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NUMERICAL INVESTIGATION OF THE PROPAGATION OF A PULSE OF RADIATION WITH λ = 10.6 μm THROUGH ABSORBING MEDIA

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The passage of electromagnetic radiation through gaseous media is of special interest when reasonantly absorbing impurities are present in the gas. The interaction of radiation with such a medium can lead, for example, to a temporal decrease of the gas temperature or to its strong heating [1-3]. At the same time the index of refraction in the channel of the light beam is altered, which leads to a deviation of the light rays from the initial direction. The main characteristics of such thermal self-action within the framework of linear absorption theory for steady and nonsteady processes have been discussed in [4-12]. Nonequilibrium processes in the medium upon absorption of resonant radiation were not taken into account. The effect of the kinetics of vibrational energy exchange on the state of a medium upon the propagation of radiation through a mixture of CO_2 and N_2 gases was first considered in [2, 13, 14]. However, the simplest models of vibrational energy exchange were used, and saturation of the absorbing transition P20 [$10^0 0 \rightarrow 00^0 1$] in the CO_2 molecule was not taken into account. Thus linearized equations of vibrational kinetics were used in [13], and only one channel of relaxation of asymmetric vibrations of CO_2 and excited nitrogen was considered in [14]. The propagation of a pulse of radiation with λ = 10.6 μm through an absorbing medium is investigated and the influence of the saturation effect and nonlinear processes of vibrational energy exchange on the self-action of light beams of Gaussian profile is studied in this paper.

1. Theoretical Model. The propagation of electromagnetic radiation in a medium with dielectric constant ε in the quasioptics approximation is described by the following equation for the complex amplitude A(z, r, t):

$$2ik\left(\frac{\partial}{\partial z}+\frac{n_0}{c}\frac{\partial}{\partial t}\right)A+\Delta_\perp A+k^2\left(\frac{\varepsilon}{n_0^2}-1\right)A=0,$$
$$k=\frac{\omega}{c}n_0,\quad \Delta_\perp=\frac{1}{r}\frac{\partial}{\partial r}\left(r\frac{\partial}{\partial r}\right),\quad \varepsilon=\left(n+i\frac{c}{2\omega}k_\nu\right)^2, \tag{1.1}$$

where n is the index of refraction (a subscript "0" refers to the unperturbed medium), ω is the frequency of the wave, c is the speed of light, and k_ν is the absorption coefficient.

A change in the state of the medium upon absorption will lead to a change in both the real and imaginary parts of the dielectric constant. Actually, when the temperature of a gas varies, its density ρ changes, which is related to the index of refraction by the Gladstone–Dale law $\delta n = (n_0 - 1)\delta\rho/\rho_0$. On the other hand, the absorption of photons of an electromagnetic field will result in disruption of the initial distribution of molecules by energy levels of the internal degrees of freedom, and as a consequence, in a change in the absorption coefficient k_ν, whose value determines the imaginary part of the dielectric constant. When

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describing a change in the state of a medium under the action of resonant radiation, we shall simulate the vibrations of CO_2 and N_2 molecules with a harmonic oscillator, and we shall assume the distribution over rotational energy levels to be the equilibrium one.

Let us consider the propagation of a pulse whose duration τ_I is much longer than the propagation time of a sound wave through the transverse cross section of the beam.

In this case $\delta\rho = -\rho_0\delta T/T_0$, and the following equations are valid for determination of the state of the medium at any time:

$$\rho C_p \frac{\partial T}{\partial t} = \varkappa\Delta_\perp T + k_\nu I - \rho\sum_{i=1}^{M}\gamma_i\frac{R}{\mu}\sum_{j=1}^{s}\Theta_{ij}\frac{d\varepsilon_{ij}}{dt}, \tag{1.2}$$

$$C_p = \frac{R}{\mu}\left(\frac{5}{2}+\sum_{i=1}^{L}\gamma_i+\frac{3}{2}\sum_{i=L+1}^{M}\gamma_i\right), \quad I = \frac{cn}{8\pi}|A|^2;$$

$$\frac{d\varepsilon_{ij}}{dt} = f_{ij} + \left(\frac{d\varepsilon_{ij}}{dt}\right)_I \equiv F_{ij}, \tag{1.3}$$

where T is the temperature of the translational and rotational degrees of freedom of the gas molecules, $\varkappa$ is the thermal conductivity coefficient, R is the universal gas constant, μ is the molecular weight of the mixture $\left(\mu = \sum_{i=1}\mu_i\gamma_i\right)$, L is the number of molecular components composed of linear rigid molecules, M is their total number, γ_i is the molar fraction of the i-th component in the mixture (i = 1 refers to CO_2, and i = 2 refers to N_2), s is the number of types of vibrations in a molecule of the i-th component, $\varepsilon_{ij} = r_{ij}[\exp(\Theta_{ij}/T_{ij})-1]^{-1}$, r_{ij} is the multiplicity of the degeneracy of the j-th vibration, Θ_{ij} and T_{ij} are the characteristic and local vibrational temperatures of the j-th mode, f_{ij} is the term responsible for V–V' and V–T processes in the mixture of gases under discussion, and $(d\varepsilon_{ij}/dt)_I$ determines the variation of the number of photons in the j-th mode as a result of induced transitions.

The absorption of radiation whose frequency is resonant to the frequency of the vibrational–rotational transition P20 [$10^0 0 \rightarrow 00^0 1$] of the CO_2 molecule (λ = 10.6 μm) has been discussed. The widely accepted model of local vibrational temperatures T_j, where the values j = 1, 2, 3, 4 correspond to symmetric, deformational, and asymmetric types of vibrations of CO_2 and vibrations of N_2, was used in the determination of f_{ij} [15, 16]. The relaxation scheme of processes in a CO_2–N_2 mixture was taken to be the same as in [15]. The finite rate of interaction between symmetric and deformational vibrations of CO_2 was taken into account in accordance with [17]. In the model under discussion

$$\left(\frac{d\varepsilon_{ij}}{dt}\right)_I = (\delta_{1j}-\delta_{3j})\frac{k_\nu I\mu}{\rho N_A h\nu_I\gamma_1},$$

where δ_{ij} is the Kronecker delta, h is the Planck constant, ν_I is the frequency of the radiation, and N_A is Avogadro's number. The value of the absorption coefficient was calculated by the formulas of [16]. It was assumed that the impact broadening cross sections do not depend on temperature. The values of the rate constants of elementary processes were determined from the data of [15-18].

The system of equations (1.1)-(1.3) was solved numerically for beams with a plane phase front under the following boundary and initial conditions:

$$A(z=0, r, t) = A_0\exp\left(-\frac{r^2}{2a^2}\right)g(t), \tag{1.4}$$

$$A(z, r=\infty, t) = 0, \quad \frac{\partial}{\partial r}A(z, r=0, t) = 0,$$

$$T(z, r=\infty, t)=T_0, \quad \frac{\partial}{\partial r}T(z, r=0, t)=0,$$
$$T(z, r, t=0)=T_0, \quad \varepsilon_{ij}(t=0)=\varepsilon_{ij}(T_0),$$

where a is the characteristic size of the beam, A_0 is the maximum amplitude, and g(t) is a function which characterizes the temporal modulation of the amplitude.

2. Method of Numerical Integration and Computational Results. The main difficulty with numerical integration of the system (1.1)-(1.3) is the necessity of calculating high-frequency oscillations of the phase and the variation of the amplitude of the electromagnetic wave which arises upon propagation of the beam. A transformation of the complex amplitude which uses an exact analytic solution for Gaussian beams propagating in media with a constant absorption coefficient permits eliminating this difficulty and restricting ourselves to calculation of the variation of the amplitude and phase of the electromagnetic wave caused only by nonlinear effects [10]. This method, which has received the name "nonadaptive", has been used in this paper.

Introducing the dimensionless variables

$$r'=r/a\sqrt{D}, \quad t'=\frac{t-zn_0/c}{t_T}, \quad dz'=\frac{dz}{ka^2D^2},$$
$$D=\left(\frac{z}{ka^2}\right)^2+\left(1-\frac{z}{f}\right)^2, \quad t_T=\frac{\rho C_p a^2}{\varkappa},$$

where f is the radius of curvature of the phase front, one can represent the system of equations (1.1)-(1.3) with the boundary conditions (1.4) in the following form (the primes are omitted from here on):

$$2i\frac{\partial A_1}{\partial z}+\Delta_\perp A_1+A_1\left[2-r^2+k^2a^2D\left(\frac{\varepsilon}{n_0^2}-1\right)\right]=0; \tag{2.1}$$

$$\frac{\partial T}{\partial t}=\frac{1}{D}\Delta_\perp T-\frac{t_T R}{\mu C_p}\sum_{i=1}^{M}\sum_{j=1}^{s}\Theta_{ij}f_{ij}\gamma_i; \tag{2.2}$$

$$\frac{d\varepsilon_{ij}}{dt}=t_T F_{ij}; \tag{2.3}$$

$$A_1(z, r=R_a, t)=0, \quad A_1(z=0, r, t)=g(t)\exp\left(-\frac{r^2}{2}\right), \tag{2.4}$$
$$\frac{\partial}{\partial r}A_1(z, r=0, t)=0, \quad \frac{\partial}{\partial r}T(z, r=0, t)=0, \quad T(z, r=R_a, t)=T_0,$$
$$T(z, r, t=0)=T_0, \quad \varepsilon_{ij}(t=0)=\varepsilon_{ij}(T_0),$$

where R_a is the beam radius and A_1 is the complex amplitude transformed in accordance with [10].

Conditionally stable symmetric implicit difference schemes of second-order accuracy [19] were used in the numerical integration of Eqs. (2.1)-(2.3). The solution of the system of difference equations was carried out by an iteration method. In each iteration difference analogs of Eqs. (2.1) and (2.2) were solved by the righthand sweep method, and of Eq. (2.3), by Newton's method [19].

A specific analysis of the effect of saturation of an absorbing transition on the propagation of a pulse of radiation with $\lambda = 10.6$ μm was performed by the example of CO_2—N_2 media containing different amounts of CO_2: $\gamma_1 = 1$, $\gamma_1 = 0.25$, and $\gamma_1 = 0.05$. The unperturbed parameters of the medium (temperature T_0 and pressure p_0) were taken equal to 293°K and 10^{-3} MPa. With the selected T_0 and p_0 the relaxation times and thermal conductivity time t_T are comparable, and the absorption coefficients for all the media under discussion are equal to 0.11, 3.12×10^{-2}, and 6.48×10^{-3} m^{-1}, respectively. The maximum intensity of the radiation is $I_0 = 5 \times 10^7$ W/m², and the effective beam radius $a = 1.3 \times 10^{-2}$ m. The time dependence of the amplitude was determined by the formula $g(t) = 1.1 - \exp(-10^5 t)$. The path length was 100 m ($z' = 1$). With the selected value of z one can consider the indicated media to be strongly, moderately,

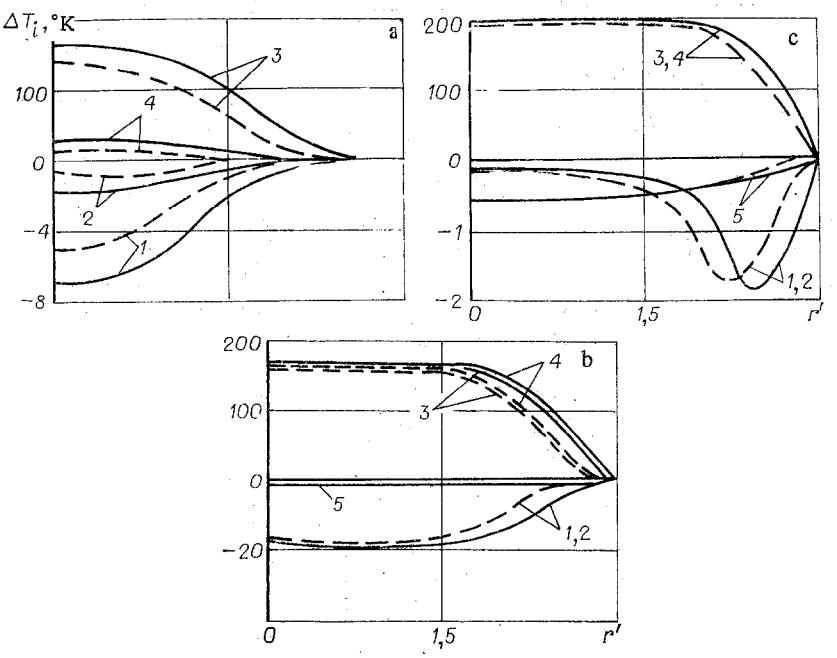

Fig. 1

and weakly absorbing, respectively. The characteristic absorption lengths for these media are equal to 9.1, 32, and 155 m, respectively. We note that the propagation conditions under discussion and the parameters of the medium are typical in the conduct of experiments with laser beams.

First we shall consider the change in state of the medium in time upon the action of resonant radiation. The distribution of the vibrational T_i ($i = 1, 2, 3, 4$, curves 1-4, respectively) and the translational T (curve 5) temperatures over the relative radius of the beam r' is shown in Fig. 1a, b, and c for characteristic times τ = 8.3, 85.7, 807 μsec in a gas with $\gamma_1 = 0.05$. Here the solid curves correspond to the parameters in the cross section z' = 0, and the dashed curves correspond to those in the cross section z' = 1. It is evident from the distributions presented that due to absorption by the gas of radiation whose frequency is resonant to the frequency of the vibrational-rotational transition P20[$10^0 0 \rightarrow 00^0 1$] in the CO_2 molecule a decrease occurs in the populations of the levels of the symmetric mode of CO_2 (temperature T_1), and on the contrary an increase occurs in the population of the levels of the asymmetric mode (temperature T_3) (see Fig. 1a). We note the absence of equilibrium at τ = 8.3 μsec between the vibrations of N_2 and the asymmetric mode of CO_2 ($T_3 \neq T_4$) and also between the deformational and symmetric vibrations of CO_2.

The breakdown of thermodynamic equilibrium between the deformational vibrations and the translational random motion of the CO_2 molecules leads to the appearance of a flow of energy from the translational degrees of freedom to the deformational mode of CO_2, and as a consequence, to a decrease in the gas temperature (see Fig. 1b). We note that at τ = 85.7 μsec the depth of kinetic cooling of the three media under discussion is a maximum for the gas with $\gamma_1 = 0.05$ ($\Delta T = 0.5$°K). This circumstance is explained by the fact that in the medium with a higher CO_2 content the absorbing transition is very rapidly saturated, and in the medium with $\gamma_1 = 0.05$ no such saturation occurs at τ = 85.7 μsec. Therefore at this time k_ν is greatest for the gas with $\gamma_1 = 0.05$, and consequently, the flow of energy from the translational degrees of freedom to the deformational vibrations of CO_2 is a maximum. Figure 2, in which the distribution of k_ν over the beam radius at τ = 8.3, 85.7, and 807 μsec (curves 1-3, respectively) is presented, illustrates the variation in time of the absorption coefficient due to saturation. As before, here the solid curves correspond to the parameters in the cross section z' = 0, and the dashed curves correspond to those in the cross section z' = 1. A decrease in the value of k_ν due to saturation leads, first of all, to a decrease in the flow of energy to the vibrational degrees of freedom of the mixture molecules, and secondly, to a more uniform variation of temperature over beam radius. The variation of the translational temperature in the propagation plane (r' × z') for τ = 85.7 μsec calculated on the assumption of the absorption coefficient being independent of light intensity (there is no saturation effect) (Fig. 3a) and with this effect taken into account (Fig. 3b) is shown for comparison in Fig. 3a and b. It is evident that when the approximation of a constant absorption coefficient

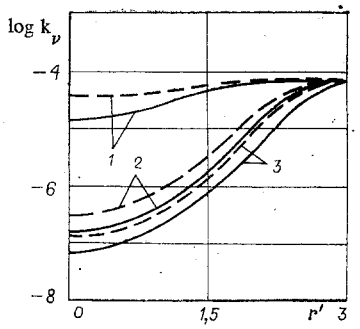

Fig. 2

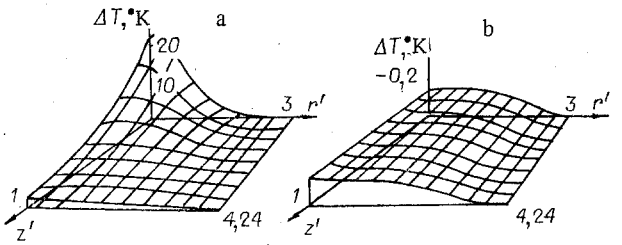

Fig. 3

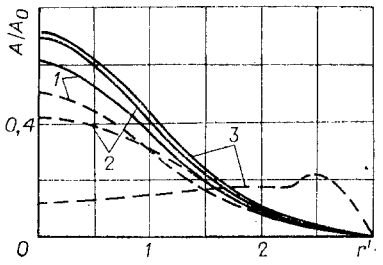

Fig. 4

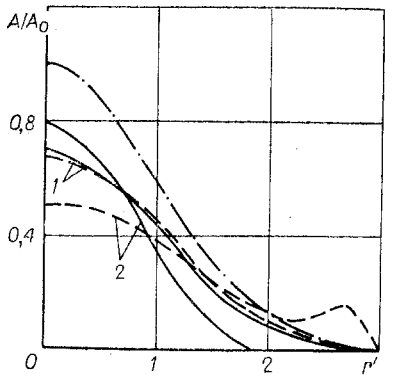

Fig. 5

is used the temperature field in the beam channel is nonuniform, and in addition heating of the medium is observed instead of cooling.

The indicated peculiarities lead to the fact that defocusing of the beam occurs appreciably earlier in time when the approximation of a constant gain coefficient is used than predicted by the theory which takes into account saturation of the absorbing transition. Figure 4, in which the variation of the relative amplitude of the electromagnetic wave A/A_0 over beam radius r' for τ = 8.3, 85.7, and 807 μsec (curves 1-3, respectively) calculated with account taken of saturation of the absorbing transition (solid curves) and on the assumption k_ν = const (dashed curves) is shown, illustrates this. The numerical calculations performed have also shown that at the time the medium is illuminated, when $k_\nu \approx 0$, and all the absorbed energy is located in the vibrational degrees of freedom, one can use the analytic solution of [10] for calculation of the amplitude of the electromagnetic wave. The approximation k_ν = const gives complete defocusing of the beam up to this time.

An increase in the CO_2 content in the mixture (γ_1 = 0.25 and 1) leads to earlier (in time) saturation of the absorbing transition, and as a consequence, to more rapid heating of the medium. Thus in a medium with γ_1 = 1 intense relaxation of the energy of asymmetric vibrations into translational degrees of freedom starts already at τ = 85.7 μsec and k_ν = 0 as a result of intramolecular V—V' exchange, and the gas temperature increases. In the following equilibrium is established between the processes of induced absorption and V—V' exchange, and heating of the gas occurs mainly due to liberation of the energy defect in connection with V—V' exchange. Further action of radiation on the medium will result only in its heating.

At $\tau = t_T$ the variation of the translational temperature over beam radius becomes more uniform. More rapid heating in media with an increased CO_2 content (for example, $\gamma_1 = 1$) will lead to the earlier formation of a thermal lens and spreading of the beam. The effect of the processes under discussion on the propagation of a radiation pulse is illustrated by Fig. 5, in which the distribution of the relative amplitude A/A_0 over beam radius in the $z' = 1$ plane for weakly and strongly absorbing media ($\gamma_1 = 0.05$ and 1, curves 1, 2, respectively) at $\tau = 85.7$ μsec (solid curves) and $\tau = t_T$ (dashed curves) is shown; for comparison the initial distribution of the amplitude at $z' = 0$ is shown by the dot-dash curve. It is evident that due to the more rapid saturation of the absorbing transition at $\tau = 85.7$ μsec in a medium with $\gamma_1 = 1$ the amplitude on the beam axis ($r' = 0$) is higher than in a medium with $\gamma_1 = 0.05$, although the depth of kinetic cooling in a medium with $\gamma_1 = 1$ is less.

At $\tau = t_T$ the situation changes. Here on the contrary the maximum amplitude at $r' = 0$ is observed in a weakly absorbing medium. Strong heating of the medium with $\gamma_1 = 1$ leads to thermal defocusing and to a "displacement" of energy from the axis to the periphery of the beam.

Thus the computational analysis performed has shown that upon the propagation of a radiation pulse in gaseous resonantly absorbing media it is necessary to take account of both excitation of the different types of vibrations of the mixture molecules and the finite rate of thermalization of the absorbed energy as well as the saturation effect of the absorbing transition. Ignoring these effects can lead to significant errors in the determination of the amplitude–phase characteristics of a light beam.

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