PARAMETERS OF A FLUORESCENT LIDAR FOR REMOTE SENSING OF ATMOSPHERIC MOLECULAR IODINE

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Received November 3, 1997

Based on the numerical solution of the lidar equation the feasibility for sounding of iodine isotopes in the atmosphere using the fluorescence method has been studied. As a fluorescence excitation source the Nd–*YAG laser second harmonic generation is taken; the variants of sensing by other laser sources are analyzed. The minimum detectable value of iodine concentration for remote sensing at distances from* 0.5 *to* 10 km *is* 10^{10} cm^{-3} *at peak power of a sounding pulse of* 10 MW.

The YAG–Nd laser radiation and especially its third and fourth harmonics are widely used in the remote fluorometer systems[1] that enables one to obtain the pulses of 10 ns duration with the energy from 1 to 100 mJ at a pulse repetition rate up to 50 Hz. The harmonic generation of such a laser may be used for remote sensing of molecular iodine in the atmosphere with a fluorescent lidar (FL).

The investigations of gas-discharge lasers stabilized by the iodine absorption[2–5] have made it possible to improve the data of the iodine atlas[6] and to give the supplementary data[7,8] for the iodine atlas containing hundreds of thousand lines.[9]

The iodine-vapor laser fluorescence has been studied earlier.[10,11]

The copper-vapor lasers may also have extensive applications to the fluorescence sensing of atmospheric molecular iodine.[12] The copper-vapor laser pulses of 10 ns duration with the pulse repetition rate up to 20 kHz may have the peak powers up to 130 kW.[13] Besides, the use of the accumulation regime of N pulses when recording a fluorescent signal makes it possible to have the sounding distance corresponding to an effective power and being equal to $PN^{1/2}$ (see Ref. 13).

For a comparison the results are presented for a XeCl excimer laser which is widely used in the FL sensing systems. It delivers the pulses of 10 ns duration and energy up to 0.1 J at the wavelength of 308 nm at a repetition frequency up to 50 Hz.[1]

In this connection, numerically solving lidar equation for the $^{127}I_2$ molecule and its isotopes is of a considerable interest when choosing among all the above-mentioned lasers a transmitter for an FL to provide for detection of iodine molecules.

The lidar equation for the iodine molecular fluorescence is of the form[1]

$$P(\lambda_1, R) = P_0(\lambda_0)\, K_1\, \Delta R A_2 \times$$
$$\times\, T(\lambda_1)\, T(\lambda_0)\, \frac{d\sigma_\rho}{d\Omega}\, N_a I R^{-2}, \qquad (1)$$

where $P(\lambda, R)$ is the FL signal power at a photoreceiver at a wavelength λ_1 coming from the distance R; $P_0(\lambda_0)$ is the laser power and its wavelength; K_1 is the lidar constant; ΔR is the distance step; A_2 is the receiving telescope area; $T(\lambda_0)$, $T(\lambda_1)$ are the atmospheric transmission at a laser radiation wavelength and at a FL signal wavelength; $(d\sigma_\rho/d\Omega)$ is the fluorescence differential cross-section of the molecule being studied; N_a is the molecular concentration; R is the distance to the volume sounded.

The wavelength of the fluorescence band maximum for I_2 molecules lies at 589.5 nm, the fluorescence differential cross-section at this wavelength equals $6.1\cdot10^{-22}$ cm^2/sr, the fluorescence quenching factor is 10^{-3} and the fluorescence quenching time is 1000 ns.[1]

Let us next remove from the constant K_1 the factor $\xi_\rho(\lambda)$ that describes the photocathode wavelength sensitivity by writing the former as

$$K_1 = K_2\, \xi_\rho(\lambda)\ . \qquad (2)$$

The remaining factors in Eq. (1) are the following: $\Delta R = 200$ m for the measurement time $\tau = 1000$ ns; $A_2 = 0.008$ m^2; $K_2 = 0.495$, as measured at the wavelength of 532 nm; the peak power of a laser pulse $P_0 = 10$ MW; the remote sensing distance $R = 0.5$; 1; 2; 5 and 10 km; the molecular concentration is 10^{16} cm^{-3}; the value of spectral sensitivity of FEU–79 photocathodes at the working wavelength of 589.5 nm was taken from Ref. 14 and it is given in the second column of Table I.

The atmospheric transmission is calculated as in Ref. 1 using the formula

$$T(\lambda, R) = \exp\left[-\int_0^R k(\lambda)\, dR\right] \qquad (3)$$

based on the values of the extinction coefficient k, taken from Ref. 15. The extinction coefficients at the

0235-6880/98/ 02-03 211-03 $02.00

corresponding wavelengths are presented in the second column of Table I.

TABLE I. The values of the extinction coefficient in the atmosphere; the photomultiplier relative spectral sensitivity and spectral brightness of solar background radiation calculated for laser wavelengths and the iodine molecular fluorescence band.

λ, nm	k, km^{-1}	ξ_ρ	S_b, 10^3 W/m^2 sr·nm
589.5	0.159	0.54	16.0
532	0.16	–	–
355	0.31	–	–
266	0.785	–	–
578	0.158	–	–
510	0.172	–	–
289	0.53	–	–
271	0.70	–	–
308	0.45	–	–

Given this value of the molecular concentration, selected wavelengths, and the pulse power of all lasers we have carried out numerical calculations, using the above data and Eq. (1), of the FL power in the sensing range from 0.5 to 10.0 km. The calculations were aimed at seeking an optimal version of the lidar system. The results of calculations for the I_2 molecule are given in Table II. From Table II it follows that with the distance increase the FL signal decreases by one order in the range of the first km and then it decreases by four orders of magnitude in the range of the next 9 km. Analysis of these results shows that the optimal version is the use of the YAG–Nd laser second harmonic with 532 nm wavelength and the copper-vapor laser radiation with the wavelengths at 510 and 578 nm. These lasers enable us to obtain the maximum FL power from iodine molecules in the range from 0.5 km to 10 km.

However, all these calculations were performed for the case of no background radiation present or for the case of night-time sensing. The sky background due to solar radiation essentially contributes to the optical power recorded with a FL lidar during day-time sensing. So, we have calculated the background power $P_b(\lambda, R)$ at a photoreceiver to estimate the effect of the background radiation on the lidar potentialities.

The spectral brightness values of solar radiation in different seasons, time of a day, and different meteorological conditions were taken from Ref. 16. In so doing we have chosen the most unfavorably conditions of lidar operation, namely, a bright sunny day. Using literature data[1,17,18] (because of the uncertainty in the direction of a telescope axis relative to the direction to the Sun) the background radiation brightness $S_b(\lambda)$ was determined. It is given in the last column of Table I. Using these values and the equation

$$P_b(\lambda, R) = S_b(\lambda)\, T(\lambda, R)\, K_2\, \xi_\rho(\lambda)\, A_2\, \Omega(R)\, \Delta\lambda\,, \qquad (4)$$

where $\Omega(R)$ is the solid angle of the lidar receiving telescope field-of-view, $\Delta\lambda$ is the spectral band width of the lidar optical receiver, similar to that in Ref. (18), the values of the background power $P_b(\lambda, R)$ for the signal being studied are calculated. According to Ref. 1 the minimum permissible signal-to-noise (S/N) ratio is considered to be equal to 1.5. Then we may determine the minimum power P detectable by a lidar as follows:

$$P_m = (S/N)\, P_b(\lambda, R)\ . \qquad (5)$$

TABLE II. Results of calculations of I_2 *FL power, for the wavelengths of all the lasers having* 10 MW *power, the molecular concentrations* 10^{16} cm^{-3}*, and the background power for the sensing distances from* 0.5 *to* 10 km.

R, km	0.5	1	2	5	10
			I_2		
λ, nm	P_m, μW	P_m, μW	P_m, μW	P_m, nW	P_m, nW
532	707.9	150.9	2.742	1685.0	84.71
355	656.8	129.9	2.031	795.8	18.89
268	517.9	82.4	0.785	74.03	0.16
578	708.2	151.1	2.748	1694.0	85.51
510	703.7	149.1	2.677	1578.0	75.11
289	588.3	104.2	1.309	265.1	2.10
271	540.4	87.9	1.304	113.3	0.38
308	612.3	112.9	1.535	395.3	4.66
			Background		
λ, nm	P_m, μW	P_m, nW	P_m, pW	P_m, pW	P_m, fW
589.5	1.517	350.2	74.68	7.417	837.2

Thus calculated results are presented in the last line of Table II. When comparing these results with the data from Table III we have drawn the conclusion that the largest excess of the FL power over the background one is obtained for the same wavelengths in the entire range of distances. In this case the molecular concentration from $2\cdot10^{10}$ to 10^{11} cm^{-3} can be detected using a laser power of 10 MW at the above wavelengths at any distance within this range, whereas for the same power at other wavelengths the value of the molecular concentration is from $2\cdot10^{10}$ to $5\cdot10^{13}$ cm^{-3}. In addition, we must keep in mind that the high repetition rate of a copper-vapor laser makes it possible the $N^{1/2}$ times increase in the FL power when accumulating N-pulse signal without the loss of space resolution. The subsequent enhancement of the lidar sensitivity can be obtained by increasing the receiving telescope diameter, but this may result in a sharp rise of the mass and dimensions of the lidar system. The use of corner-cube retroreflectors or topographic targets in the FL system could enable an increase in the power by a factor of 1.2–1.3 but will result in the loss spatial resolution[1] of FL measurements.

TABLE III. Results of calculations of minimum detectable I_2 *concentration using a lidar for all the above lasers having* 10 MW *power and the sensing distances from* 0.5 *to* 10 km.

R, km	0.5	1	2	5	10
λ, nm	N_a, cm^{-3}				
532	$2.14\cdot10^{10}$	$2.32\cdot10^{10}$	$2.72\cdot10^{10}$	$4.40\cdot10^{10}$	$9.88\cdot10^{10}$
355	$2.31\cdot10^{10}$	$2.70\cdot10^{10}$	$3.68\cdot10^{10}$	$9.32\cdot10^{10}$	$4.43\cdot10^{11}$
266	$2.93\cdot10^{10}$	$4.25\cdot10^{10}$	$9.51\cdot10^{10}$	$1.00\cdot10^{12}$	$5.12\cdot10^{13}$
578	$2.14\cdot10^{10}$	$2.32\cdot10^{10}$	$2.72\cdot10^{10}$	$4.38\cdot10^{10}$	$9.79\cdot10^{10}$
510	$2.16\cdot10^{10}$	$2.35\cdot10^{10}$	$2.79\cdot10^{10}$	$4.67\cdot10^{10}$	$1.12\cdot10^{11}$
289	$2.58\cdot10^{10}$	$3.36\cdot10^{10}$	$5.71\cdot10^{10}$	$2.80\cdot10^{11}$	$3.99\cdot10^{12}$
271	$2.81\cdot10^{10}$	$3.98\cdot10^{10}$	$5.73\cdot10^{10}$	$6.55\cdot10^{11}$	$2.22\cdot10^{13}$
308	$2.48\cdot10^{10}$	$3.10\cdot10^{10}$	$4.86\cdot10^{10}$	$1.88\cdot10^{11}$	$1.80\cdot10^{12}$

Thus the results obtained show that it is possible to choose an optimal laser radiation wavelength for remotely sensing the molecular iodine in the atmosphere at a given distance. In this case, even under most unfavorable background conditions, a laser wavelength can be selected that would enable detecting a molecular species at a desired level of concentration.

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