THE POSSIBILITY OF USING LIDARS FOR STUDYING ARTIFICIAL CLOUDS OF BARIUM AND BARIUM OXIDE IN THE UPPER ATMOSPHERE

D.V. Vlasov, O.K. Kostko, N.N. Kostko, and G.F. Tulinov

Soyuzgiprovodkhoz Institute, Moscow
Received July 26, 1989

The cross sections for resonant scattering of laser radiation by barium atoms and ions and barium oxide molecules are calculated for three wavelengths: 553.7, 455.4, *and* 535.1 nm. *The profiles of the transmission of laser radiation by the atmosphere at these wavelengths when sounding from the earth's surface are calculated. The integral ion times necessary for measuring the indicated components at altitudes of* 100–300 km *with fixed accuracy are determined for specific parameters of the receiving-transmit ting apparatus of the lidar.*

1. On of the main methods for studying the atmosphere is the method of artificial glowing clouds (AGCs). In this method an artificial cloud, consisting, as a rule, of alkali-metal vapors is ejected from a rocket at a given altitude (usually the altitudes range from 80 to 300 km). The experiment is conducted at dawn or dusk.

The atoms in the cloud are irradiated by rays of the rising or setting sun, they reemit resonantly, and the entire cloud glows against the background of the crepuscular sky. The artificial glowing cloud is photographed and photometric measurements are performed on it by ground-based optical means, and the temperature, the turbulent and molecular diffusion coefficients, the density, the wind in the atmosphere, and the relative concentration of some components at fixed altitudes are determined from the observations[1].

In the last few years AGCs consisting of barium vapors have been used in many experiments. Barium oxide and barium atoms and ions, emitting in the visible region of the spectrum, are formed in the atmosphere. The observation of the dynamics of the barium ions yields information about processes in the ionosphere, in particular, it makes it possible to measure the magnitude and direction of the electric fields.

One of the important drawbacks of the method of AGCs is that the experiment can be performed only at dawn or dusk, and this limits the observation time, for example, to 10–20 min at middle latitudes. In addition, the wind is measured by the method of triangulation, which requires organization of several observation points separated from one another by tens of kilometers. Such experiments are impossible in inaccessible and at the same time geophysically interesting regions, like, for example, the Arctic. Moreover, at high latitudes, owing to the specific nature of the solar irradiation, the experiments can be performed only during the spring or fall.

Lidar measurements make it possible largely to eliminate these drawbacks of the method of AGCs and sometimes to obtain additional information. We shall study in greater detail the possibility of such measurements.

2. For the further calculations it is necessary to know the cross section of resonance scattering (RS). The wavelength of a resonant transition to the ground state $^1S_0$ in the barium atom is equal to 553.7 nm and the wavelength for the transition of the barium ion into the state $^2S_{1/2}$ has the wavelength 455.4 nm. These wavelengths correspond to the main lines of the emission of Ba and Ba^+, since the oscillator strengths for these transitions are maximum (f is equal to 1.59 and 0.74, respectively).

At the altitude studied ($H \geq 150$ km) the Doppler half-width of the line is much greater than the natural half-width of the line and the width due to elastic collisions. At altitudes in the range 150–300 km ($T \sim 10^3$–$1.4 \cdot 10^3$ K) the values of $\Delta\nu_d$ for 553.7 nm and 455.4 nm are equal to 10^9 Hz ($\Delta\lambda_d = 1.1 \cdot 10^{-3}$ nm) and $1.3 \cdot 10^9$ Hz ($\Delta\lambda_d = 8.8 \cdot 10^{-4}$ nm for 10^3 K), where $\Delta\lambda_d$ is the Doppler broadening of λ_0. There are no fundamental difficulties in obtaining laser radiation in the indicated region of the spectrum with half-width $\Delta\nu_1 \simeq 10^9$ Hz. For $\Delta\nu_1 = \Delta\nu_d$ it may be assumed that the cross section for resonance scattering is determined by the well-known formula

$$\sigma^0_{RS} = \frac{\sqrt{\pi}\, e^2}{m_e c} \cdot \frac{f\lambda_0\sqrt{M}}{\sqrt{2RT}} \cdot \exp(-\ln 2) = 2.89 \cdot 10^{-14} f\lambda_0 \sqrt{\frac{M}{T}}. \quad (1)$$

In Eq. (1) σ^0_{RS} has the dimension cm^2, if λ is expressed in cm, and $R = 8.31 \cdot 107$ ergs/mole · K.

0235-6880/97/03 271-06 $02.00

Assuming that the resonance scattering is isotropic $\left(\sigma^0_{\pi RS} = \frac{\sigma^0_{RS}}{4\pi}\right)$ for the maximum cross section for resonance backscattering we obtain

$$\sigma^0_{\pi RS}(553.7\ \text{nm}) = 7.1\cdot10^{-13}\ \text{cm}^2\cdot\text{cp}^{-1};$$

$$\sigma^0_{\pi RS}(455.4\ \text{nm}) = 2.7\cdot10^{-13}\ \text{cm}^2\cdot\text{cp}^{-1}$$

at $T = 10^3$ K.

We shall now calculate the cross section for resonance scattering for the BAO molecule. For this we shall study the spectral transition to the ground state $A^1\Sigma \to X^1\Sigma$. The projection of the orbital angular momentum of the electrons on the axis of the molecule Λ for the state Σ is equal to zero. The degeneracy is equal to one and therefore the quantum number $S = 1$ (since $2S + 1 = 1$). In the transition $\Sigma \to \Sigma$ $\Delta J = 0$ is forbidden, i.e., there is no Q branch, and since the transition is a singlet transition, it has two branches P_1 and R_1.

Values of the population of the lower level for vibrational quantum numbers $v'' = 0$–0.978, $v'' = 1$–0.021, $v'' = 2$–$5\cdot10^{-3}$ at T = 220 K are presented in Ref. 2. As the temperature of the atmosphere increases as the altitude increases the population of the vibrational levels with $v'' = 1$, 2, etc. increases. Estimates nonetheless show that for $T \simeq 10^3$ K the population of the level with $v'' = 0$ remains highest. For this reason, for all of the subsequent calculations we shall use transitions from the lower level $X'\Sigma(\ v'' = 0)$.

The most intense emission is the emission of BaO owing to the transitions $v' = 3$, $v'' = 0$ and $v' = 4$, $v'' = 0$. This emission is observed when BaO is ejected at an altitude of 107 km[2]. The Frank-Condon factor is maximum for the transition (4, 0): $q_{4,0} = 0.1616$ (Ref. 3) (correspondingly the oscillator strength connected with the Frank-Condon factor is also maximum: $f_{4,0} = 5.72\cdot10$–3 (Ref. 4)). We shall calculate for the transition (4, 0) the frequencies of the transition lines corresponding to different rotational quantum numbers J.

The main spectroscopic constants of the transitions are: $v_{0,0} = 16722.3\ \text{cm}^{-1}$, the rotational constant $B_{v''=0} = 0.3119$, $B_{v'=4} = 0.2534$, the frequencies between the levels $v' = 0$ and $v' = 1$ correspond to 496.4 cm^{-1}; $v' = 1$, $v' = 2$–$493.1\ \text{cm}^{-1}$; $v' = 2$, $v' = 3$–$489.8\ \text{cm}^{-1}$; $v' = 3$, $v' = 4$–$486.6\ \text{cm}^{-1}$.[3] Therefore $\nu_{4,0} = 18688.2\ \text{cm}^{-1}$ ($\lambda_{4,0} = 535.1$ nm).

The transition frequencies for the P_1 and R_1 branches are calculated according to the following formulas:

$$\nu = \nu_{4,0} + F'(J-1) - F''(J) \text{ —— } P_1\text{-branch};$$

$$\nu = \nu_{4,0} + F'(J+1) - F''(J) \text{ —— } R_1\text{-branch}; \quad (2)$$

$$F'(J) = 0.2534\ J\ (J+1);$$

$$F''(J) = 0.3119\ J\ (J+1),$$

where $F'(J)$ and $F''(J)$ are the terms of the upper and lower states, respectively. The results of the calculations performed are presented in Fig. 1.

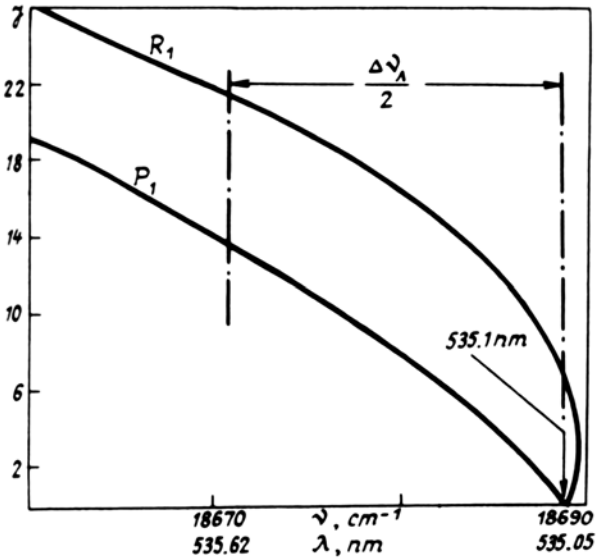

FIG. 1. The computed transition frequencies for the P_1 and R_1 branches of the BaO molecule ($A^1\Sigma$–$X^1\Sigma$, $v' = 4$, $v''= 0$).

The cross section for resonance backscattering for the chosen transition of the BaO molecule can be written in the form

$$\sigma^0_{\pi RS} = \frac{1}{4\pi}\frac{\pi e^2}{m_e c} f_{4,0}\frac{A_{4,0}}{\sum\limits_{v''} A_{4,v''}}\frac{\theta}{\Delta\nu_L}, \quad (3)$$

where $A_{v',v''}$ is the Einstein coefficient for spontaneous emission and Θ is found as follows:[5]

$$\theta = \frac{1}{4\sum\limits_{J''=0}^{\infty}(2J''+1)\cdot e^{-F''_1(J'')\frac{hc}{kT}}} \times$$

$$\times\left[\sum_{J''=0}^{21}(J''+2)\cdot e^{-F''_1(J'')\frac{hc}{kT}} + \sum_{J''=0}^{13}(J''-1)\cdot e^{-F''_1(J'')\frac{hc}{kT}}\right]. \quad (4)$$

The summation on the P_1 and R_1 branches is performed in Eq. (4) over the quantum numbers for

which the transition frequencies fall into the spectral width of the propagating radiation.

For the calculations we chose the spectrum width of the organic-dye laser (ODL) $\Delta\lambda_1 = 1$ nm. For this spectral width the energy of the ODL is of the order of $E_0 = 1$ J less than the value of $\Delta\lambda_1$ obtained using several Fabry-Perot interferometers, etc., and leads to a sharp drop in the radiation energy. As one can see from Fig. 1, for this value of $\Delta\lambda_1$ in Eq. (4) it is necessary to sum up to $j'' = 21$ for the R_1 branch and $j'' = 13$ for the P_1 branch. At $T = 10^3$ K the calculation gives the value $\Theta \simeq 4 \cdot 10^{-2}$.

We shall now study the ratio $\dfrac{A_{4,0}}{\sum\limits_{v''} A_{4,v''}}$. The numerator is given by the expression[4]

$$A_{4,0} = \frac{64\pi^4}{3h} \lambda_{4,0}^{-3} R_{e,4}^2(\bar{r}) q_{4,0}, \qquad (5)$$

where the matrix element of the dipole moment $R_{e,4}^2(r)$ ($v' = 4$) is equal to $1.98 \cdot 10^{-18} \mathrm{g}^{1/2}\mathrm{cm}^{S/2} \cdot \mathrm{c}^{-1}$. The lifetime of the excited state τ_v, for $v' = 4$ is equal to $2.75 \cdot 10^{-7}$ sec.[4]

Using the relation $\sum\limits_{v''} A_{v'v''} = \dfrac{1}{\tau_{v'}}$ and the formula (5) we obtain

$$A_{v'v''} \Big/ \overline{\sum_{v''} A_{v'v''}} = 0.36.$$

Finally, we obtain for the transition (4, 0),

$$\sigma^0_{\pi RS} \simeq 1.6 \cdot 10^{-19} \ \mathrm{cm}^2 \cdot \mathrm{sr}^{-1}.$$

We note that in the range of altitudes studied the quenching of the radiation by collisions is insignificant, since the collision frequency $\nu_{col} \ll A_{4,0}$. (At $H = 150$ km, $\nu \simeq 2 \cdot 10^1\ \mathrm{c}^{-1}$, $A_{4,0} = 3.6 \cdot 10^6\ \mathrm{c}^{-1}$).

TABLE I.

Parameters σ^0_M, $\sigma^0_{\pi M}$ and $\sigma^0_{O_3}$ used in calculations.

λ , nm	455.4(Ba$^+$)	535.1(BaO)	553.7(Ba)
σ^0_M, cm^2	$1.14 \cdot 10^{-26}$	$5.72 \cdot 10^{-27}$	$4.98 \cdot 10^{-27}$
$\sigma^0_{\pi M}$, cm·sr^{-1}	$1.37 \cdot 10^{-27}$	$6.87 \cdot 10^{-28}$	$5.99 \cdot 10^{-28}$
$\sigma^0_{O_3}$, cm^2	$1.7 \cdot 10^{-22}$	$2.2 \cdot 10^{-21}$	$2.7 \cdot 10^{-21}$

3. For the further calculations it is necessary to determine the transmission of the atmosphere on the sounding path for the chosen wavelengths. Table I gives the values employed in the calculations for the cross sections of the molecular scattering and absorption by ozone.

In the calculations the values of the transmission

$$q \simeq \exp\left\{-2 \int_0^H \left[\sigma_A(h) + \sigma^0_M \rho(h) + \sigma^0_{O_3} N_{O_3}(h)\right] dh\right\} \qquad (6)$$

the aerosol models of Ref. 6 (σ_A is the aerosol scattering coefficient) were employed and the middle-latitude data of Ref. 7 were employed for the number density of the atmosphere ρ and the ozone concentration N_{O_3}. . The results of the calculations of q show that the main attenuation occurs up to the altitude 30 km. The computed values of q are equal to 0.17 at 455.4 nm, 0.28 at 535.1 nm, and 0.3 at 553.7 nm.

4. For the further calculations it is necessary to evaluate the background radiation for the chosen wavelengths. Using the well-known expression

$$\bar{N}_b(\lambda) = B(\lambda) S \Omega \Delta\lambda \qquad (7)$$

and the data of Ref. 8 on the airglow of the night sky, we obtain the values of $\bar{N}_b$. As the calculation showed, these values vary from 30 to 40 photoelectrons per second for the wavelength studied. In the formula (7) $B(\lambda)$ is the brightness of the night sky, S is the area of the receiving optical antenna, and $\Omega = \pi\gamma^2/4$, where γ is the angle of the field of view of the antenna, and $\Delta\lambda$ is the spectral half-width of the interference filter. In the calculations the following values were employed: $S = 7.85 \cdot 10^3\ \mathrm{cm}^3$ ($D = 1$ m), $\gamma = 0.5$ mrad, $\Delta\lambda = 0.5$ nm, and the quantum efficiency of the photomultiplier $\eta = 0.1$.

5. Usually, from the approximately 10 kg of barium ejected from the rocket about 0.2 kg of barium vapor is formed. According to data from ground-based measurements, by the tenth second after ejection the diameter of the AGC is equal to about 10 km, and by the 200th second it increases up to 30 km. Assuming that the vapor-like substance expands in all directions uniformly, it is possible to calculate the concentration of the components after ejection from the rocket. The calculations are presented in Table II.

TABLE II.

The concentration of the gaseous components (in cm^{-3}) and the backscattering signals (in numbers of photoelectrons per sec).

		Ba	Ba$^+$	BaO
t, s	10	$1.7 \cdot 10^6$	$1.7 \cdot 10^6$	$1.6 \cdot 10^6$
	100	$2.2 \cdot 10^5$	$2.2 \cdot 10^5$	$2.0 \cdot 10^5$
	200	$6.2 \cdot 10^4$	$6.2 \cdot 10^4$	$5.6 \cdot 10^4$
$\bar{N}_c$, km	100	$1.7 \cdot 10^7$	$2.9 \cdot 10^6$	1.2
	200	$4.1 \cdot 10^6$	$7.0 \cdot 10^5$	$3.8 \cdot 10^{-1}$
	300	$1.9 \cdot 10^6$	$3.2 \cdot 10^5$	$1.6 \cdot 10^{-1}$

In the calculation the following quantities were used: the concentration of the components was equal to 10^3 cm^{-3}, $\Delta H = 1$ km, the energy of the radiation of the ODL was equal to 500 mJ per pulse, the total transmittance of the optical systems was equal to 0.2, the pulse repetition frequency of the ODL $f_1 = 0.1$ Hz, $S = 7.85 \cdot 10^3$ cm^2, and $\eta = 0.1$. The small magnitudes of the backscattering signals (the second part of Table II) for barium oxide are explained by the insignificant resonant scattering cross section for this molecule compared with Ba and Ba^+.

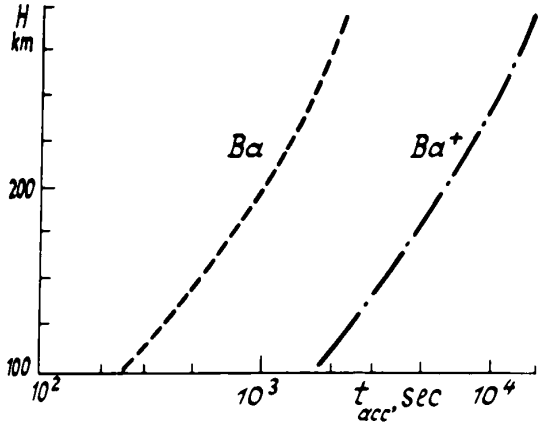

FIG. 2. The accumulation times necessary for determining the backscattering signals with an accuracy of 1%. The parameters of the lidar are presented in the text.

For the parameters presented above the accumulation times t_{acc}, necessary for recording the signal with a given accuracy δ (in %) was also calculated:

$$t_{acc} = \left(\frac{100}{\delta}\right)^2 \cdot \frac{\bar{N}_c + 2\bar{N}_n}{\bar{N}_c^2} \cdot \frac{2}{t(\Delta H) f_L} \qquad (8)$$

where N_n is the noise signal (the background radiation, dark current), $t(\Delta H)$ is the gating time, assumed in the calculations to be equal to 6.67 μs ($\Delta H = 1$ km). The computed values of t_{acc} for $\delta = 1\%$ are presented in Fig. 2. The calculations also show that it is impossible to measure BaO with a ground-based lidar. Even for BaO concentrations of 10^6 cm^{-3}, the radiation energy of the ODL 1 J and $\delta = 10\%$ the accumulation time of the signal from an altitude of 100 km with $\Delta H = 5$ km is equal to about 3 h.

The barium-vapor cloud expands and it is necessary to study the dynamic picture of the changes in the backscattering signal. In sounding the expanding cloud the backscattering signal is distorted by the Poisson statistics of the photocounts. To analyze the real situation of the expansion of the barium cloud we developed a numerical model that includes the altitude profiles of the molecular and aerosol scattering and absorption by ozone. The calculations were performed on an IBM PC-AT personal computer for the lidar parameters presented above. Figure 3 illustrates selected data from the calculations for three times after ejection of barium at an altitude of 150 km, 0.2 km of which is vaporized. The backscattering signal is so strong that Poisson statistics introduces distortions into the scattering signal and the barium concentration profile being determined only starting with the 60th second after ejection.

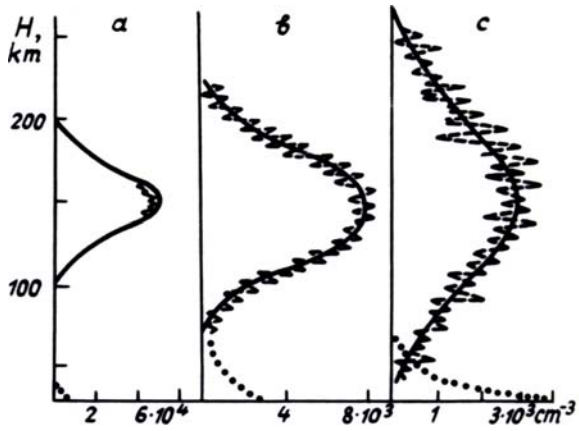

FIG. 3. The distortion of the barium concentration profile owing to the Poisson statistics of the photocounts. a) 60 sec, *b)* 110 sec, *and c)* 150 sec *after ejection. The solid curve is the barium distribution and the broken curve shows the measurements, distorted by the Poisson statistics of the photocounts, and the dots represent the molecular scattering.*

Finally, we shall make some remarks regarding the accuracy of the determination of the concentration of barium atoms and ions. To determine the product of the instrumental factor of the lidar by the atmospheric transmission K^*q it is necessary to use the method of calibration according to the layer of the atmosphere where the scattering is assumed to be molecular scattering ($H > 30$ km). In this case, using, for example, the atmospheric data, K^*q can be determined with an error of several percent. Then the main error in determining the concentration will be determined by the accuracy with which the signal is measured.

Thus the calculations performed show that ground-based nighttime lidar measurements of clouds of barium and barium ions in the upper atmosphere are extremely efficient. Available organic-dye lasers already permit building lidars for observing these components.

REFERENCES

1. M. McIven and L. Phillips, *Atmospheric Chemistry* [Russian translation] Mir, Moscow (1978).
2. G.T. Best and H.S. Hoffman, J. Quant Spectrosc. Radiat. Transfer **13**, 69–78 (1973).
3. T. Wentink and R. Spindler, J. Quant. Spectrosc. Radiat. Transfer **12**, 129–148 (1972).
4. S.E. Johnson, J. Chemical Phys. **56**, No. 1, 149–154 (1972).

5. O.K. Kostko and E.A. Chayanova, Tr. Tsentr. Aerol. Obs. No. 99, 81–89 (1971).
6. V.E. Zuev and G.M. Krekov *Optical Models of the Atmosphere*, Gidrometeoizdat, Leningrad (1986).
7. I.I. Ippolitov, V.S. Komarov, and A.A. Mitsel', in: *Methods for Sounding the Atmosphere* [in Russian], Nauka, Novosibirsk (1985).
8. A.L. Broadfoot and K.R. Kendall, J. Geoph. Res **73**, No. 1, 426–428 (1968).