## EFFECT OF KINETICS OF MOLECULAR ABSORPTION OF RADIATION ON THE PROPAGATION OF A PULSE WITH $\lambda$ = 10.6 µm IN THE ATMOSPHERE

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Much attention has been given to the problem of propagation of wave packets in the atmosphere, which is related to the important use of optical systems for the transfer of energy and information [1, 2]. The transmission of intense radiation through the atmosphere is associated with many nonlinear effects [1], which, for radiation at a wavelength of  $\lambda = 10.6 \mu m$  include thermal self-action in the presence of the resonance absorption component of carbon dioxide.

As noted in [3-8], a consideration of resonance optical excitation requires a detailed account of the kinetics of molecular absorption and thermalization of molecular gas, which, in particular, results in optical brightening and kinetic cooling of the medium. However, we know of no study that investigates the effects of various processes on the parameters of a wave packet propagated along an extended, nonuniform path through the atmosphere.

We will consider nonsteady-state thermal self-action of a collimated Gaussian beam of radiation with a wavelength of 10.6  $\mu$ m transmitted through standard atmosphere [9], which does not contain dust particles or aerosols, along a beam path up to a height of 20 km above the ocean taking into account resonance absorption of radiation by atmospheric carbon dioxide. The initial transmittance through atmospheric CO<sub>2</sub> is ~0.6624 and through water vapor at 100% relative humidity is ~0.828, i.e., the attenuation of radiation related to absorption is substantial. We use the kinetic model in [4] for describing the kinetics of molecular absorption and thermalization in a mixture of CO<sub>2</sub>-N<sub>2</sub>-O<sub>2</sub>-H<sub>2</sub>O. The effect of kinetic cooling and optical brightening of the medium on the form of the beam for different values of the radiation intensity and atmospheric humidity is analyzed. The pulse duration of a rectangular pulse is determined at the moment the beam becomes defocused, where the beam changes over a range from 0.1 to 2 msec.

<u>Theoretical Model.</u> Propagation of a beam of electromagnetic radiation through the atmosphere in the quasioptical approximation is described by the parabolic equation

$$2i\frac{\partial A}{\partial z'} + \frac{k_0}{k}\Delta_{\perp}A + i\frac{\partial \ln k}{\partial z'} + k_0 a^2 \left(2k\frac{\delta n}{n} + i\alpha\right)A = 0,$$

$$k(z) = \frac{\omega}{c}n_0(z), \ k_0 = k(0), \ \Delta_{\perp} = \frac{1}{r'}\frac{\partial}{\partial r'}\left(r'\frac{\partial}{\partial r'}\right),$$
(1)

where  $A = A(r', z', t)/A_0$  is the complex amplitude of the electric field normalized at the maximum of the absolute value of the amplitude of the input radiation along the axis of the beam; the transverse coordinate is normalized over the characteristic radius of the beam r' = r/a. while the longitudinal coordinate is normalized over the diffraction length;  $z' = z/k_0a^2$ ;  $\omega$ , c are the circular frequency of the wave and the velocity of light in a vacuum; n,  $\delta n$  are the index and perturbation index of refraction; the subscript 0 pertains to the unperturbed atmosphere; and  $\alpha = \alpha_1 + \alpha_2$  is the absorption coefficient, which is a sum of the absorption coefficients for carbon dioxide and water vapor.

The density of the gas is related to the index of refraction by the Gladstone-Dale law  $n = 1 + \beta \rho / \rho_S$ , where  $\beta = 2.92 \cdot 10^{-4}$  for air and  $\rho_S$  is the density of the air under standard conditions, i.e., for a pressure of  $p_S = 10^5$  Pa and a temperature of  $T_S = 288.15$  K.

The radiation is absorbed by the carbon dioxide gas due to the transition P(20) [10°0  $\rightarrow$  00°1] and by water vapor at the wing of the absorption band at 6.3 µm. The initial

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dependences of  $\alpha_1$  and  $\alpha_2$  on height above sea level is determined with experimental data in [3]. It is assumed that the saturation of  $\alpha_2$  is absent for absorption of radiation by the H<sub>2</sub>O molecules, since the time for rotational relaxation of H<sub>2</sub>O is elss than the time for VT=relaxation of CO<sub>2</sub> and the offset tuning from the center of the line is high. Because the concentration of CO<sub>2</sub> molecules in the atmosphere is small ( $\xi_1 = 3.18 \cdot 10^{-4}$ ), the process of radiation absorption by carbon dioxide is given on the basis of the kinetic model in [4], i.e., the balance equations for the relative populations of the lower vibrational levels  $x_i$  for the CO<sub>2</sub> and N<sub>2</sub> molecules are

$$\begin{aligned} \frac{\partial x_1}{\partial t'} &= -\frac{1}{4} t_p w \left( x_1 - x_2 \right) - \frac{3}{2} t_p P_{10} \left( x_1 - x_1^0 \right), \\ \frac{\partial x_2}{\partial t'} &= t_p w \left( x_1 - x_2 \right) - t_p Q \left( x_2 - x_3 \right) - t_p P_{20} \left( x_2 - x_2^0 \right), \\ \frac{\partial x_3}{\partial t'} &= \delta t_p Q \left( x_2 - x_3 \right) - t_p P_{30} \left( x_3 - x_3^0 \right), w = \frac{\sigma I}{hv}. \end{aligned}$$

( )

Here,  $x_1$ ,  $x_2$ ,  $x_3$  pertain to the vibrational levels  $10^{\circ}0$ ,  $00^{\circ}1$  (CO<sub>2</sub>) and v = 1 (N<sub>2</sub>);  $x_1^{\circ} = \exp(-\Theta_i/T_0(z))$  are the equilibrium values of these populations;  $\Theta_i$  are the energies (in degrees) of the corresponding levels; I is the intensity of the radiation: h is Planck's constant; v is the frequency of the wave;  $P_{10}$ ,  $P_{20}$ ,  $P_{30}$  are the probabilities of collisional deactivation for the levels  $01^{\circ}0$ ,  $00^{\circ}1$  (CO<sub>2</sub>) and v = 1 (N<sub>2</sub>);  $\Omega$  is the probability of an excitation transition from  $00^{\circ}1$  (CO<sub>2</sub>) to v = 1 (N<sub>2</sub>); w is the probability of optical excitation of CO<sub>2</sub> due to the radiation;  $t_p$  is the pulse duration (t' = t/t<sub>p</sub>); and  $\delta$  is the ratio of the concentrations of CO<sub>2</sub> and N<sub>2</sub>.

The absorption coefficient of carbon dioxide is calculated from the equation  $\alpha_1 = N_1 \sigma(x_1 - x_2)$ , where  $\sigma$  is the cross section for the absorption of radiation by CO<sub>2</sub>, and N<sub>1</sub> is the concentration of CO<sub>2</sub> molecules.

Reaction with a wavelength of 10.6  $\mu$ m corresponds to a transmission window in the atmosphere ( $\alpha$  for 30% relative humidity changes from  $8.6 \cdot 10^{-7}$  at sea level to  $4 \cdot 10^{-8}$  at a height of 18 km [3]). Hence, the perturbation introduced by the beam is weak. For a stationary medium small changes in the density in all the remaining gas parameters are determined by the linearized system of equations for gas dynamics. Therefore,  $\rho = \rho_0(z)(1 + \rho')$ ,  $p = p_0(z)(1 + p')$ ,  $T = T_0(z)(1 + T')$ , where  $\rho_0(z)$ ,  $p_0(z)$  and  $T_0(z)$  are the initial density distribution, pressure, and temperature in terms of height for standard atmospheric conditions, and  $\rho'$ , p', T' are the relative perturbations of these parameters, in dimensionless variables, which yields the following

$$\begin{aligned} \frac{\partial \rho'}{\partial t'} &+ \frac{t_{\mathbf{p}}}{t_{s}} \operatorname{div}_{\perp} v' = 0, \ \frac{\partial v'}{\partial t'} &+ \frac{t_{\mathbf{p}}}{t_{s}} \operatorname{grad}_{\perp} p' = 0, \\ \frac{\partial T'}{\partial t'} &= (\gamma - 1) \left[ \frac{t_{\mathbf{p}}}{t_{c}} \Delta_{\perp} T' - \frac{t_{\mathbf{p}}}{t_{s}} \operatorname{div}_{\perp} v' + F \right], \\ p' &= \rho' + T', \ v' &= v/\sqrt[4]{RT_{0}(z)}, \ \operatorname{grad}_{\perp} &= \frac{\partial}{\partial r'}, \ \operatorname{div}_{\perp} &= \frac{1}{r'} \frac{\partial}{\partial r'}(r'). \end{aligned}$$
(3)

Here, it is assumed that in the quasioptical approximation, the rate of change of the parameters along the propagation axis of the beam is much less than the rate of change in the transverse direction, and, therefore, one can ignore the derivatives with respect to z;  $t_s = a/\sqrt{RT_0(z)}$ ;  $t_c = \rho_0(z)a^2R/\kappa$ ; R is the gas constant;  $\kappa$  is the coefficient of thermal conductivity for air; and  $\gamma$  is the ratio of the specific heats for constant pressure and volume.

The intensity of the distributed thermal source F, due to the absorption of radiation by carbon dioxide molecules and water vapor, has the form

$$F = t_{p} \alpha_{2} I / P_{0}(z) + F_{1},$$
  

$$F_{1} = (t_{p} \xi_{1} / T_{0}(z)) \left\{ 6\Theta_{1} P_{10} \left( x_{1} - x_{1}^{0} \right) + \Theta_{2} \left[ P_{20} \left( x_{2} - x_{2}^{0} \right) + \delta P_{30} \left( x_{3} - x_{3}^{0} \right) \right] \right\}.$$

The system of equations (1)-(3) can be solved for beams with two-dimensional phase fronts for the following initial and boundary conditions

$$A|_{z=0} = \exp(-r'^{2}), A|_{r=\infty} = \frac{\partial A}{\partial r}\Big|_{r=0} = 0,$$

$$x_{1}|_{t=0} = x_{1}^{0}, x_{2}|_{t=0} = x_{3}|_{t=0} = x_{2}^{0},$$

$$v' = p' = \rho' = 0 \text{ when } t = 0 \text{ and } r = \infty,$$

$$\frac{\partial v'}{\partial r} = \frac{\partial p'}{\partial r} = \frac{\partial T'}{\partial r} = 0 \text{ when } \dot{r} = 0.$$
(4)

The parabolic equation (1) is solved with the implicit conservative scheme of secondorder accuracy [10], while the equations of gas dynamics are solved by the modified explicit difference scheme in which the dynamic and thermodynamic functions are related to different (integer or "half-integer") points on the grid [11, 12].

Analytic tests are applied for preliminary control of the results. The modulus of the program used for solving the propagation equation is tested by comparing the numerical results with the analytic solution of system (1), (4) for an unperturbed medium, the kinetic modulus is tested with the approximation solutions of the system (2), (4) for a given radiation intensity obtained in [4], and the gas-dynamic modulus is tested with the asymptotic solution of system (3), (4) for t << t<sub>s</sub> [2] for a given intensity profile of the distributed thermal source F.

<u>Calculation Results</u>. Upon integration of Eqs. (1)-(3), the dimensionless parameter for the distortion of the pulse at the input to the medium  $\tau = (t_p/t_s)^2(t_pP_{10})(t_pw)$  varies over a range from  $10^2$  to  $10^3$ , the rectangular pulse duration varies from 0.1 to 2 msec, and the relative humidity of the air  $\eta$  varies from 0 to 100%. The length of the vertical beam path is 20 km, and the radius of the beam is determined by the condition that the dimensionless length of the beam path is z' = 0.5.

The discrete characteristics that describes the change over time of the beam's intensity distribution at the output cross section include the intensity of radiation transmitted across a square of a different radius

$$P_{0}(t) = 2\pi \int_{0}^{0.5} I(r', 0.5, t) r' dr', P_{1}(t) = 2\pi \int_{0}^{1} I(r', 0.5, t) r' dr',$$

where r' = 1 coincides with the diffraction width for the beam intensity. The moment in time that correspond to the maximum of the function  $P_0(t)$  occurs at the final stage in the focusing of the beam  $t_1$ , if there is focusing, and the moment at which the condition  $P_1(t) < P_1(0)$  is initially satisfied occurs during the initial stage of beam defocusing  $t_2$ . Naturally, the division of the propagation process into two stages is conditional. The intensity  $P_0(t)$  and  $P_1(t)$  will be related to the initial values  $P_0'(t) = P_0(t)/P_0(0)$ ,  $P_1'(t) = P_1(t)/P_1(0)$  (we will omit from now on the primes over  $P_0$  and  $P_1$ ).

We will consider propagation of a beam with an intensity of  $\tau = 2.54 \cdot 10^2$  for  $\eta = 0$ . The energy flow F<sub>1</sub> between the translational and vibrational degrees of freedom leads to a decrease in the temperature of the gas within the beam, which changes the density and the index of refraction of the medium. The distribution  $\rho'(r')$  for  $\eta = 30\%$  at z = 0, 6, and 14 km is illustrated in Fig. 1a with solid, dashed, and dot-dash lines, where 1, 2 are the initial moments in time for focusing t<sub>1</sub> and defocusing t<sub>2</sub>. It is evident that kinetic cooling leads to the formation of a density distribution with a negative gradient near the axis of the beam, i.e., to the formation of a gas lens. A change in the optical properties of the medium changes the form of the beam.

We will trace the evolution of the beam using the plots for the changes in the powers  $P_0$ ,  $P_1$  over time (Fig. 1b, where the solid, dashed, and dot-dash lines correspond to  $\eta = 0$ ; 30; 100%). The basic mechanism behind self-action of the beam is nonlinear refraction. The beam is initially focused and the value of  $P_0$  increases up to 1.72; 1.73; 1.94 for  $\eta = 0$ ; 30; 100%, respectively, after which the beam is rapidly defocused.

However, it is evident from Fig. la that there are no substantial distortions of the field density gradient between the time the focusing ends and the time the defocusing begins. We will consider an amplitude distribution |A(0, z, t)| over the beam axis (Fig. 1c) at the times t<sub>1</sub> and t<sub>2</sub> (curves 1 and 2), where the dashed and dot-dash lines corresponds to  $\eta = 30$  and 100%. It follows from the amplitude distributions of the wave along the z axis that the







collimated beam is focused in a plane that is far from the assigned output cross section (at a height of 6-7 km for  $\eta$  = 30% and 9-10 km for saturated air).

The difference between the amplitude distributions of the wave over the axis for saturated air is due to the fact that kinetic cooling is not observed up to a height of  $\sim 2.5$  km, and heating of the medium due to the flow of energy over the instantaneously relaxing water vapor is greater than the output of energy in the vibrational degrees of freedom for the molecule (F > 0).

It follows from the condition [2]  $(L/\lambda)\beta\rho' \geq 1$ , where L is the length of the beam path, that the relative change in  $\rho'$  at which significant refraction of the beam begins is on the order of  $2 \cdot 10^{-6}$ . As is evident from Fig. 1a, for a relative change in  $\rho'$  over the first half of the beam path, there arises a quasisquared profile for the index of refraction in the central region (r' < 1)

$$n(r) = n(0) \left( 1 - \frac{k_2}{2k_0} r^2 \right)$$
(5)

 $(k_2 \text{ is a constant, } k_2 > 0)$ . If the real distribution n(r, z) over the first half of the beam path is substituted by an effective distribution of the form (5) that is uniform over height, then one can construct an analytic solution for the Gaussian beam in this lens-like medium, where it is found that the coordinate of the plane in which the beam has minimal width is  $z_x = (\pi/2) k_0/k_2$ , and the amplitude at the axis in this plane is

$$A_{*} = k_{0}a^{2} \sqrt{\frac{k_{2}}{k(z_{*})}} \exp\left(-\frac{1}{2}\int_{0}^{z_{*}} \alpha dz\right).$$

For  $\langle \rho'(0, z) - \rho'(0, 8, z) \rangle = 3 \cdot 10^{-6}$  we obtain  $z_* = 7.2 \text{ km}$ ,  $A_* = 2.26$ , which corresponds with the results given in Fig. 1c. Defocusing of the beam occurs earlier than the end of kinetic cooling. In this event, one can use the method of amplitude-phase beam correction.

With an increase in the parameter  $\tau = 6.84 \cdot 10^2$ ,  $\eta = 0\%$  while holding the remaining parameters of the medium and the beam constant, the propagation of the beam is qualitatively the same as it was earlier. According to the plots, a change in the intensity gives rise to

a boundary layer that corresponds to a process of resonance brightening of the atmosphere. The beam is then focused, where the maximum value of the power is  $P_0 = 2.2$  for  $\eta = 100$ . Defocusing of the beam at the output cross section occurs, as in the previous case, due to the small focal length of the gas lens formed in the beam.

If the parameter  $\tau$  is increased to a value of  $2.72 \cdot 10^3$  for  $\eta = 0$ , the propagation of the beam in the atmosphere changes dramatically. The change in the powers  $P_0$  and  $P_1$  over time is shown in Fig. 2a. Intense radiation causes rapid resonance brightening of the medium. Then, an adjacent section is formed that corresponds to propagation of the beam in an optically brightened medium with weakly perturbed density. A change in the density of the medium for  $\eta = 0$  leads to slight focusing of the beam, but only defocusing occurs for  $\eta = 30$  and 100%.

If the condition t <<  $t_s$  is satisfied for a rectangular pulse, it follows from system (3) that the density is a function of the energy flow F according to

$$\rho' \sim \frac{t^3}{3!} \Delta_\perp F.$$

For the indicated value of the distortion parameter of a beam  $\tau$  in a brightened region, one observes "complete saturation" [4], where the energy flow is not a function of the intensity of the radiation and  $\Delta_{\perp} F \approx 0$ . This peculiarity in the distribution F(r) results in a density field with a positive gradient near the axis of the beam. The density distribution at z = 0 and 6 km (the solid and dashed lines on Fig. 2b, respectively, where 1, 2 are the times  $t_1$  and  $\tau_2$ ;  $\eta = 0$ ) indicates the maximum density is off the axis at a distance approximately equal to the diffraction width of the beam for an intensity where the rays deviate from the axis to the periphery within this radius. A ring-like region is formed farther away from the axis in which the density gradient is negative, and, therefore, rays that are incident in this region will deviate toward the axis of the beam. Because of this, the beam is distorted. The distribution for the absolute value of the wave amplitude on the z axis is illustrated in Fig. 2c, where the solid, dashed, and dot-dash lines pertain to  $\eta = 0$ ; 30; and 100%, and 1, 2 are the times  $t_1$  and  $t_2$ .

Therefore, for comparatively small radiation intensities, the predominant mechanism behind self-action of a beam in the atmosphere is nonlinear refraction caused by kinetic cooling. If the intensity of the radiation is increased, nonlinear saturation of the absorption coefficient for carbon dioxide plays a greater role. If  $\tau \sim 10^3$ , complete saturation of the energy flow between translational and vibrational degrees of freedom within the beam gives rise to zones with alternating signs for the refraction gradient, which distorts the beam. The presence of water vapor increases the degree of focusing and delays the time at which defocusing of a collimated Gaussian beam begins, while for high intensities, these characteristics decrease.

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## LITERATURE CITED

- 1. V. E. Zuev, Propagation of Laser Radiation in the Atmosphere, Radio i Svyaz', Moscow (1982).
- 2. Propagation of Laser Radiation in the Atmosphere [Russian translation], Mir, Moscow (1981).
- A. D. Wood, M. Camac, and F. T. Gerry, "Effects of 10.6 µm laser-induced air chemistry of the atmospheric refractive index," Appl. Optics, <u>10</u>, 1877 (1971).
   B. F. Gordiets, A. I. Osipov, and R. V. Khokhlov, "Cooling of gas with the transmission
- B. F. Gordiets, A. I. Osipov, and R. V. Khokhlov, "Cooling of gas with the transmission of intense radiation from a CO<sub>2</sub> laser through the atmosphere," Zh. Tekh. Fiz., <u>44</u>, 1063 (1974).
- 5. F. G. Gebhardt and D. C. Smith, "Kinetic cooling of a gas by absorption of  $CO_2$  laser radiation," Appl. Phys. Lett., <u>20</u>, 129 (1972).
- 6. V. A. Vysloukh and L. I. Ognev, "Resonance self-focusing in a mixture of  $CO_2$  and  $N_2$ ," Prikl. Mekh. Tekh. Fiz., No. 4 (1980).
- 7. V. D. Gora, N. N. Karamzin, and A. P. Sukhorukov, "Self-action of light beams with resonance absorption," Kvant. Elektronika, <u>7</u>, 720 (1980).
- 8. V. A. Levin, V. V. Netesov, and A. M. Starik, "Analysis of the propagation of a radiation pulse with  $\lambda = 10.6 \mu m$  through an absorbing medium," Prikl. Mekh. Tekh. Fiz., No. 3, (1984).

9. Tables of Physical Quantities. Handbook, ed. I. K. Kikoin, Atomizdat, Môscow (1976).

- 10. N. S. Bakhvalov, Ya. M. Zhileikin, and E. A. Zabolotskaya, Nonlinear Theory of Sonic Pulses, Nauka, Moscow (1982).
- 11. R. D. Richtmyer and K. W. Morton, Difference Methods for Initial-Value Problems, 2nd ed., Wiley (1967).
- 12. A. A. Samarskii and Yu. P. Popov, Difference Methods for Solving Problems of Gas Dynamics, 2nd ed., Nauka, Moscow (1980).

## TRANSFER OF GAS-ION MOMENTUM AND ENERGY TO AN ELECTRICALLY CONDUCTIVE SURFACE PARTIALLY COATED BY A THIN DIELECTRIC LAYER

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The dynamic interaction of bodies with a rarefied gas flow is characterized to a significant extent by exchange of momentum and energy, or the corresponding accommodation coefficients. The momentum and energy accommodation coefficients are used to determine aerodynamic characteristics and heat exchange of bodies in a rarefied medium and are an important element of the computation relationships no matter what pattern of gas-atom interaction with the surface is chosen.

The present study will examine interaction of gas atoms with purely crystalline structures. There are available a significant number of studies which have performed numerical simulations of atomic particle collisions with a solid surface and offered approximate analytical solutions characterizing the mechanism of gas-atom momentum and energy transfer to ideal crystalline surfaces [1, 2]. In practical targets with an ideal single-crystal structure are rarely found. In the majority of cases the surface flowed over a polycrystalline structure with individual crystallites randomly oriented. In numerical study of collision of atomic particles with an atomically smooth polycrystalline surface it is necessary to average the interaction characteristics, which significantly complicates the problem [3]. The situation becomes even more complicated for technological materials such as alloys with complex structure and surface relief.

The literature provides an insufficient volume of data on calculated and experimental values of gas particle accommodation coefficients for the velocity range  $u_{\infty} \cong 10$  km/sec which is of practical interest in aerodynamics. Experimental values of the accommodation coefficients are few in number, and refer to varied experimental conditions. Calculations of aerodynamic characteristics and heat exchange of bodies moving in a rarefied medium require knowledge of a complex of parameters characterizing the dynamic interaction of the body with the incident flow. In connection with this it becomes necessary to perform general studies to determine the gas-particle momentum and energy accommodation coefficients of a number of factors characterizing interaction of a gas with a surface upon the values of the momentum and energy accommodation coefficients of the surface of an aluminized polymer film with a conductive face surface coated by a glass screen (dielectric grid with transparency coefficient of  $\sim 0.12$ ), the outer surface of vacuum-screen thermal insulation [4].

1. The force action of a flow of partially ionized gas of low density on a surface being flowed over having a "floating" potential is determined by bombardment of electrons, ions, rapid and slow neutral particles produced by ion charge exchange with the residual gas, metastable particles, etc.:

$$F_{2} = F_{e} + F_{i} + F_{n} + F_{0} + F_{m} + \dots = F_{e}(V) + F_{i}(V) + \Delta F.$$

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