USE OF NONLINEAR SPECTROSCOPIC EFFECTS TO MEASURE OPTICAL PARAMETERS OF THE ATMOSPHERIC AEROSOL

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We present here some results of the numerical simulations of the methods of single- and double-frequency sounding of atmospheric aerosol optical parameters using a nonlinear spectroscopic effect of saturation of the resonance absorption. We consider also the influence of interferences of the channel of propagation and errors in spectroscopic data on the accuracy of reconstruction of the above-mentioned parameters.

1. INTRODUCTION

When determining the atmospheric aerosol optical parameters a problem arises on separating the backscattering and extinction coefficients.

Inversion of sounding data aimed at obtaining aerosol microphysical parameters is possible at multifrequency sounding. However, it is worth using this method if the bulk of measurement data is limited (the sounding is performed at two or three wavelengths, and an object being studied refers to the class of a well studied aerosol, since the information on the type of solution is used as *a priori* information.[1] When sounding at one or two wavelengths *a priori* information about the lidar ratio is often used. In practice of the multifrequency sounding, the complicated questions arise on selection of wavelengths at which the measurements should be made so that they are informative relative to the aerosol under study.[1]

In Ref. 2 we described a possibility of measuring separately aerosol optical parameters using nonlinear effects. The purpose of this paper is to simulate single- and double-frequency sounding of the aerosol optical parameters under conditions of saturation of resonance absorption by the molecular component of the medium. Selection of a nonlinear effect is caused by its lower threshold compared to other effects under atmospheric conditions.[3,4]

2. STATEMENT OF THE PROBLEM

To describe the structure of energy levels of a molecule we used a two-level model of a resonance-absorbing gas particle.

In the case of a nonlinear stationary interaction of radiation with the resonance component of the medium, i.e. with a gas, as well as in the case of a linear interaction of radiation with aerosol, the equation of propagation for a plane wave is written in the known form:

$$\mathrm{d}I/\mathrm{d}z = -\beta_{\mathrm{ex}}^{\mathrm{m}}\, I/(1 + I/I_{\mathrm{s}}) - \beta_{\mathrm{ex}}^{\mathrm{n}}\, I\,, \tag{1}$$

where $\beta_{\mathrm{ex}}^{\mathrm{m}}$ and $\beta_{\mathrm{ex}}^{\mathrm{n}}$ are the volume coefficients of resonance absorption and nonresonance losses; I_{s} is the intensity of a resonance transition saturation.

Solution of the problem (1) was represented as an implicit function and was sought by the iteration method of bisection.[5]

The analysis has shown[2] that the inverse problem is resolvable when sounding the medium by three sounding pulses with a variable initial intensity. For example, as the measurement conditions one can select the conditions of linear, weakly nonlinear, and strongly nonlinear interactions.

Under these conditions the scattering signals can be represented as

$$P_1(\lambda_1) = \beta_\pi(z)\; W_1(0)\, \exp[-2(\tau^{\mathrm{m}} + \tau^{\mathrm{n}})]\,, \tag{2a}$$

$$P_2(\lambda_2) = \beta_\pi(z)\; W_2(z, \tau^{\mathrm{m}}, \tau^{\mathrm{n}}, I_{\mathrm{s}})\, \exp[-(\tau^{\mathrm{m}} + \tau^{\mathrm{n}})]\,, \tag{2b}$$

$$P_3(\lambda_1) = \beta_\pi(z)\; W_3(z, \tau^{\mathrm{m}}, \tau^{\mathrm{n}}, I_s)\, \exp[-(\tau^{\mathrm{m}} + \tau^{\mathrm{n}})]\,, \tag{2c}$$

where W_2 and W_3 are the energies of high-power laser pulses propagated at a distance z in the medium, which are calculated from the corresponding problem of propagation taking into account nonlinear interaction; τ^{m} and τ^{n} are the optical depths of resonance and nonresonance components of the medium; P_j are the measured backscattering signals.

The functional relationships for the unknown optical parameters of the atmosphere are determined assuming that the scattering occurs mainly on particles of the atmospheric aerosol (neglect of the Rayleigh scattering): $\beta_\pi^{\mathrm{m}}(z) \ll \beta_\pi^{\mathrm{n}}(z)$, that, as a rule, is valid in the visible and IR ranges.

As the initial data for solving the inverse problem of sounding we used the ratios of backscattering signals P_1/P_2, P_1/P_3, P_2/P_3. It should be noted that these ratios depend only on τ^{m} and τ^{n}. When simulating sounding process, these signals were calculated from

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Eqs. (1) and (2) for the selected in advance optical parameters of the medium.

The solution of the inverse problem was sought by the partition method using *a priori* preset intervals of values of τ_x^m and τ_x^n: for different values of τ_x^m and τ_x^n from these intervals the backscattering signals of P_{1x}, P_{2x}, and P_{3x} were calculated using known regularities of the optical radiation propagation in the medium. The reconstructed values of the medium parameters τ^m and τ^n were calculated from the condition of maximum coincidence of the ratios P_1/P_2 and P_{1x}/P_{2x}; P_1/P_3 and P_{1x}/P_{3x}; P_2/P_3 and P_{2x}/P_{3x}, respectively. In the calculations this problem was reduced to the problem of minimization of the following function:

$$F = (P_1/P_2 - P_{1x}/P_{2x})^2 + (P_3/P_1 - P_{3x}/P_{1x})^2 + (P_2/P_3 - P_{2x}/P_{3x})^2 . \quad (3)$$

Reconstruction of the backscattering coefficient β_π is possible from Eq. (2a) provided that τ^m and τ^n are measured.

3. RESULTS

3.1. Uniform layer of a medium

The method described here can be used in both single-frequency and double-frequency sounding.

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

The investigations into the dependence of the accuracy of optical depth reconstruction on the intensity of high-power sounding pulses I_1 and I_2 have shown that the value of the reconstruction error weakly depends on the radiation intensity when I_2 and I_3 exceed I_s and the ratio of these intensities is more than two.

Below we present some results of reconstruction of the optical depth of resonance and nonresonance components of the medium at single-frequency sounding of a uniform layer of the medium at the initial maximum intensity of high-power sounding pulses $I_2/I_s = 0.5$ and $I_3/I_s = 2$ (the pulses were of Gaussian shape).

Figure 1 shows the dependences of the mean error of reconstruction of the optical depth of the medium components $(\delta\tau^m + \delta\tau^n)/2$ in the absence of interference of the propagation channel (additive error) δ^a. The figure shows that reliable reconstruction of the medium parameters using the initial data begins with the value of optical depths of resonance and nonresonance components of the order of 0.2.

Figure 2 illustrates the dependence of the mean error of reconstruction of the medium parameters $(\delta\tau^m + \delta\tau^n)/2$ on the value of additive error. It is clear that the reconstruction error is of the order of additive error and decreases with the increase of the optical depth of the medium.

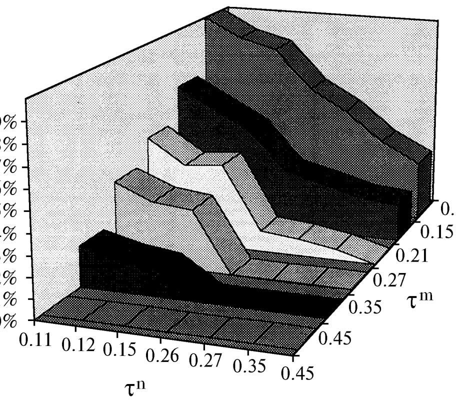

FIG. 1. Mean error of reconstruction of optical depths of the medium components $(\delta\tau^m + \delta\tau^n)/2$ *depending on their values. Conditions of calculation:* $\delta^a = 0$.

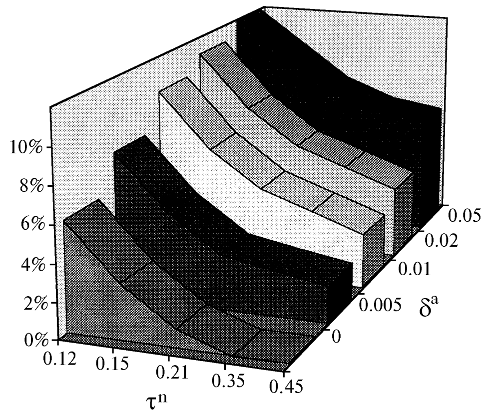

FIG. 2. Mean error of reconstruction of the optical depths of components of the medium $(\delta\tau^m + \delta\tau^n)/2$ *depending on the values of* τ^n *and* δ^a. *Conditions of calculation:* $\tau^m = 0.26$.

We have also investigated the influence of the accuracy of the spectroscopic data used, in particular, the medium saturation intensity, on the reconstruction error. We have revealed that the reconstruction error slightly depends assignment on the accuracy of this parameter.

The calculations demonstrated that the reconstruction error is of the same sign as the introduced additive error. Consequently, at zero mathematical expectation of the additive error one can assume that the averaging of reconstructed parameters of the medium over a series of measurements will essentially decrease the influence of this error.

The saturation effect of resonance absorption can be used also in the double-frequency laser sounding of atmospheric aerosol as a modification of the differential absorption method.[2] In this case in addition to the sounding at wavelengths λ_1 and λ_2 under conditions of linear interaction, as in the differential absorption method, the third high-power pulse is transmitted at the wavelength λ_1. The scattering signals from the first two pulses give, as a rule, the data on the optical depth of gas component τ^m and the third pulse makes it possible to determine the aerosol parameters β^n_π, τ^n.

For implementing the method, the wavelength λ_1 must be within the contour of a resonance absorption line of the medium, and the wavelength λ_2 must be in the adjacent atmospheric microwindow.

Below we present the dependences of reconstruction error of optical depths of the components of a uniform layer at double-frequency sounding.

The calculations show that the reconstruction error decreases with the increase of the optical depth.

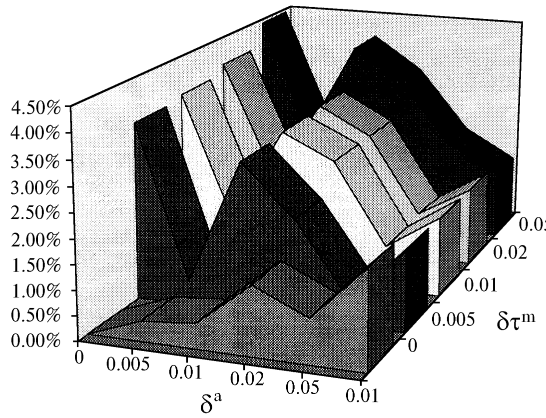

FIG. 3. Calculation of the error of $\delta\tau^n$ depending on the values of $\delta\tau^m$ and δ^a. Conditions of calculation: $\tau^m = 0.35$, $\tau^n = 0.49$.

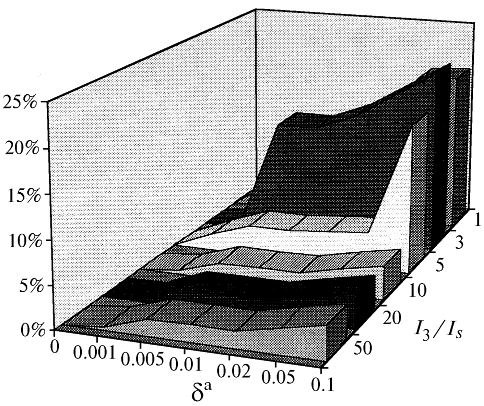

FIG. 4. Calculation of the error of $\delta\tau^n$ depending on the values of δ^a and I_3/I_s. Conditions of calculation: $\tau^m = 0.35$; $\tau^n = 0.49$.

Figure 3 shows the dependence of the reconstruction error of optical depth of nonresonance component of the medium on the additive error δ^a and the reconstruction error τ^m using the method of differential absorption under conditions of linear interaction of radiation with the medium $\delta\tau^m$. The calculations were made for $I_3/I_s = 5$. Note that the value $\delta\tau^m$ slightly affects the result of reconstruction.

Figure 4 shows the influence of the initial intensity value of a high-power pulse on the result of reconstruction. The data illustrate that the influence of additive error decreases considerably with the increase of this intensity.

3.2. Nonuniform layer of the medium

For inhomogeneous atmospheric paths the above algorithm of sounding can be used for layer-by-layer reconstruction of the medium parameters.

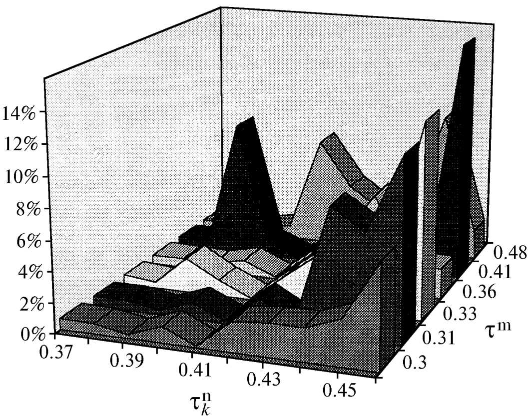

FIG. 5. Calculation of the error of $\delta\tau^n$ in the inhomogeneous medium depending on the values of optical depths τ^m, τ^n_k, where k is the layer number. Conditions of calculation: $\delta^a = 0.001$, $I_3/I_s = 10$.

Figure 5 shows an example of such a layer-by-layer reconstruction of optical depths of components of an inhomogeneous medium at double-frequency sounding. In this case the calculations were carried out for a uniform distribution of a molecular component of a medium, and for a nonuniform distribution the calculations were made for aerosol component. The calculations show that in the presence of additive error the reconstruction error increases with the increase of the layer number, in this case the larger the layer optical depth, the less is the influence of additive error. Note that the uniformity of spatial distribution of the molecular component is not the basic limitation of the method, such a choice is connected with the convenience of graphical representation of the information.

4. CONCLUSION

The paper considers the single-frequency and double-frequency methods of sounding of atmospheric aerosol optical parameters with the use of the saturation effect of resonance absorption by a molecular component of the medium.

The results of calculations have shown that when the investigations fall outside the scope of linear interaction, this makes it possible to perform separate measurements of backscattering coefficients and atmospheric aerosol extinction with the acceptable level of error in reconstructed data. The data obtained show that the reconstruction of these parameters begins to be quite reliable with the values of optical depth $\tau^m \ge 0.1-0.3$.

Now we consider an example of the implementation of sounding methods using the saturation effect. If the sounding is performed by means of a CO_2 laser, the atmospheric CO_2 can be used as a resonance component of the medium. In this case, taking into account the fact that the above mentioned gas is stable and homogeneously distributed in space, the bulk of data measured can be decreased. Typical values of the aerosol extinction coefficient for a weakly turbid atmosphere in this spectral range are of the order of 0.1 km^{-1} (see Ref.2) and, hence, the spatial resolution of this method, by our estimates, is about 1 to 3 km.

To improve the spatial resolution we should use the radiation sources in the visible spectral range with an appropriate choice of resonance gases, since there the aerosol extinction coefficient value is much greater. The spatial resolution of the method will be increased with the growth of the atmospheric turbidity.

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