RESONANT SELF-FOCUSING IN A CO_2-N_2 MIXTURE

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The resonant absorption of pulsed CO_2 laser radiation in a mixture of CO_2 and N_2 is accompanied by the kinetic cooling effect [1]. This effect has been studied experimentally [2-4] and theoretically [4, 5] in mixtures containing more than 1% carbon dioxide. The dependence of the temperature drop (decrement) and duration of the cooling effect on the parameters of the pulse and the medium has been investigated [4, 5] on the basis of the kinetic equations for the inventories of vibrational quanta in each mode. It has been shown [6] that the cooling decrement increases appreciably as the temperature of the mixture is raised to 500-600°K. The latter study was carried out in the approximation of a specified radiation field, which is valid over short paths.

On the other hand, it is clear that the increased density in the axial zone of the beam due to kinetic cooling tends to suppress diffraction spreading and can elicit self-focusing, which, in turn, affects the parameters of the medium. The present study is devoted to a numerical analysis of the self-consistent problem of the interaction of CO_2 laser radiation with a mixture of carbon dioxide and nitrogen. The characteristic parameters are consistent with laboratory experimental conditions [4]. The principal aim is to determine the conditions under which self-focusing due to kinetic cooling can be recorded in natural experiments.

1. The self-induced effect is analyzed in the quasioptical approximation [7] on the basis of the "parabolic" equation for the complex amplitude of the electric field $\mathscr{E}(r, z, t)$:

$$2ikn\left(\frac{\partial \mathscr{E}}{\partial z} + \frac{1}{v}\frac{\partial \mathscr{E}}{\partial t}\right) = \Delta_\perp \mathscr{E} + 2k^2\delta n\mathscr{E} - ika\mathscr{E}, \tag{1.1}$$

where $k = 2\pi/\lambda$ is the wave number, v is the group velocity of light, α and n are the absorption coefficient and refractive index of light in the medium, δn is the variation of the refractive index of the medium, and $\Delta_\perp = 1/r \cdot \partial/\partial r(r\partial/\partial r)$. The first term on the right-hand side of Eq. (1.1) describes diffraction, and the last term describes absorption in the medium. The absorption process is assumed to be linear, i.e., the optical brightening effect in the medium is disregarded. The self-induced effect created by variation of the refractive index of the medium in kinetic cooling is described by the second term on the right-side of (1.1). The variation of the refractive index is assumed to be proportional to the density perturbation $\delta n = (n_0 - 1)\delta\rho/\rho_0$. The relative variation of the density of the gas $\rho(r, z, t)$, in turn, obeys the wave equation

$$\frac{\partial^2 \rho}{\partial t^2} - c_0^2\Delta_\perp\rho = c_0^2\beta\Delta_\perp T, \tag{1.2}$$

in which β is the coefficient of (cubical) thermal expansion, c_0 is the velocity of sound, and $T(I_0, t)$ is the temperature source function. Equation (1.2) is written on the assumption that the longitudinal density gradients are negligible in comparison with the transverse gradients. The temperature source function is determined from the equation

$$\partial T/\partial t = \Phi(I_0, t)/(C_p\rho_0), \tag{1.3}$$

in which $\Phi(I_0, t)$ is the power density of the cooling (heating) sources in connection with energy transfer between the translational and vibrational degrees of freedom of the molecules, ρ_0 is the unperturbed density of the gas, and C_p is the heat capacity of translational and rotational degrees of freedom of the gas at constant pressure. It is assumed in the formulation of Eq. (1.3) that the influence of thermal conduction and forced convection is small in the time scales of interest here.

Thus, the characteristic transition times to fully developed convection τ_K and thermal conduction τ_T are defined as [5]

$$\tau_K \sim r(\alpha I_0 r^2 \beta g/\rho_0 C_p)^{-1/3}, \quad \tau_T \sim r^2/(4\chi).$$

For values of the parameters consistent with laboratory experimental conditions [4] $\tau_K \geqslant 10^{-3}$ sec, and $\tau_T \geqslant 0.1$ sec. These estimates refer to the case of instantaneous thermalization of absorbed energy $\Phi(I_0, t) = \alpha I_0$, where $I_0 = cn|\mathscr{E}_0|^2/8\pi$. In the investigated case of kinetic cooling $|\Phi(I_0, t)| < \alpha I_0$, because the energy transferred into molecular vibrational levels from the translational degrees of freedom of the gas is lower than the absorbed energy. Consequently, the estimate for τ_K is too low. On the other hand, the duration of kinetic cooling $\tau \leqslant 10^{-4}$ sec, which is at least an order of magnitude shorter than τ_K or τ_T, justifying the approximations used. The power density of the cooling sources $\Phi(I_0, t)$ is given by the relation

$$\Phi(I_0, t) = N\left[A(\varepsilon_2) E_{010} P(\varepsilon_2, \varepsilon_2^0) - (E_{001} - A(\varepsilon_2) E_{030}) W_3(\varepsilon_1, \varepsilon_2)\right]. \tag{1.4}$$

This quantity is the difference between the energy flux from translational degrees of freedom to the (01^10) level [first term on the right-hand side of (1.4)] and the energy flux released as heat in three-quantum decay of the (00^o1) level [second term in (1.4)]. Heating is a consequence of the energy deficit associated with the transition $(00^o1) \to (03^10)$. The notation in (1.4) corresponds to that used in [5]: N is the density of carbon dioxide molecules;

$$A(\varepsilon_2) = 2(1 + \varepsilon_2)/(2 + 6\varepsilon_2 + 3\varepsilon_2^2); \quad P(\varepsilon_2, \varepsilon_2^0) =$$
$$= P_{20}(\varepsilon_2 - \varepsilon_2^0)(1 - \exp(-E_{010}/T_0));$$
$$W_3(\varepsilon_1, \varepsilon_2) = \frac{P_3}{8}\left[\exp(-500/T_0)\varepsilon_2^3(1 + \varepsilon_1) - \varepsilon_1(\varepsilon_2 + 2)^3\right];$$
$$P_{20} = P_c K_1 + p_a K_2; \quad P_3 = p_c K_3 \text{ s } p_a K_4;$$

K_i denotes the kinetic coefficients; p_c and p_a are the partial pressures of the carbon dioxide and nitrogen in the mixture; P_{20} and P_3 have the significance of the reciprocal times of vibration—translation relaxation and three-quantum decay, respectively; ε_1 is the average number of quanta of the asymmetrical mode of CO_2 and vibrations of N_2; ε_2 is the same for the combined symmetrical and strain modes of CO_2 in mutual equilibrium; ε_2^0 is the equilibrium value of ε_2; and T_0 is the temperature of the gas. The inventories of vibrational quanta ε_1 and ε_2 satisfy the kinetic equations

$$\frac{d\varepsilon_1}{dt} = X\left[W_3(\varepsilon_1, \varepsilon_2) + E(I_0, \varepsilon_1, \varepsilon_2)\right]; \tag{1.5}$$

$$\frac{d\varepsilon_2}{dt} = A(\varepsilon_2)\left[-3W_3(\varepsilon_1, \varepsilon_2) - 2E(I_0, \varepsilon_1, \varepsilon_2) - P(\varepsilon_2, \varepsilon_2^0)\right]; \tag{1.6}$$

in which $X = p_c/(p_c + p_a)$; $E(I_0, \varepsilon_1, \varepsilon_2)$ is an optical pumping term [8]:

$$E(I_0, \varepsilon_1, \varepsilon_2) = (\sigma_0 I_0/\hbar\omega)\left[\varepsilon_2^2/(2 + \varepsilon_2)^2 - \varepsilon_1/(1 + \varepsilon_1)\right]16(1 + \varepsilon_2)/[(1 + \varepsilon_1)(2 + \varepsilon_2)^4],$$

$\hbar\omega = E_{001} - E_{100}$ is the optical transition energy, and σ_0 is the cross section for absorption of radiation by a CO_2 molecule.

Thus, the investigation of the self-induced effect under kinetic cooling conditions entails an analysis of the quasioptical equation (1.1) in conjunction with the equations describing the variation of the density (1.2), heat withdrawal (1.3), and the kinetic behavior of the inventories of vibrational quanta (1.5) and (1.6).

2. For numerical analysis it is practical in (1.1)-(1.6) to transform to dimensionless variables and functions $r' = r/a_0$, $z' = z/(ka_0^2)$, $\tau = (t - z/v)/t_0$, $\mathscr{E}' = \mathscr{E}/|\mathscr{E}_0|$, $\rho' = \rho/\rho^*$, $T' = T/T^*$, where a_0 is the initial beam radius, t_0 is the pulsewidth, $\mathscr{E}_0$ is a characteristic value of the field amplitude at the entrance to the medium, and ρ^* and T^* are the scales for measurement of the density and temperature source function. In the new variables we have[†]

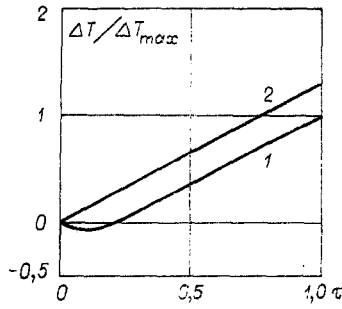[†]From now on we drop the primes from the dimensionless variables.

Fig. 1

$$i\frac{\partial \mathscr{E}}{\partial z} = \frac{1}{2}\Delta_\perp \mathscr{E} + R\rho\mathscr{E} - ia\mathscr{E}. \tag{2.1}$$

The nonlinearity parameter R is proportional to the pulse energy W and depends on the charac-
teristics of the medium:

$$R = W\frac{k^2\,(n_0-1)\,X}{\pi T_0}\frac{E_{010}}{\hbar\omega}\frac{\sigma_0 N_A}{(7/2)\,R_0}\frac{\varepsilon_1^0\left(1+\varepsilon_2^0\right)}{\left(1+\varepsilon_2^0/2\right)^4}.$$

where N_A is Avogadro's number, R_o is the universal gas constant, and $\alpha = \alpha_0 X k a_0^2/2$ is the re-
duced absorption. The wave equation takes the form

$$\partial^2\rho/\partial\tau^2 - c^2\Delta_\perp\rho = c^2\Delta_\perp T, \tag{2.2}$$

where $c = t_0 c_0/a_0$ is the acoustic parameter or ratio of the pulsewidth to the characteristic
equalization time of the pressure over the beam cross section. The density scale $\rho^* = \rho_0 j t_0\gamma$
and scale of the temperature source function $T^* = T_0 j t_0\gamma$, where (jt_0) has the significance of
the ratio of the energy transmitted across the absorption cross section of the CO_2 molecule
in time t_0 to the optical transition energy, $j = \sigma_0 I_0 (1 + \varepsilon_2^0)/[\hbar\omega(1 + \varepsilon_2^0/2)^4]$, $\gamma = NE_{010}\varepsilon_1^0/$
$(\rho_0 C_p T_0)$. With good accuracy, Eqs. (1.3)-(1.6) are linearized with respect to the deviations
of the inventories of vibrational quanta from their equilibrium values $\Delta\varepsilon_1 = \varepsilon_1 - \varepsilon_1^0$ and $\Delta\varepsilon_2 =$
$\varepsilon_2 - \varepsilon_2^0$. In dimensionless variables the linearized equations assume the form

$$\partial T/\partial\tau = \lambda_1 e_1 + \lambda_2 e_2; \tag{2.3}$$

$$\partial e_1/d\tau = X[v - e_1 t_0(\mu + j) + e_2 t_c\zeta(\xi + j)]; \tag{2.4}$$

$$de_2/d\tau = -2v + 2e_1 t_0(3\mu/2 + j) - 2e_2 t_0\eta(\theta + j), \tag{2.5}$$

where $e_i = \Delta\varepsilon_i/(\varepsilon_i^0 j t_0)$. The constant coefficients in Eqs. (2.3)-(2.5) depend in a complex
way on the molecular constants; in particular, for $T_0 = 500°K$ we have $\xi \sim 0.1P_3$, $\mu \sim 1.6P_3$,
and $\theta \sim 0.8P_3 + 8P_{20}$, where $\theta > \mu > \xi$. If the medium is far from saturation $(j \ll \xi)$, then
in the period of three-quantum decay of the $(00°1)$ level the energy transmitted across the
cross section σ_0 is much smaller than $\hbar\omega$. In this case the kinetic cooling decrement is
proportional to the pulse energy W. If the energy transmitted across σ_0 during the vi-
bration—translation relaxation time $\tau \sim 1/P_{20}$ is much greater than $\hbar\omega$ $(j \gg \theta)$, then the cool-
ing depth attains a maximum and any further increase in the intensity merely accelerates the
transition process.[†]

A typical time dependence of the reduced cooling decrement is given in Fig. 1. It has
been obtained from the solution of the system (2.3)-(2.5) in the approximation of a specified
intensity $I_0 = const$ (curve 1). Curve 2 corresponds to nonresonant absorption.

3. The system of equations (2.1)-(2.5) has been solved on the basis of a specially
developed set of programs. For the integration of the quasioptical equation (2.1) we use

[†]Different values are given in the literature for the kinetic relaxation constants in CO_2
at elevated temperatures. Here we use the data of [6]. If the constants from [10] are used,
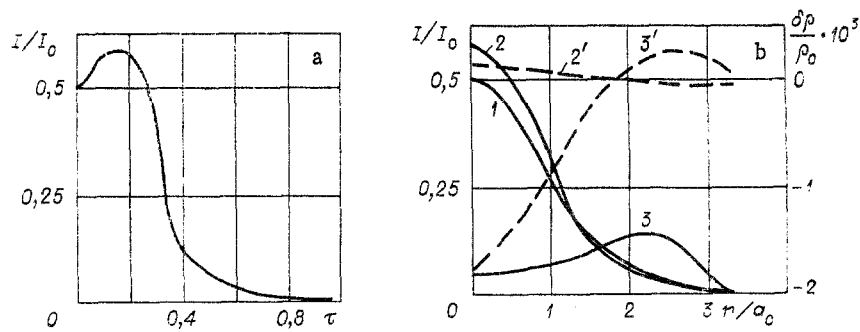the cooling depth is smaller by 2/3 to 1/2.

Fig. 2

the method of finite elements [9], and for the wave equation (2.2) the discrete Fourier—Bessel transform method. During the computational process the space—time distributions of the intensity for various values of z are printed out, along with the profiles of the density perturbations and temperature source function T.[†]

From the point of view of recording the self-focusing effect in a natural experiment an important factor is the relative increment of the intensity at the observation point $\Delta = (I(t^*, r, z) - I(0, r, z))/I(0, r, z)$. On the other hand, a suitable parameter for assessing the self-focusing efficiency is the intensity at the observation point, normalized to the input intensity: $I(t, r, z)/I(0, 0, 0)$. Both of these parameters are used in the present study, where Δ denotes the relative intensity increment on the beam axis. The time t^* corresponds to maximum self-focusing.

Typical results corresponding to room temperatures ($T_0 = 300°K$, $X = 0.5$, $t_0 = 90$ μsec, $\alpha = 0$, $R = 0.14$, value of $n_0 - 1$ taken from [11]) are given in Fig. 2. The initial conditions correspond to a time-square pulse with a Gaussian intensity profile $I(t, r, z) = I_0 \exp(-(r/a_0)^2)$ and a plane phase front. Figure 2a represents the time variation of the normalized intensity on the beam axis at a distance equal to the diffraction length from the entrance to the medium. It is seen that the leading edge is subjected to diffraction spreading (Fig. 2b, curve 1, $\tau = 0$).

At times t of the order of the cooling duration the intensity is observed to grow (curve 2, $\tau = 0.25$), and then in the course of transition from cooling to heating defocusing is observed (curve 3, $\tau = 0.5$). The radial distributions of the normalized density perturbation at a distance equal to the diffraction length are represented by dashed curves in Fig. 2b. Curves 2' and 3' correspond to times $\tau = 0.25$ and 0.5. The negative values of the density perturbations for curve 2' and the positive values for curve 3' at the periphery of the beam are attributable to hydrodynamic pressure-equalization effects. In the given example the relative intensity increment Δ at the diffraction length attains 20%.

As already mentioned, the kinetic cooling decrement increases at elevated temperatures. It is therefore sensible to consider the self-induced effect at a temperature of the mixture $T_0 = 500°K$. The characteristic parameter in the theory of transient self-focusing is the pulse energy [12]. Consequently, in the first series of numerical experiments we fix the pulse energy $W = 1$ J ($R = 0.25$) and vary the beam radius a_0. In order for the effect of diffraction to be identical within the entire series of experiments the observation point is situated at a distance $z = ka_0^2$ or $ka_0^2/2$. The pulsewidth is chosen to maximize the increment Δ at the time $2t_0/3$ (with allowance for hydrodynamic effects). The CO_2 concentration is selected with regard for the dependence of the cooling decrement ΔT on the CO_2 content of the mixture at a specified pulse energy and for the absorption of radiation in the medium, since absorption is significant at high temperatures.

The optimal concentration X is taken to be that at which the product of the cooling decrement and the radiation attenuation coefficient $M(X) = \Delta T \exp(-\alpha_0 Xz)$ attains a maximum.

[†]The accuracy of the calculations is tested according to the value of the total radiated power in the beam $P = 2\pi \int_0^\infty I(t, r, z)r\,dr$. The power decays with distance from the entrance to the medium in accordance with linear absorption. It has been shown that with halving of the integration step the numerical value of the solution varies within the limits of a few percent.
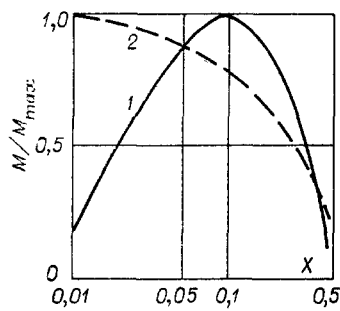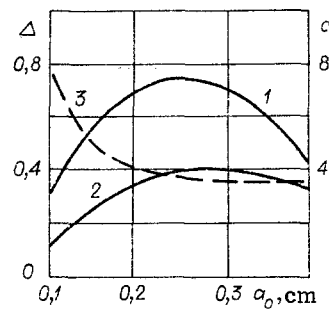
Fig. 3   Fig. 4

Here z is the observation distance. If the pulsewidth is made to the time to attain maximum cooling, then for a beam with $a_0 = 0.2$ cm and $W = 1$ J the function $M(X)$ has a maximum at $X = 0.1$ (Fig. 3, curve 1). Consequently, the numerical experiments of the first series are carried out for $X = 0.1$.

The dependence of Δ on the initial beam radius a_0 is shown in Fig. 4. The observation points are situated at the diffraction length (curve 1) and at half that length (curve 2). The variation of the acoustical number c within the experimental series is represented by the dashed curve 3. It is apparent from the graph that the dependence of the relative intensity increment Δ on a_0 has characteristic maxima. The reduction of the self-focusing for large radii a_0 is attributable to the influence of absorption, since the observations are made at large distances. Characteristically, the maximum of curve 2 in Fig. 4 is shifted toward larger values of a_0 in comparison with the maximum of curve 1. The decrease of Δ observed for small radii a_0 is explained by the fact that saturation effects begin to set in at high intensity. As a result, the increase in the diffraction spreading for small a_0 can no longer be compensated by the growth of the cooling decrement due to increased intensity.

The second series of numerical experiments is carried out for a fixed intensity I_0 and a fixed pulsewidth $t_0 = 50$ µsec. It is assumed that the observations are made at distances of the order of 1–4 m, which are consistent with natural experimental conditions. For a fixed radiation intensity the cooling decrement increases with a decrease in the content of CO_2 in the mixture (due to prolonging of the cooling period). Accordingly, the self-focusing effect must be more appreciable at low CO_2 concentrations. The function $M(X)$ in this case is represented by curve 2 in Fig. 3. However, the pulsewidth is limited by the structure of the radiation sources. We therefore choose a concentration $X = 0.05$. Figure 5 gives the normalized intensity on the beam axis I/I_0 as a function of z ($0 \leqslant z \leqslant 400$ cm) for two initial radii $a_0 = 0.25$ (curves 1 and 1') and $a_0 = 0.5$ cm (curves 2 and 2'). Curves 1 and 2 correspond to the initial time, and curves 1' and 2' to the time of maximum self-focusing $\tau = 0.4$. It is important to note that with an increase in the beam radius a_0 the pulse energy and, hence, the nonlinearity parameter R increase. For $a_0 = 0.25$ cm the nonlinearity parameter $R = 0.6$, and for $a_0 = 0.5$ cm we have $R = 2.4$. However, as is evident from Fig. 5, if the observation distance is shorter than 2 m, then the increase in I/I_0 is more appreciable for a beam of smaller radius. This result is attributable to the fact that for small beam radii nonlinear effects are felt at shorter distances. A typical plot of the self-focusing parameters Δ and I/I_0 as a function of the initial beam radius for a fixed value of the nonlinearity parameter $R = 0.5$ is given in Fig. 6. The observation distance is 1 m. The relative increment
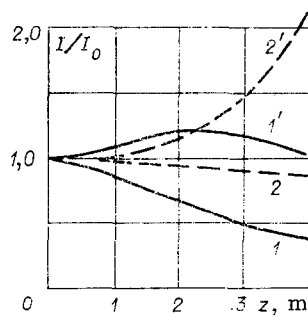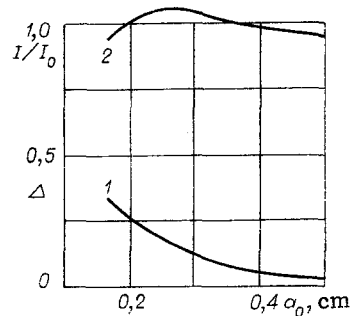

Fig. 5   Fig. 6

Δ decreases monotonically with increasing radius a_0, since the path length z/ka_0^2 decreases. This monotonic behavior is violated in the case of I/I_0 as a function of the initial radius (curve 2). Clearly, for narrow beams the increasing diffraction can no longer be compensated in the investigated self-focusing mechanism. We note that numerical experiments with beams having a "super-Gaussian" profile $I = I_0 \exp(-(r/a_0)^6)$ at distances $z = 0$–100 cm do not indicate any appreciable intensification of the self-focusing effect in comparison with Gaussian beams.

The self-focusing efficiency can be affected by the shape of the leading edge of the radiation pulse, because it has been shown [4] that the kinetic cooling decrement is greater in the case of a pulse with a steep leading edge. However, at times $t \leqslant P_3^{-1} + P_{20}^{-1}$ the kinetic cooling decrement is completely determined by the energy of the radiation transmitted through the medium [4]. Consequently, for pulses having a rise time much shorter than the kinetic cooling period in the medium the self-focusing efficiency will be determined mainly by the pulse energy W (i.e., by the value of the nonlinearity parameter R).

The numerical simulation results have shown that the transient self-induced thermal effect created by the kinetic cooling effect in a gas has a significant effect on the shape of the pulse that propagates in the medium. At high temperatures this effect can be appreciable at distances of the order of 1 m. It is therefore reasonable to anticipate the possibility of recording the given type of self-induced effect experimentally.

Experiments on the self-induced action of resonant radiation under kinetic cooling conditions can provide a criterion for the proper selection of the kinetic rate constants of vibrational relaxation in carbon dioxide molecules.

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