SUPER-SHORT LASER PULSE DISPERSION IN THE ATMOSPHERE

V.P. Kandidov and M.P. Tamarov

M.V. Lomonosov State University, Moscow
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The fine structure of the absorption spectral lines of the air gas components plays a significant role in the process of the pico- and subpicosecond laser pulses dispersion when they propagate through the atmosphere. It is shown by means of numerical solution of exact wave equation jointly with the media material equations. In particular, the account for the fine structure of lines allows the improvement of the spatial resolution of the optical ranging systems. The results of the numerical simulation of a satellite experimental ranging are presented.

The development of femtosecond lasers attracted much interest to the problems of super-short pulses (SSP) propagation in natural media. The systematic error connected with the dispersion in the atmosphere is about 1 cm at satellite laser ranging.[1] To reduce this error, multiwave schemes of picosecond lidars with a common detector are being developed.

Theoretical study of SSP propagation in gaseous media requires new simulations of optical radiation interaction with the matter. The possibilities of the method of slowly changing amplitudes are essentially restricted in the optics of the femtosecond range. In fact, the assumption that the change of the wave packet amplitude is slow remains valid up to the pulse duration of $t_p \approx (3-10)T_0$ where T_0 is the period of optical oscillations.[2] But this method permits one to use only approximations of the dispersion theory whose applicability depends on the properties of the medium. In the second approximation, the pulse blurring is described in the parabolic approximation, and the modulation of light pulse fronts is described within the cubic approximation.[3] The approximations of the dispersion theory are applicable to improvement of the dispersion dependence in the wings of absorption lines, e.g. in the analysis of pulse propagation in optical fibers.[4]

In gaseous media, the spectral band of pico- and subpicosecond pulses can span several tens and even hundreds of absorption lines. The approach based on the approximations of the dispersion theory is not applicable despite slow change of the light pulse amplitude. In order to consider the fine structure of the dispersion characteristics of gaseous media, one should take into account the resonant response on collections of a large number of spectral lines. To analyze SSP propagation, it is necessary to consider directly the system of Maxwell equations with the material equations of the medium.

The Maxwell equations were used in the numerical study of some simulation problems of nonlinear optics. The propagation of a soliton with a small number of periods of optical oscillations in a cubically nonlinear medium has been considered in Ref. 5. Duffing equation describing the response at a single spectral line was used for simulation of the medium. Direct integration of the Maxwell equations in the problem on propagation of a femtosecond pulse in a linearly absorbing half-space with the Lorentz dispersion is performed in Ref. 6. The simulation of the dispersion dependence $\varepsilon(\omega)$ corresponding to an isolated spectral line was realized by integration of second-order differential equations describing variation of electric field strength and induction in time at all points of the space. Subsequently, the method of finite difference integration of the medium and Maxwell equations was generalized to the problem on a femtosecond optical soliton propagation under conditions of Kerr nonlinearity and Raman scattering.[7] However, the approach developed in Refs. 5–7 does not enable the study of SSP propagation in a medium whose dispersion is determined by a large number of spectral lines.

In this paper we study the dispersion of femto- and subpicosecond laser pulses of the optical range at propagation in air. The investigation is performed on the basis of a numerical solution of the system of Maxwell equations in the spectral space and using the HITRAN database on vibrational-rotational absorption lines of molecules of the atmospheric gases.

For a linearly polarized plane wave, the system of Maxwell equations has the form

$$\begin{cases} \dfrac{\partial H_z(x,t)}{\partial t} = -c\dfrac{\partial E_y(x,t)}{\partial x}, \\ \dfrac{\partial D_y(x,t)}{\partial t} = -c\dfrac{\partial H_z(x,t)}{\partial x}. \end{cases} \tag{1}$$

The material equation establishing the connection between the electric field induction $D_y(x, t)$ and strength $E_y(x, t)$ is convenient to be described in the spectral space:

$$\tilde{D}_y(x, \omega) = \varepsilon(\omega)\tilde{E}_y(x, \omega). \tag{2}$$

To obtain the dispersion dependence of the permittivity on the frequency $\varepsilon(\omega)$, we assume that the spectral lines have Lorentzian shape

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$$k_M(\omega) = \frac{\omega}{c}\left(1 + \sum_{m=1}^{M} \frac{A_m}{\omega_{0m}^2 - \omega^2 - i\,2\,\delta_m\omega}\right)^{\frac{1}{2}}, \quad (3)$$

where ω_{0m}, δ_m, A_m are parameters of the mth line, M is the number of lines. This assumption is valid for most of isolated lines in the visible range for the near-ground atmospheric layer where the collisional broadening dominates.[8] According to experimental data,[8] the Lorentzian profile is preserved for the central part of the line providing the maximal contribution to the dispersion dependence for a gas mix. The presentation of $\varepsilon(\omega)$ in the form (3) corresponds to the medium simulation as a set of linear oscillators whose natural frequencies and attenuation coefficients correspond to the central frequencies ω_{0m} and widths δ_m of the spectral lines.

The solution of the system of equations (1) and (2) in the spectral space has the form

$$\tilde{E}_y(x, \omega) = \tilde{E}_{0\omega}\, e^{i\,(k(\omega)x - (\omega/c)x)}\, e^{-i\omega\tau}, \quad (4)$$

where $\tilde{E}_{0\omega} = \tilde{E}_y(x = 0, \omega)$ are complex amplitudes of the pulse spectral components at the beginning of the propagation; $k(\omega) = \left(\frac{\omega}{c}\right)\sqrt{\varepsilon(\omega)}$ is the wave number. The solution (4) is written in the running time $\tau = t - \frac{x}{c}$. To obtain the strength of the pulse field $E_y(x, t)$ at a given distance x, it is necessary to perform the inverse Fourier transform of the expression (4).

When describing the medium response we took into consideration the whole array of spectral lines of the gas components lying within the frequency band of a pulse. The spectroscopic database HITRAN[9] contains much information about the parameters of the molecular absorption lines of 31 gaseous components of the atmosphere in the frequency range from 0 to 23000 cm^{-1}. In particular, it contains data on the position of an absorption line center ν_m [cm^{-1}], self-broadening coefficient $\nu_{m\,\mathrm{self}}$ [cm^{-1}/atm], and air broadening $\gamma_{m\,\mathrm{air}}$ [cm^{-1}/atm] of a line at 296 K, its integral intensity reduced to a single molecule S_m [cm^{-1}/mol·cm^{-2}] (Ref. 10):

$$S_m = \frac{1}{N}\int \alpha(\nu)\, d\nu. \quad (5)$$

The spectral absorption coefficient $\alpha_m(\nu)$ of the mth line of a gaseous component is equal to[10]

$$\alpha_m(\nu) = \frac{S_m\, N}{\pi}\,\frac{\gamma_m}{(\nu - \nu_m)^2 + \gamma_m^2}, \quad \alpha_m(\nu) = 2\ \mathrm{Im}\ k(\nu), \quad (6)$$

where N is the number density of this molecular component. The halfwidth γ_m of the line is defined by the formula

$$\gamma_m = (\gamma_{m\,\mathrm{self}}\,\beta + \gamma_{m\,\mathrm{air}}(1 - \beta))\,P, \quad (7)$$

where β is the partial content of the gas in the air; P is the air pressure, atm.

Expressions (5), (6), and (7) enable one to find the dispersion relations $k(\omega)$ for gaseous components of the air mixture under given conditions using the spectroscopic database HITRAN.

In the numerical calculations, the wavelength of laser radiation was taken to be equal to that of a dye laser $\lambda = 0.65$ μm ($\nu = 15384.6$ cm^{-1}). The pulse was assumed to be spectrally bounded Gaussian one

$$E_y(x = 0,\ t) = E_0 \exp\,(-\,2t^2/t_p^2). \quad (8)$$

Its duration t_p is 20 – 200 fs. Thus the pulse occupied the spectral region from 100 to 1000 cm^{-1}. In this spectral region the strongest absorption lines belong to water vapor ($S \geq 10^{-25}$ cm^{-1}/mol·cm^{-2}). The width of the absorption lines of water vapor was determined for the relative humidity of 70% ($\beta = 0.015$) and normal pressure corresponding to the mean stochastic model of the atmosphere for summer of a mid-latitude zone.[11] As to other gases of the air mixture, this spectral region is either a transmission region, or the intensity of their absorption lines is essentially less and their contribution to the dispersion can be neglected. Nevertheless, the number of lines producing a significant contribution to the dispersion dependence $k(\nu)$ was about 10–100. For instance, the number M of lines for which $S \geq 10^{-25}$ cm^{-1}/mol·cm^{-2} is 110 for the pulse of $t_p = 20$ fs. The initial spectrum $G_0(\nu)$ of a Gaussian pulse of the duration $t_p = 200$ fs at the wavelength $\lambda = 0.65$ μm, the absorption spectrum $\alpha_m(\nu)$ of water vapor in the atmosphere, and the spectrum of the pulse $G(\nu)$ after passing a horizontal path of length $x = 3000$ m are presented in Fig. 1. One can see narrow dips at the frequencies of water vapor absorption lines in the pulse spectrum $G(\nu)$ at the end of the path.

A large number of narrow spectral lines in the frequency band of the considered pulse essentially complicates the numerical investigation of the above formulated problem. Indeed, the spectral resolution of the computation grid must be sufficiently high to reflect the profile of separate absorption lines of air gaseous components. This means that the discretization step in the spectral region $\Delta\nu$ is less than the width of a separate line γ_m

$$\Delta\nu \leq \gamma_m \approx 0.2 - 0.5\ cm^{-1}. \quad (9)$$

At the same time, the time step Δt must be less than the optical oscillation period $\frac{2\pi}{\omega}$ in order to reproduce the oscillations:

$$\Delta t < (2\pi/\omega). \quad (10)$$

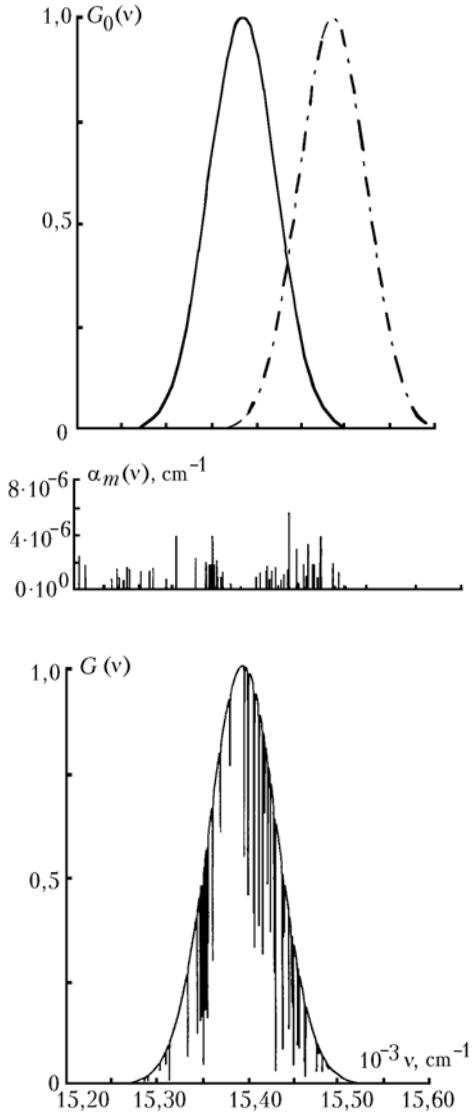

FIG. 1. The initial spectrum $G_0(\nu)$ *of Gaussian pulses* t_p = 200 fs *at the wavelengths* λ = 0.65 μm *(solid line) and* λ = 0.64 μm *(dot-and-dash line), absorption spectrum* $\alpha_m(\nu)$ *of water vapor in the atmosphere, and the spectrum of the pulse* $G(\nu)$ (t_p = 200 fs, λ = 0.65 μm) *after passing a horizontal path of length* x = 3000 m.

Assuming $\Delta t \approx 0.2\frac{2\pi}{\omega}$ or $\Delta t \approx 0.2\frac{\lambda}{c}$ we obtain that the upper boundary Ω of the spectral region satisfies the condition

$$\Omega \geq 5\ (1/\lambda)\ \text{cm}^{-1}. \qquad (11)$$

The number of points in the spectral space for a given step $\Delta\nu$ is

$$N = \Omega/(\Delta\omega). \qquad (12)$$

We obtain the following estimation for the parameters considered

$$\Delta t = 0.43 \cdot 10^{-15}\ \text{s},\ \ \Omega = 77000\ \text{cm}^{-1},\ \ N = 2^{19}.$$

The N-dimensional array of points covers the spectral range from 14000 to 17000 cm^{-1} involving about 300 most intense absorption lines. The temporal behavior of the field intensity in the pulse was considered on the interval $N\Delta t = 2.2\cdot10^{-10}$ sec what made it possible to reproduce the formation of long tails at the pulse trailing edge in numerical experiments with a sufficient reliability.

Large dimension of the arrays processed requires a computer with no less than 8 Mb RAM. The simulation of a separate version of SSP propagation on a PC–486DX4/100 took about 30 min.

Let us consider dispersion distortions of pico- and subpicosecond pulses in the atmosphere. The loss of energy caused by absorption at narrow water vapor lines is small for a pulse of duration t_p = 200 fs. The integral energy loss is 4.4% along a horizontal path of length 3000 m at 70% humidity. The dispersion effect is shown as a long tail of a small amplitude which is formed at the trailing edge (Fig. 2*a*).

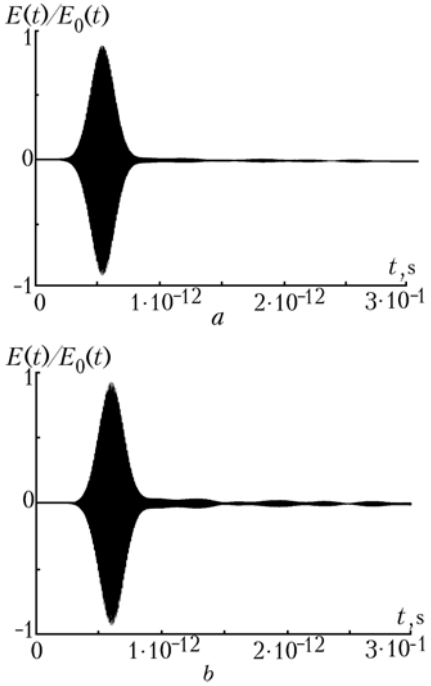

FIG. 2. The electric field strength in the Gaussian pulse t_p = 200 fs, λ = 0.65 μm *after passing a horizontal path of length* x = 3000 m *at air humidity of 70% (a) and the electric field strength of the Gaussian pulse* t_p = 200 fs, λ = 0.65 μm *after passing a horizontal path of length* 6000 m *at air humidity of 70% (b).*

Let us define the root-mean-square duration of a pulse as follows:

$$\tau_{rms} = (\langle\tau^2\rangle - \langle\tau\rangle^2)^{1/2},$$

where $\langle\tau^k\rangle = \left(\int t^k |E(t)|^2\,dt\right) \Big/ \left(\int |E(t)|^2\,dt\right)$, k = 1, 2.

For a Gaussian pulse (8), the rms duration τ_{rms} is connected with the duration t_p by the expression

$t_p = 2\sqrt{2}\ \tau_{rms}$.

The relative increase of rms duration is 2% for the considered parameters.

Figure 2*b* presents a 200 fs pulse passed a distance of 6000 m. The loss of pulse energy increased to 6.9%. In spite of a small absorption, the shape of the pulse was considerably distorted. The amplitude of the tail increased, and the tail took the shape of low-frequency beatings caused by superposition of responses of a large number of oscillators whose frequencies fall within the spectral region of the pulse. The rms duration increased by 4.8%.

The result of SSP dispersion essentially depends on the fact whether the pulse spectrum falls within the absorption band of a gas or not. The numerical experiment shows that the distortion of the pulse shape can be neglected if the whole pulse spectrum is in the band of air transparency or the absorption lines are weak ($S < 10^{-26}$ cm^{-1}/mol·cm^{-2}). However, one should pay much attention to the cases when there are intense absorption lines on the wing of the pulse spectrum. Such a case is shown in Fig. 1 by a dot-and-dash line for a pulse with the duration 200 fs and the carrier frequency $\nu = 15484.6$ cm^{-1}. In this case, the energy loss is 3.2% after passing 6 km in the near-ground layer, the rms duration increases by 1.9%. In this connection, it should be noted that the influence of spectral lines which are far from the carrier frequency increases and the role of dispersion can be considerable for pulses of non-Gaussian shape when the pulse spectrum broadens or the pulse is frequency modulated. The character of the dispersion does not change for shorter pulses. However, the relative contribution of narrow bands of spectral lines of water vapor decreases with the increase of the spectral width of the pulse, and the effect of dispersion distortion of the pulse shape develops at longer distances. Figure 3 presents the electric field strength $E_y(x, t)$ variation within the pulse duration, 20 fs, $\lambda = 0.65$ μm after passing a horizontal near-ground path of 6000 m length and its spectrum. One can see oscillations of the field $E_y(x, t)$ with optical frequency ω_0 and field beats at the trailing edge of the pulse. The integral energy loss was 1.6%, and its rms duration increased by 0.6%. Thus, dispersion spreading only begins to play its role along this path. As the analysis shows, the simplification of the dispersion model by introducing the small number of equivalent lines corresponding to real line bands leads to a qualitatively different character of dispersion distortions.

Figure 4 presents the result of simulation of picosecond pulse propagation under conditions close to the experiment of precise satellite ranging.[1] This experiment was performed in 1993 by Prague investigators together with Austrian scientists. The ranging was performed at three wavelengths $\lambda_1 = 0.45$, $\lambda_2 = 0.532$, $\lambda_3 = 0.683$ μm simultaneously by pulses of duration $t_p = 25$ to 35 ps. The measurements of the difference in the pulse travel time at different wavelengths made it possible to improve the current atmospheric model describing the general dispersion behavior in the considered frequency range. The calibration of the scheme at the near-ground path of 6000 m length shows that the experimental differences in the pulse travel time are different by approximately 10% from theoretical ones obtained for the given meteorological conditions. To improve the ranging accuracy, it is necessary to use more accurate dispersion models of the atmosphere when processing the experimental data.[1]

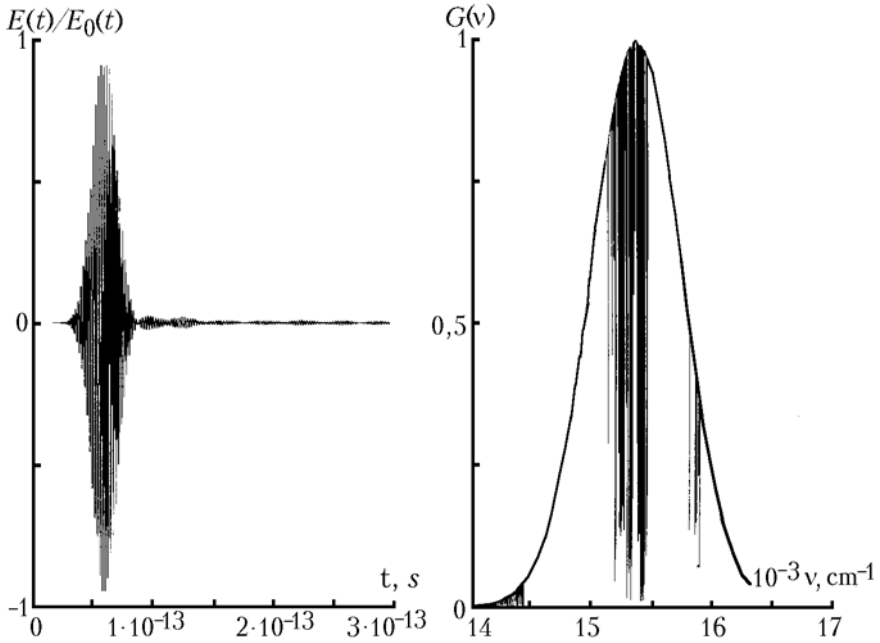

FIG. 3. The electric field strength in the Gaussian pulse $t_p = 20$ fs, $\lambda = 0.65$ μm *after passing a horizontal near-ground path of* 6000 m *length and the spectrum of the pulse.*

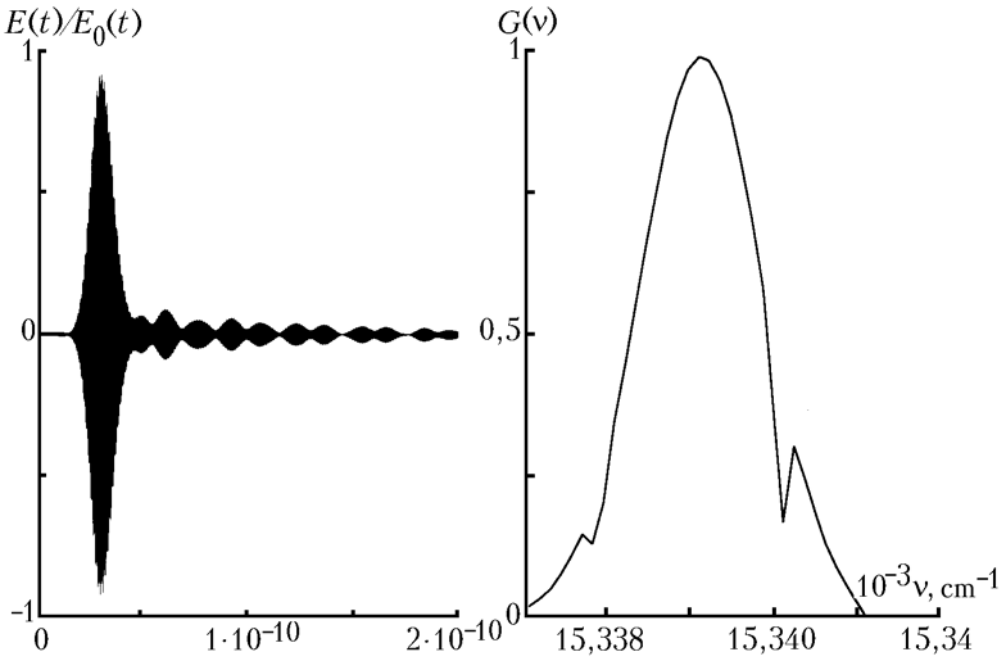

FIG. 4. The electric field strength in the Gaussian pulse t_p = 10 ps, λ = 0.65 μm *after passing a vertical path of* 10 000 m *length, and the spectrum of the pulse.*

The numerical experiment shows that three absorption lines of water vapor fall within the spectrum of a 10 ps pulse at the wavelength λ = 0.65 μm (see Fig. 4). In calculations the intensity and width of lines were assumed to vary with height in accordance with the mean stochastic models of the atmosphere. The integral energy loss is 6.3% at 10 km long vertical path. The increase of the pulse rms duration is more than 12% what is close to the experimental error.[1] Long beats of a considerable amplitude appear at the trailing edge of the pulse, and this can introduce an error into the measurements of the difference in travel time of the ranging pulses.

Thus, dispersion distortions of pulse shapes become essential with the decrease of laser pulse duration to pico- and subpicosecond range at their propagation along the atmospheric paths. To improve the accuracy, it is necessary to take into account the detailed structure of absorption bands in the pulse spectrum.

REFERENCES

1. I. Prochazka, K. Hamal, and G. Kirchner, in: *CLEO'94*, pp. 264–265.
2. K. Blow and D. Weod, IEEE Quantum Electron. **25**, 2665–2673 (1980).
3. S.A. Akhmanov, V.A. Vysloukh, and F.S. Chirkin, *Optics of Femtosecond Laser Pulses* (Nauka, Moscow, 1988), 312 pp.
4. A.V. Belov, S.I. Miroshnichenko, and V.A. Semenov, Sov. Lightwave Commun. **3**, 45–57 (1993).
5. A.V. Vederko, O.B. Dubrovskaya, V.F. Marchenko, and A.P. Sukhorukov, Vestn. Mosk. Univ., Fiz. Astron. **33**, No. 3, 4–20 (1992).
6. R.M. Joseph, S.C. Hagness, and A. Taflov, Opt. Lett. **16**, No. 18, 1412–1414 (1991).
7. P.M. Goorjian, R.M. Joseph, S.C. Hagness, and A. Taflov, IEEE Quant. Electron. **38**, No. 10, 2416–2422 (1992).
8. V.E. Zuev, Yu.S. Makushkin, and Yu.N. Ponomarev, *Spectroscopy of the Atmosphere* (Gidrometeoizdat, Leningrad, 1986), 247 pp.
9. L.S. Rothman, R.R. Gamache, R.H. Tipping, C.P. Rinsland, M.A. Smith, D.C. Benner, V. M. Devi et al., J. Quant. Spectrosc. Radiat. Transfer. **48**, Nos. 5–6, 469–507 (1992).
10. L.A. Kuznetsova, N.E. Kuzmenko, and Yu.Ya. Kuzyakov, *Probabilities of Optical Transitions in Diatomic Molecules* (Nauka, Moscow, 1980), 315 pp.
11. V.E. Zuev and V.S. Komarov, *Statistical Models of Temperature and Gaseous Components of the Atmosphere* (Gidrometeoizdat, Leningrad, 1986), 264 pp.