section, iodine molecules concentration and absorbing layer thickness multiplier is in the right part. The cuvette (4) thickness from window to window is about 10 cm. The iodine molecules concentration level N was varied from $7 \cdot 10^{14} \text{ cm}^{-3}$ up to $3.4 \cdot 10^{16} \text{ cm}^{-3}$ in our case. The sensing distance range of 1 ... 8 m has been used in this study.

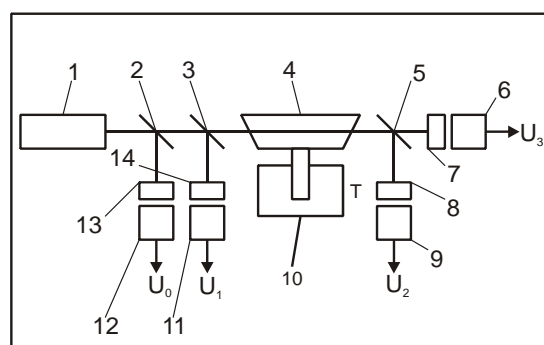


Fig.1. Experimental lidar layout

3. DA – LIDAR STUDY RESULTS

The 532 nm wavelength YAG: Nd laser radiation intensity of laboratory DAL lidar [2,6] that passed through cuvette was decreased due to iodine molecules absorption [3, 9] and has been directed to the FD-24K type photodiode with interference filter at this wavelength. The 1064 nm wavelength laser FH radiation intensity that passed through vacuum cuvette with I₂ has been recorded too. It served as reference beam in the differential measuring scheme. Four

photodiode pulse outputs of the same type were recorded by memory oscilloscope. These photodiode output measuring results at 4 m distance, for example U_0 and U_1 - the 1064 and 532 nm wavelength signals before cuvette, but U_2 and U_3 - the same wavelength signals after passing through the cuvette in Fig.1., were converted into pulse energy values. The calibration studies were at first fulfilled for the photodiode receiving module transmission coefficient measuring. This transmission coefficient value $K_f = 4.5 \pm 0.5$ mJ/V was established by photodiode output amplifier coefficient change. These results allowed us to convert the output amplitude to pulse energy E in Table 1. The DA coefficient X values are shown in the last column of Table 1. The average measurement ratio error was equal to 8.5 % in our case. The measuring results at the sensing distance range from 1 to 8 m were coincided with our measurement accuracy. The plot of DA coefficient X (in arbitrary units) versus iodine molecules concentration N in 10^{15} cm^{-3} is exhibited in Fig.2. The iodine molecules absorption cross section at 532 nm wavelength was calculated as in [1]. It equals $\sigma_0 = (1.88 \pm 0.37) 10^{-18} \text{ cm}^2$ in satisfactory confidence with data [3] - $4.6 10^{-18} \text{ cm}^2$ at 590 nm wavelength.

Table 1. The measured 1064 and 532 nm wavelengths DA pulse energy values, calculated iodine molecules concentration and DA coefficient values.

$N 10^{15}, \text{ cm}^{-3}$	$E_0, \text{ mJ}$	$E_1, \text{ mJ}$	$E_2, \text{ mJ}$	$E_3, \text{ mJ}$	$\Delta E_0, \text{ mJ}$	X
7.42	45	20	23	11.7	0.9	0.51
14.8			23	13		0.59
18.2			22	13.8		0.63
23.9			23	16		0.69
27.5			23	17		0.75
32.4			22	18		0.82

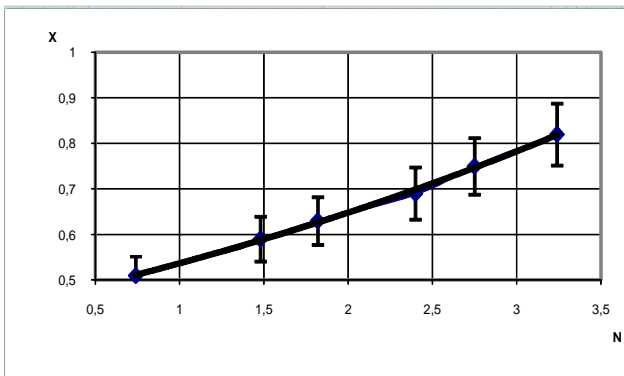


Fig.2. Plot of the DA coefficient X values vs. iodine molecules concentration N in 10^{15} cm^{-3} at sensing distance of 4 m.

4. DA-LIDAR EQUATION SIMULATION

The molecular iodine DA-lidar differential absorption coefficient versus iodine concentration dependences experimental studies can be confirmed by the DA-lidar equation computer simulation. The resonance absorption in the visible and infrared region of the spectrum has the

interaction maximal cross section [3], [8] from all of the spectroscopic effects. The molecule low level concentration at the large ranging distance measurement possibilities was determined by this absorption. As it has been shown earlier, DA-lidar has two laser beams with different wavelengths, the first wavelength lies in the molecule absorption line maximum and the second wavelength lies out of this line and weakening of these beams was determined by the iodine molecules in the atmosphere. The studied molecule concentration distribution in atmosphere data were calculated by the recorded DA-lidar signal ratio at these two wavelengths in the narrow spectral interval [1, 3].

DA-lidar equation [2, 3, 8] simulation has been fulfilled for I_2 molecules sensing possibilities estimation and molecules concentration and ranging distance ranges determination. The iodine molecules spectrum in the visible and infrared region consists of the electronic, vibration and rotation bands transitions from the basic state $X^1 \Sigma^+_{0g}$ to the state $A^3 \Pi_{1u}$, or $B^3 \Pi^+_{0u}$ and $I^1 \Pi_u$ [9] and the bands of such a transition from X to the first electronic state B , lie in the range from 499 up to 670 nm. The 532 nm wavelength YAG: Nd laser lies in this absorption band [1, 9] and such an experimental situation is of interest for DA-lidar equation computer simulation.

The lidar equation for the elastic back scattering was rewritten by [3, 8] in the form

$$P(\lambda, R) = P_L(\lambda_L) K_1 A_0 T^2(\lambda_L, R) \rho / R^2 \quad (1)$$

where $P(\lambda, R)$ - the back scattering signal power at the photodetector at the wavelength λ_L from the distance R ; $P_L(\lambda_L)$ - the peak power of the laser radiation and its wavelength λ_L ; K_1 - lidar constant; A_0 - the receiving telescope square; $T(\lambda_L, R)$ - the atmospheric transmittance at the wavelength λ_L ; ρ - the topographic target reflection coefficient. All of the information about iodine molecules concentration is in the multiplier $T(\lambda_L, R)$, meaning that the extinction coefficient $k(\lambda_L, R)$ can be exhibited in the general case as in [3, 8] in the equation

$$k(\lambda_L, R) = k_A(\lambda_L, R) + N_a(R) \sigma_0(\lambda_L) \quad (2)$$

where the first adding is the atmospheric extinction coefficient at the laser radiation wavelength without the iodine molecules and the second one is the I_2 molecules concentration and the resonance absorption cross section multiplication of these molecules.

The next step due to the differential absorption method [8] is dividing the two lidar equations (1) for the two laser radiation wavelengths λ_0 and λ_1 , where the second laser radiation wavelength is out of the I_2 absorption band, by one another. The dividing result is the differential absorption equation for the general case in the next form

$$\frac{P(\lambda_0, R)}{P(\lambda_1, R)} = \frac{P_0 K_{10} \rho_0}{P_1 K_{11} \rho_1} \exp \left\{ -2 \int_0^R [k(\lambda_0, R) - k(\lambda_1, R)] dR \right\} \quad (3)$$

The parameters in (3) in our experimental case are the next. The two laser radiation wavelengths are $\lambda_0 = 532$ nm and $\lambda_1 = 1064$ nm as in the differential absorption method. The 1064 nm laser radiation wavelength is out of the I_2 absorption band but it lies in the atmospheric transparency region [10]. The lidar receiving telescope square $A_2 = 0.011$ m², lidar constant $K_2 = 0.4$ at the 1064 nm wavelength [6] and iodine molecules absorption cross section as it has been derived from our experiments $\sigma_0 = 1.88 \cdot 10^{-18}$ cm². The laser radiation peak power values are equal to $P_L = 10$ and 100 kW and the two wavelengths' laser radiation peak power values ratio is equal to their avalanche photodiode spectral sensitivity values from [10] reverse ratio. The studied molecules concentration range is $10^{12} \dots 10^{16}$ cm⁻³ and the ranging distance was chosen from 1 up to 8 m. The atmospheric extinction coefficient values k_A were taken from [10] and for the two laser radiation wavelengths are equal to 0.17 and 0.135 km⁻¹ respectively.

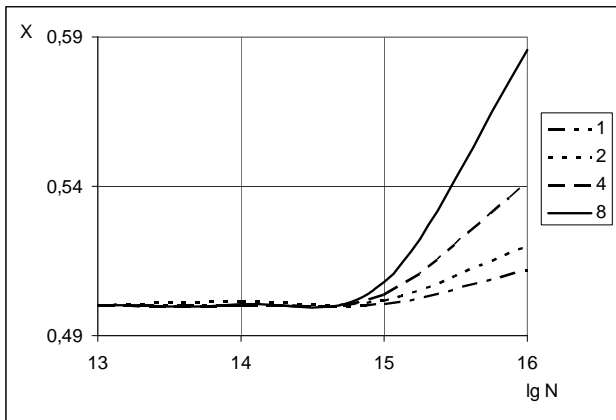


Fig.3. Plot of the DA coefficient X values vs. iodine molecules concentration N in 10^{15} cm⁻³ at the distance range 1 ... 8 m (pointed as 1, 2, 4 and 8 respectively).

DA-lidar signal powers ratio simulations by equation (3) were fulfilled using the above described parameters as a function of I_2 molecules concentration in the range from 10^{12} up to 10^{16} cm⁻³ and ranging distances from 1 to 8 m. These simulation results are exhibited in Fig.3. As results from Fig.3, the iodine molecules concentration in the range of $10^{12} \dots 10^{16}$ cm⁻³ can be sensed in all of the distance range, but concentration level of 10^{16} cm⁻³ at 8 m was exponential due to the high value of $\sigma_0 N$ as in the usual absorption spectroscopy [3, 8]. For further study would be of interest to compare the results of laser radiation power with the signal weakening as the radiation passes through the measuring volume and it does not overlap the signal recorded by DA photodetector in all of the distance ranges and molecules concentrations with exception of concentration level at $N_a = 10^{16}$ cm⁻³. All of the simulations

were fulfilled for laser radiation peak power of 10 kW. The laser radiation peak power value increasing up to 100 kW creates the sensing efficiency proportionally increased ratio. The X values differences in Fig.2 and Fig.3 are from our experimental accuracy.

5. CONCLUSION

Laboratory lidar studies and lidar equation simulation allowed us to get the iodine molecules absorption cross section at 532 nm wavelength $\sigma_0 = (1.88 \pm 0.37) \cdot 10^{-18}$ cm² in iodine molecules concentration range from $7.42 \cdot 10^{15}$ cm⁻³ to $32.4 \cdot 10^{15}$ cm⁻³ in satisfactory confidence with data [3] - $4.6 \cdot 10^{-18}$ cm² at 590 nm wavelength.

The iodine molecules absorption cross section at 532 nm wavelength was taken as the basis of the DA-lidar equation computer simulation. These simulation results corresponded with our DA-lidar experimental data.

REFERENCES

- [1] Privalov, V.E., Shemanin, V.G. (1999). Parameters of differential absorption lidar for detection of molecular iodine in atmosphere. *Rus. J. Optical Tech.* 66 (2), 40-42.
- [2] Privalov, V.E., Shemanin, V.G., Voronina, E.I. (2002). Lidar measurements of iodine molecules concentration. In *Proceedings of the SPIE*. Vol. 4900, 78-82.
- [3] Measures, R.M. (1987). *Laser Remote Sensing*. Moscow: Mir.
- [4] Vorob'eva, L.P., Evtushenko, G.S., Klimkin, V.M. et al. (1995). Cu-laser in problem of iodine radionuclides monitoring. *Rus. Atmosphere and Ocean Optics*, 8, 1648-1651.
- [5] Privalov, V.E., Shemanin, V.G. (1998). Lidars for control and measurements. In *Proceedings of the SPIE*. Vol. 3345, 6-10.
- [6] Privalov, V.E., Shemanin, V.G. (2000). Molecular iodine laser monitoring in the atmosphere. In *Proceedings of the SPIE*. Vol. 4316, 36-42.
- [7] Privalov, V.E., Shemanin, V.G., Voronina, E.I. (2001). Air quality controlling system for industrial region. In *Proceedings of the SPIE*. Vol. 4680, 112-128.
- [8] Zuyev, V.V., Kataev, M.Yu., Makogon, M.M., Mitzel, A.A. (1995). Differential absorption lidar method. Studies real status. *Rus. Atmosphere and Ocean Optics*, 8, 1136-1164.
- [9] Mironov, A.V., Privalov, V.E., Savel'ev, S.K. (1996). Iodine-127 absorption lines calculation according to cuprum vapor laser radiation lines. *Rus. Optics and Spectroscopy*, 80, 348-350.
- [10] Prokhorov, A. M. (Ed.) (1978). *Laser Handbook*. Vol. II. Moscow: Sov. Radio.

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