USE OF NONSTATIONARY INTERACTION EFFECTS TO MONITOR THE ATMOSPHERIC GASEOUS SPECIES

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Potentialities of the techniques for single-frequency sounding of atmospheric gases with the use of laser pulses of variable spectral widths are analyzed. The variable spectral characteristics of the sounding pulses are their duration and the index of phase modulation. Influence of the main errors on the accuracy of reconstructing a resonant gas concentration sounded have also been estimated.

INTRODUCTION

It is well known that extinction of pulses with the spectral width comparable to an absorption line width obeys more complicated regularities than it does in the case of a stationary interaction.[1,2] Therewith, variations of the energy, temporal, and spectral characteristics of radiation depend on the medium parameters and, consequently, may be used for retrieval of information about them. Thus, a single-frequency lidar based on parametric light oscillator using a specialized spectral selector for a broad-band radiation passing through a medium is described in Ref. 3. In Ref. 4 it is proposed to determine parameters of the absorption line with the use of deformation of the pulse shape in a medium of a large optical thickness.

The use of several sources of sounding radiation with essentially different spectral widths (authors used a laser and a thermal source) allows, as it was shown in Ref. 5, to measure concentration of the atmospheric gaseous components. This work was not developed further due to poorer performance characteristics of thermal sources in comparison with lasers.

At present, sufficiently simple technical means providing laser radiation of variable spectrum width have been developed. This allows us to develop a new approach to single-frequency sounding of the atmosphere by laser pulses whose spectral widths vary from pulse to pulse. The possibilities of such sounding techniques are analyzed in this paper.

The problem on propagation of an optical wave was described by the known Maxwell-Bloch system of equations[6] (MBE):

$$\frac{\partial \varepsilon}{\partial z} = 2\pi i k N \mu P , \qquad (1a)$$

$$\frac{\partial P}{\partial \eta} = -\Gamma P - i\varkappa\varepsilon, \qquad (1b)$$

where ε and P are the complex height of an optical pulse and induced polarization of a medium, respectively, μ is the dipole moment of the transition, N is the concentration of resonant particles, $\varkappa = 2\mu/\hbar$, $\Gamma = 1/T_2 - i\Delta$, T_2 is the time of the phase memory of a medium, Δ is the frequency shift off the resonance, $\eta = (t - zn_0/c)$, n_0 is the nonresonant part of the refractive index of a medium, z is the coordinate along the direction of propagation, $k = w/c$. The system (1) is written for a uniform layer of medium neglecting diffraction and assuming conditions of linear interaction.

THE USE OF PHASE MODULATED PULSES

Phase modulation (PM) is one of the techniques changing spectral width of optical pulses. For example, this effect is achieved in nonlinear interaction of radiation with some liquids or in fiber-optical waveguides.[7]

Potentialities of the long path absorption method with the use of PM pulses estimated for concentration measurements are considered below. Formulation of the direct problem as well as a method of its solution (the use of the Fourier transform) was analogous to that in Ref. 1. Therewith, solution of the system (1) within the spectral region can be represented in the form (see, for example, Ref. 6):

$$\varepsilon(z, \nu) = \varepsilon(0, \nu) \exp[-A(\nu) z] , \qquad (2)$$

where $\varepsilon(z, \nu)$ is the Fourier transform of a complex field amplitude varying slowly at the input to medium;

$$A(\nu) = \frac{2\pi N k \mu \varkappa}{1/T_2 + i(\Delta - \nu)} .$$

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In calculations we used the model of Gaussian pulses with linear variation of a carrier frequency with time:

$$\varepsilon(0, t) = \varepsilon_0 \exp [-t^2(1 - i f)/\tau_p^2] , \qquad (3)$$

where τ_p is the pulse duration, f is the index of phase modulation. Such pulses, for example, may be obtained when using a clearing up filter inside laser resonator,[8] non-linear interaction of radiation in the optical waveguide also leads to a linear variation of the frequency in the central portion of the pulse.[9] Dependence of the spectral width of the pulse described by Eq. (3) on the value of f is shown in Fig. 1.

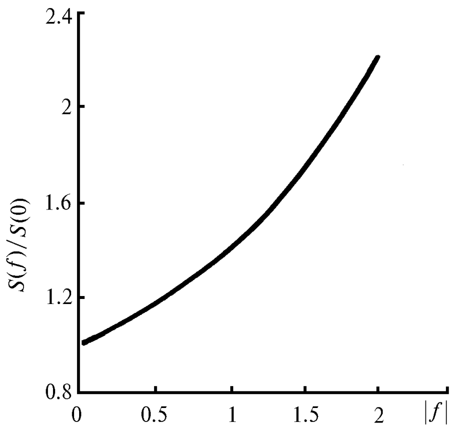

FIG.1. Dependence of spectral width of the Gaussian pulse S(f) on the index of phase modulation f; S(f) is normalized to the spectral width of a similar pulse without the phase modulation (f = 0).

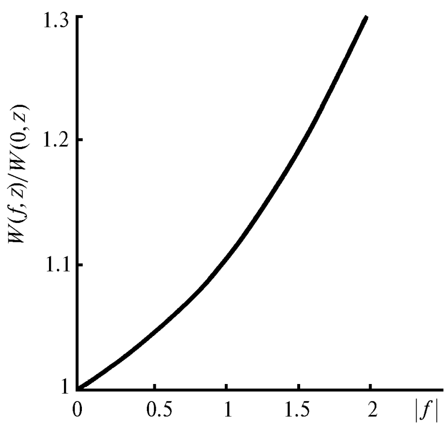

FIG. 2. Energy of Gaussian pulse having passed the 10 km vertical layer of the atmosphere. It is normalized to the energy of the similar pulse without the phase modulation (f = 0). Calculation parameters: $\lambda = 0.69438$ μm, $\tau_p = 1$ ns, $\Delta = 0$, the mid-latitude summer atmospheric model (Δ is the frequency shift from resonance).

The influence of phase modulation on the atmospheric transmission is also interesting. An example of such calculations is presented in Fig. 2. These calculations were based on equations like Eq. (2) but taking into account inhomogeneity of the propagation path[1] and were performed for a ruby laser with $\lambda = 0.69438$ μm and $\tau_p = 1$ ns, a vertical path with the length 10 km, and the atmospheric model for mid-latitude summer. The spectrum distortions estimated for the PM CO_2-laser pulses propagating along the vertical atmospheric paths are considered in Ref. 10.

To estimate the medium absorption characteristics which, in their turn, are determined by the concentration of gas absorbing in resonance, let us consider variation of any energy parameter of radiation along the propagation path. The pulse energy, its spectral density, and their functionals may be used as such parameters. The functional of the energy spectral density is used below as such a parameter:

$$\tilde{W}(z) = \int_{\infty}^{-\infty} |\varepsilon(z, \nu)| \, d\nu .$$

The unknown concentration N may be found from a comparison of the parameters W_k measured at the medium exit for pulses with different phase modulation index, f:

$$\varepsilon_k(0, t) = \varepsilon_0 \exp [-t^2(1 - i f_k)/\tau_p^2] ,$$

for example, from the equation:

$$P(N, z) = 0 , \qquad (4)$$

where

$$P(N, z) = \frac{\tilde{W}_1(z)}{\tilde{W}_2(z)} - \frac{\int_{\infty}^{-\infty} \beta(\nu) \, G(\nu, N, z) \, d\nu}{\int_{\infty}^{-\infty} G(\nu, N, z) \, d\nu} ;$$

$$\beta(\nu) = |\varepsilon_1(0, \nu)| / |\varepsilon_2(0, \nu)| ;$$

$$G(\nu, N, z) = |\varepsilon_2(0, \nu)| \exp[- \mathrm{Re}\, A(\nu) \, z] .$$

Solution of Eq. (4) is possible if spectral widths of the pulses significantly differ and if, in addition, at least one of them has the width comparable with the width of the absorption line. It should be noted that, for decreasing interference between the adjacent absorption lines, the spectral widths of sounding pulses, at least, should not exceed the absorption line width, which is equal to ~ 0.1 cm^{-1} in the near ground atmospheric layer.[11] Fulfilment of these conditions allows Eq. (4) to be solved by any methods of search within an arbitrarily chosen interval $N \in [N_{min}, N_{max}]$, if unknown value $N = N_0$ falls within this interval.

Stability of such a scheme for solution of the sounding inverse problem has been analyzed for the case of equation:

$$P(N, z) + \delta_a \frac{\tilde{W}_1(z)}{\tilde{W}_2(z)} = 0 , \quad (5)$$

where δ_a is the additive error of experimental data due to different noise and the radiation interference in the propagation and receiving channels.

Solution of Eq. (5) for a homogeneous medium layer is analyzed below. Simulation is carried out in the following way. Solution of the direct problem in correspondence with the propagation equation (2) is performed for the chosen medium parameters determining its optical thickness and for the initial parameters of sounding pulses (the index of phase modulation and the pulse duration). Values of $\tilde{W}_i$ obtained in such a way were used as the initial information for solution of an inverse problem, i.e., as the "experimental data." The optical thickness of a medium resonance component $\tau_0 = \sigma N_0 z$ (σ is the cross section of a medium resonant absorption) was unknown parameter in solution of the inverse problem. In other words, the relation (5) was considered as an equation for a given parameter. Solution of this equation has been found by the bisection method, and iterations were performed within the interval $\tau \in [0.2–6]$. This method consists in localization of the region of variations of independent variable N, in which the function $P(N, z)$ is sign-variable. Localization is performed by bisection.

Relative error of retrieval of concentration of a medium resonant component δ_N estimated in such a way is shown in Fig. 3. It is seen from the figure that the algorithm used for retrieval of N_0 is stable to additive errors. We also should note that the value δ_N is minimum for narrow-band laser pulses, when $\tau_p\gamma >> 1$ (γ is the half-width of an absorption line).

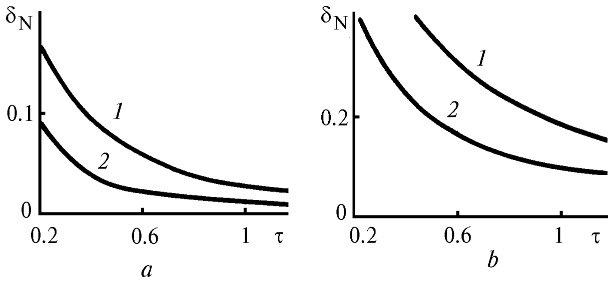

FIG. 3. Relative error in retrieval of concentration of the medium resonant component δ_N at δ_a =1 (a) and 5% (b). Calculation parameters: $f = 0.5$, $\tau_p\gamma = 100$ (1) and 1 (2).

Accuracy of the determination of the pulse carrier frequency relative to the absorption line center is the most important from the spectroscopy information used in this case as well as for the differential absorption method. Calculations have shown that influence of this factor on the final result is negligible, for example, error of determination of a pulse frequency $\delta_\omega = 0.1\gamma$ leads to the error in determination of concentration $\delta_N < 1\%$ at $\tau \geq 0.2$ and $\tau_p = 100/\gamma$.

THE USE OF PULSES WITH VARIABLE DURATION

Other way of change of a pulse spectral width is connected with variation of its duration. Therewith, deviation from the Bouguer law depends on the pulse shape and may occur already at duration $\tau_p \sim (3–10)/\gamma$ (Ref. 12).

The system (1) was used for analysis of the corresponding direct problem. The following model of the initial pulse shape was taken in calculations

$$\varepsilon_i(0, t) = [\sin(\pi t/\tau_{p,i})]^q , \quad t \in [0, \tau_{p,i}] ,$$
$$\varepsilon_i(0, t) = 0 \quad t \notin [0, \tau_{p,i}] .$$

It was varied from a quasi-Gaussian to a quasi-square one depending on the parameter q.

It is obvious that in this case solution of the corresponding inverse problem may be found from the equation analogous to Eq. (4). Difference is in the fact that parameters $\tilde{W}_i$ ("experimental data") correspond to energy of pulses with different initial duration $\tau_{p,i}$, passed through a medium, and calculation of the second term in this equation should be performed considering the model chosen for the propagation process (Eq. (1)). It should be mentioned that solution of the inverse problem may be reduced to the problem of minimization of the goal function $|P(\tau)|$. This function is analyzed below.

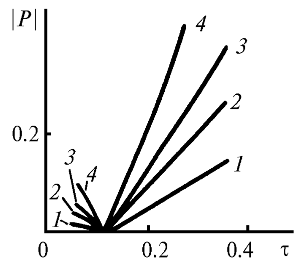

FIG. 4. Dependence of the absolute value of the goal function $|P|$ on the medium thickness τ at $\tau_{p,1}$ = 1.7 ns and $\tau_{p,2}$ = 3.3 ns (1); $\tau_{p,1}$ = 1 ns and $\tau_{p,2}$ = 3.3 ns (2); $\tau_{p,1}$ = 1.7 ns and $\tau_{p,2}$ = 10 ns (3); $\tau_{p,1}$ = 0.3 ns and $\tau_{p,2}$ = 10 ns (4). Calculation parameters: $\tau_0 = N_0\sigma z = 0.1$ and $\delta_a = 0$.

In Fig. 4., the dependence of the absolute value of the goal function $|P|$ on the value τ is shown at different duration $\tau_{p,1}$ and $\tau_{p,2}$. It is seen from the figure that this dependence is most strong if duration of the first pulse corresponds to conditions of stationary interaction: $\tau_{p,1} >> 1$, and duration of the second pulse, which is nonstationary, $\tau_{p,2} \sim 1$. In addition, it is seen that this goal function is nonmonotonic with respect to the value $\tau = \sigma N z$. Hence, solution of the inverse problem may be obtained by any exhaustive method within *a priori* chosen interval as was described above. We note that occurrence of additive error leads to the shift of the goal function whose value weakly depends on τ.

CONCLUSION

Potentialities of single frequency sounding of the atmospheric gases by laser pulses whose spectral width varies from pulse to pulse have been analyzed in this paper. Changes of the pulse duration as well as their index of phase modulation are considered as mechanisms of varying spectral characteristics of sounding pulses. Analysis has shown that the algorithms considered allow one to retrieve concentration of a resonantly absorbing component of a medium. It should be mentioned that they are highly stable relative to influence of noise in the propagation channel as well as to the errors in the initial spectroscopic data.

Applicability limits and the bulk of *a priori* information needed for the considered algorithms coincide practically with that for the method of differential absorption because they are based on the same effect: resonance absorption. So, analysis of possible sources of measurement errors, not discussed in this paper, may be based on the corresponding data for the method of differential absorption (see, for example, Ref. 13).

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