SOME POTENTIALITIES OF USING NONLINEAR SPECTROSCOPIC EFFECTS FOR SOUNDING ATMOSPHERIC AEROSOL

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The peculiarities of nonlinear spectroscopic effects use for sensing optical parameters of the atmospheric aerosol are investigated taking the effect of saturation of resonance absorption as an example. The direct problem was simulated by lidar sounding equations and the equation of a plane wave propagation, and by calculating nonlinear interaction with a resonant medium component and the linear one with a nonresonant component. The inverse problem is reduced to a minimization. Use of various nonlinear spectroscopic effects is studied. The potentialities of their use are analyzed with the corresponding goal (minimized) function.

1. INTRODUCTION

In connection with the problems of climate change forecast and climate formation, as well as influence of industrial plants on the environment, it is necessary to analyze the state and dynamics of various atmospheric processes and, particularly, to have information about the spatial distribution of aerosol.

Aerosol plays an important role in processes of radiation field formation in the atmosphere, by taking part in various physical conversions including the conversions connected with the atmospheric pollution by products of industrial activities.

A complicated spatial structure makes it difficult to obtain the data about optical characteristics of an aerosol field by usual contact methods. So it is expedient to apply laser sensing methods for the investigation of their distribution.

Remote methods of determining aerosol parameters are based on the phenomenon of laser radiation backscattering. The interpretation of return signals from the atmosphere is based on the lidar equation:

$$P(z) = I_0 \frac{c\tau}{2} \beta_\pi(z) A z^{-2} \xi \exp[-2\int_0^z \beta_{\rm ex}(z')\mathrm{d}z'], \qquad (1)$$

where $P(z)$ is the signal power at the receiver; I_0 is the sounding pulse power; c is the speed of light; τ is the pulse duration; A is the receiving area; ξ is the receiving system efficiency; $\beta_\pi(z)$, $\beta_{\rm ex}(z)$ are the volume coefficients of backscattering and extinction in the medium.

The right-hand side of the equation (1) contains at least two unknown aerosol parameters, so one should redefine the problem in order to solve the equation with respect to one of them. Different ways of solving Eq. (1) exist depending on the *a priori* information available.[1]

For instance, there exists multifrequency sounding method based on the solution of the inverse problem of laser sounding and inversion of the results. In this case the redefinition can be done on the basis of *a priori* information about the form of the solution. This method is expedient for being applied if the bulk of measurement information is highly restricted (sounding at two or three wavelengths) and the investigated object refers to the class of well studied aerosols (see, for instance, Ref. 2).

Complications connected with the choice of wavelengths at which one should perform the measurements arise in practice of the multifrequency sounding.[3]

The method of Raman scattering is also used for determining aerosol optical parameters. The method is based on the analysis of the laser pulse radiation scattered by the atmosphere. The determination of these values is performed from aerosol and Raman lidar returns from N_2 and O_2 molecules (see, for instance, Ref. 4). In spite of the principal simplicity of the method, its instrumental realization meets some difficulties. First of all, these are due to small effective cross sections of the backscattering.

2. THEORETICAL BACKGROUND FOR APPLYING NONLINEAR SPECTROSCOPIC EFFECTS TO SOUNDING ATMOSPHERIC AEROSOL

Analysis of known sounding methods based on linear interaction of the radiation with a medium shows that either the use of large volumes of *a priori* information about optical parameters of the aerosol component of a medium is inherent in the methods, or they are complicated in technical realization.

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Below it is shown that the volume of *a priori* information can be reduced if beyond the limits of the linear interactions.

Optical characteristics of the atmosphere under conditions of nonlinear interaction of radiation with a medium depend on the incident radiation intensity. The possibility of solving the inverse problem is determined both by the character of local variations in optical characteristics under the field action and by transformation of laws of optical radiation evolution in the medium under the nonlinear interaction.

The solution of the direct sounding problem in the nonlinear case can be presented in the form

$$P(z) \sim \beta_\pi(z, I(z))\; W(z)\; \exp\left[-\int\limits_0^z \beta_{ex}(z')\mathrm{d}z'\right]; \qquad (2)$$

$$F(I(0), I(z), \beta_{ex}(z)) = 0, \qquad (3)$$

where the coefficients β_π and β_{ex} are determined by all components of the atmosphere; $W(z)$ and $I(z)$ are radiant energy and intensity of the light propagated the path z in the medium; Eq. (3) describes the law of optical radiation propagation in the medium.

A separate measurement of the backscattering coefficients of different components of the atmosphere is possible if, for example, the scattering cross sections vary in a different way.

The effect of saturation of resonance absorption which, for example, takes place for the atmospheric CO_2 in the range near λ=10.6 μm at the intensity $I_s \sim$ 0.2–0.5 MW/cm^2 (Ref. 5) is the most low-threshold nonlinear effect in the atmosphere.

Another effect capable to change the absorption in the central portion of the line is the change of the intermolecular interaction potential under the action of radiation. The threshold of the effect is $I_p \sim$ 2–3 MW/cm^2 (Ref. 6) what is approximately the tenfold value of the absorption saturation intensity for the atmospheric CO_2 in this spectral range.

The laser radiant energy absorbed in the medium and transformed into the heat kinetic energy also causes a nonlinear change of the atmospheric optical characteristics. The appearance of a “laser spark,B propagation of shock waves and ionization fronts, screening of the laser beam due to strong absorption and nonlinear light scattering in thus formed plasma accompany this phenomenon. This effect imposes restrictions on limiting radiant energy which can be transferred through the atmosphere. Experimental data on the influence of pulsed radiation of a ruby laser on the water aerosol demonstrate that the optical breakdown arising at average radiant intensity $I_b \sim 10^2$ MW/cm^2 (Ref. 6) (what also exceeds the saturation intensity I_s) plays a certain role in the mechanism of drop explosion.

A two-level model of a resonant-absorbing gas particle is used as a physical model for describing the structure of molecular energy levels.

The calculations have been performed under the plane wave approximation. In this case, one should take into account the diffraction effects. It is known that the beam width increases by the law $a(z) \sim \sqrt{1+D_0^2}$, where $D_0 = 2z/ka^2$ is the dimensionless diffraction length. For typical parameters of the problem we have z=10 km, λ=1 μm, $a(0)$ = 10 cm $\Rightarrow D_{02} \cong 0.3$. Since $D_0 \ll 1$, the diffraction can be neglected. The use of the plane wave model causes also the neglection of nonlinear refraction (inhomogenity of the refractive index).

We also restrict ourselves by nonlinear interaction of radiation with a resonant component of the medium, i.e., with the gas, and by linear interaction with the aerosol.

The propagation equation (3) in the case of stationary interaction can be written in a well-known form

$$\mathrm{d}I/\mathrm{d}z = -\beta_{ex}^{m} I/(1 + I/I_s) - \beta_{ex}^{n}, \qquad (4)$$

where β_{ex}^{m} and β_{ex}^{n} are volume coefficients of resonance absorption and those of various resonant losses; I_s is the saturation intensity.

Solution to the problem (4) was obtained analytically as an implicit function which can be represented explicitly by the iteration bisection method.[7] The algorithm of the bisection method is realized in the program.

The analysis demonstrates that the inverse problem is solvable if one transmits three sounding pulses with different powers to the medium, e.g., the conditions of linear, weakly nonlinear, and strongly nonlinear interactions can be chosen as conditions of the measurements.

In this case the scattering signals are as follows:

$$P_1(\lambda_1) = \beta_\pi(z) W_1(0)\exp[-2(\tau^m + \tau^n)], \qquad (5=)$$

$$P_2(\lambda_2) = \beta_\pi(z) W_2(z, \tau^m, \tau^n, I_s)\exp[-(\tau^m + \tau^n)], \qquad (5b)$$

$$P_3(\lambda_1) = \beta_\pi(z) W_3(z, \tau^m, \tau^n, I_s)\exp[-(\tau^m + \tau^n)], \qquad (5c)$$

where $W_{2,3}$ is the energy of powerful laser pulses propagated the path z in the medium which is calculated from the corresponding propagation problem with allowance for nonlinear interaction (4); τ^m and τ^n are optical thicknesses of the resonant and nonresonant medium components; P_j are the measured backscattering signals.

The functional relations for unknown optical parameters are determined under the assumption that the scattering mainly occurs by aerosol particles (neglecting the Rayleigh scattering: $\beta_{ex}^{m}(z) \ll \beta_{ex}^{n}(z)$). As a rule, this assumption is valid in the visible and IR ranges.

By combination of equations (5), the solution of the inverse problem can be reduced to the problem of free minimization of a positively definite goal function depending on the radiation parameters and on the ratio of the received scattering signals to unknown optical parameters τ_x^m and τ_x^n of the medium. The information about the values of τ^m, and τ^n is in scattering signals. If the statement of the minimization problem is correct, the minimum of the goal function is obtained at $\tau_x^m = \tau^m$, $\tau_x^n = \tau^n$.

The parameter β_π is obtained from a separate relation similar to Eq. (5a) and can be found once the values τ^m and τ^n are obtained.

3. RESULTS

The results of calculations of the above–mentioned goal function for a homogeneous layer of the medium are presented below. The values τ^m and τ^n were the initial ones in the general case.

The calculations were performed with allowance for errors of two types: additive (caused by background disturbances and internal noises) and multiplicative (*a priori* information errors). The calculations show that the goal function has many extrema; nevertheless, the global minimum coincides with the parameters sought.

The described method can be applied to both single-frequency and double-frequency sounding.

In the case of single-frequency sounding $\lambda_1=\lambda_2$, the wavelength must, on the one hand, fall within the region of resonant absorption of the medium and, on the other hand, it must be in the region of efficient interaction of radiation with the aerosol under study.

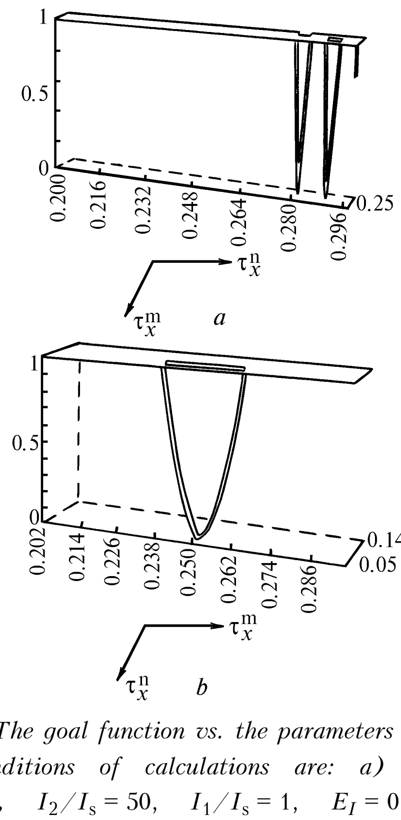

FIG. 1. The goal function vs. the parameters τ_x^m and τ_x^n. *The conditions of calculations are: a)* $\tau^m = 0.26$, $\tau^n = 0.29$, $I_2/I_s = 50$, $I_1/I_s = 1$, $E_I = 0$, $E_a = 0$; *b)* $\tau^m = 0.08$, $\tau^n = 0.25$, $I_2/I_s = 50$, $I_1/I_s = 10$, $E_I = 0$, $E_a = 0$; I/I_s *is the initial power of a sounding pulse normalized to* I_s, E_a *is the value of the additive error,* E_I *is the error of the assignment of the medium saturation intensity.*

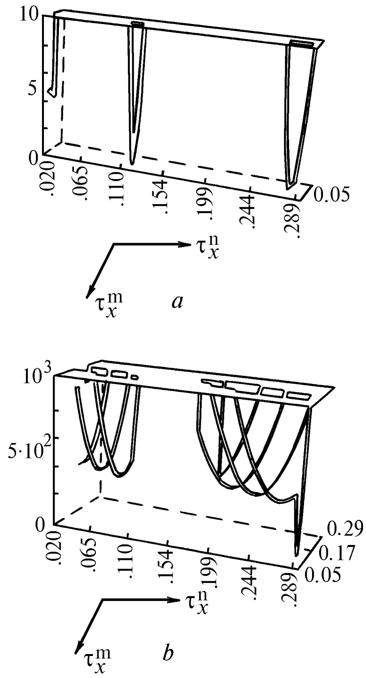

FIG. 2. The goal function vs. the parameters τ_x^m and τ_x^n. *The conditions of calculation are: a)* $\tau^m = 0.14$, $\tau^n = 0.283$, $E_a = 0.01$, $E_I = 0$, $I_2/I_s = 50$, $I_1/I_s = 10$; *b)* $\tau^m = 0.14$, $\tau^n = 0.283$, $E_a = 0.05$, $E_I = 0$, $I_2/I_s = 50$, $I_1/I_s = 10$.

Figure 1 shows an example of the calculation of the function minimized at $\lambda_1=\lambda_2$ without allowance for errors. It is seen that the value of the error in medium parameter reconstruction weakly depends on intensity if only $I_{1,2}$ are greater than I_s and $I_2/I_1>2$. The influence of the additive error E_a is illustrated by Fig. 2.

The effect of saturation of resonance absorption can also be used in double–frequency laser sounding of the atmospheric aerosol as a modification of the differential absorption method. In this case the third (powerful) pulse is transmitted at the wavelength λ_1 in addition to sounding at the wavelengths λ_1 and λ_2 under the conditions of linear interaction like in the method of differential absorption. The scattering signals of the first and second pulses give the information about the optical thickness of the gas component τ^m, and the third one makes it possible to determine the aerosol parameters β_π^n and τ^n. In this case the goal function depends only on the value τ^n.

In order to implement the method, the wavelength λ_1 must fall within the line contour of the resonance absorption of the medium, and the other wavelength λ_2 must fall within the atmospheric transmission microwindow neighboring to the line.

Figures 3–5 show the results of the corresponding goal function calculation for different initial parameters of the problem.

Comparing Fig. 3*a* and 3*b* one can see that the depth of lateral minima decreases with the increase of optical thickness τ^m what must lead to a greater stability of the method with respect to the additive error. The influence of the error is shown in Fig. 4. Note that the displacement of the global minimum here is on the order of this error.

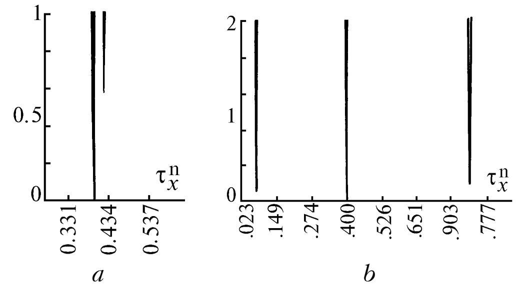

FIG. 3. The goal function vs. the parameters τ_x^m and τ_x^n. *The conditions of calculations are: a)* $\tau^m = 0.26$, $\tau^n = 0.41$, $E_a = 0$, $E_I = 0$, $I_2/I_s = 50$, $I_1/I_s = 10$; *b)* $\tau^m = 0.08$, $\tau^n = 0.41$, $E_a = 0$ $E_I = 0$, $I_2/I_s = 50$, $I_1/I_s = 10$.

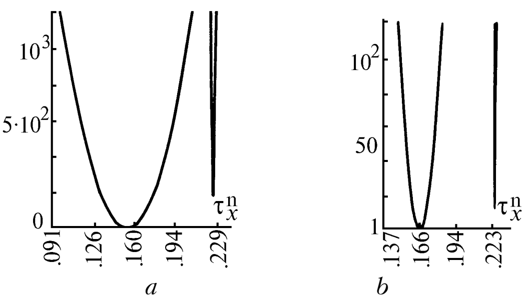

FIG. 4. The goal function vs. the parameters τ_x^m and τ_x^n. *The conditions of calculation are: a)* $\tau^m = 0.35$, $\tau^n = 0.15$, $E_a = 0.002$, $E_I = 0$, $I_2/I_s = 50$, $I_1/I_s = 10$; *b)* $\tau^m = 0.35$, $\tau^n = 0.15$, $E_a = 0.01$, $E_I = 0$, $I_2/I_s = 50$, $I_1/I_s = 10$.

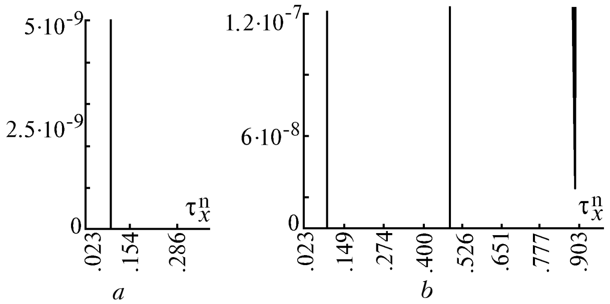

FIG. 5. The goal function vs. the parameters τ_x^m and τ_x^n. *The conditions of calculations are: a)* $\tau^m = 0.41$, $\tau^n = 0.1$, $E_I = 0$, $E_a = 0$, $I_2/I_s = 50$, $I_1/I_s = 10$; *b)* $\tau^m = 0.41$, $\tau^n = 0.1$, $E_I = 0.01$, $E_a = 0$, $I_2/I_s = 50$, $I_1/I_s = 10$.

The influence of the accuracy of *a priori* information (the accuracy of the assignment of I_s) on the behavior of the goal function is illustrated by Fig. 5. One can conclude that the error of the initial parameter reconstruction weakly depends on the accuracy of the assignment of the saturation intensity value.

4. CONCLUSION

The single-frequency sounding scheme and the modification of the differential absorption method are considered as applied to the use of nonlinear spectroscopic effects in sounding optical parameters of the atmospheric aerosol.

The estimates are obtained for a model of a plane wave in approximation of a stationary nonlinear interaction with the resonant (gas) component of the medium and the linear interaction with the aerosol one. The nonlinearity of the interaction was taken to be due to the effect of saturation of resonance absorption.

The calculations have shown that nonlinear spectroscopic effects can be used for separately determining the backscattering coefficient and the extinction coefficient due to atmospheric aerosol.

It should be noted that there exists a problem of fitness of the direct problem model used to the real situation. But this problem is common for all the methods of the optical sounding.

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