apparently condensation in gas jets. The complications occurring here have not been analyzed in detail in this paper. Let us just note that the existence of a domain of values characterizing the regime of the process of the parameter $p_0{}^{d*^{0.6}}$, where p_0 is the initial gas pressure in the jet, and d* is the nozzle diameter in which condensation is terminated by the formation of clusters with $N \sim 20-30$, for which noticeable separation can be expected from (2.17), while on the other hand the fraction of fluid far from the nozzle is almost a maximum, follows from the results in [5]. These results apparently carry over to our case as well with the difference that termination of condensation in the presence of condensation corresponds to the reduction of the effective supersaturation $\ln s' = 0$, i.e., sets in for finite supersaturations $\ln s = \mu_2 E \gamma B_0 / \overline{\Delta\varepsilon}^2 > 0$.

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THEORY OF THE KINETIC COOLING OF A CO_2–N_2 MIXTURE WITH A HIGH CO_2 CONTENT

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Resonance absorption by CO_2 molecules of radiation upon the intermodal transition (100)–(001) (or (020)–(001)) leads to nonstationary cooling of the gas [1], due mainly to the flow of energy from the translational degrees of freedom into the deformation mode of the CO_2 as a result of the process of V–T relaxation. An investigation of the effect of kinetic cooling of carbon dioxide under the action of radiation is of interest for problems in oscillation kinetics (determination of the relaxation time, analysis of the operation of molecular gas lasers) and in the propagation of radiation through gaseous media (autonomous thermal effect).

Theoretical studies have thus far been conducted on the phenomenon of kinetic cooling for atmospheric conditions (the case of low CO_2 concentrations) [2, 3]. Experimental investigations have been conducted on cooling with mixtures with partial CO_2 concentrations of $x_{CO_2} = 10^{-3}-1$ [4-7] at a gas temperature of $T_0 < 400°K$. The measured values of the kinetic-cooling parameters were in satisfactory agreement with the results of numerical calculations [7, 8] performed on the basis of a "thermodynamic" model [9]. In [10] a theoretical investigation was conducted on the channel of relaxation of the state of CO_2 (001) which made the main contribution to the variation of gas temperature.

In the present study we obtained analytic expressions for the parameters of the cooling of a mixture of gaseous CO_2 and N_2 over wide ranges of CO_2 partial concentrations and of intensities of the exciting radiation; we investigated the temperature dependence of the effect in the 200-1200°K range, which enabled us to establish the temperature region in which the phenomenon of kinetic cooling exists. We also considered the problem of optimal insertion of the energy of a light pulse for cooling a molecular system.

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1. Linearization of the Equations Describing the Kinetic-Cooling Effect. The variation in the temperature of a molecular gas for τ_m, $\tau_{rr} << t << \tau_{pe}, \tau_e$ — where τ_m, τ_{rr}, τ_{pe}, τ_e are the times for the establishment of a Maxwell distribution of molecular velocities, rotational relaxation, equalization of pressure in the medium during spatially uniform heating or cooling, and thermal conductivity, respectively — are determined by the energy flux $\Phi(t)$ from the oscillatory and translational degrees of freedom of the molecules, i.e., by the processes of oscillatory relaxation of the absorbed radiant energy:

$$\rho c_p^{RT} \frac{dT}{dt} = \Phi(t), \tag{1.1}$$

where ρ is the density of the gas and c_p^{RT} is the specific isobaric heat capacity of the translational and rotational degrees of freedom.

In order to describe the kinetic processes in the CO_2—N_2 mixture in this study, we used a "thermodynamic" model with an approximation of the modes by harmonic oscillators [8]. The rapid V—V' exchange between the antisymmetric mode of the CO_2 and the oscillations of the N_2 makes it possible to describe them in terms of the single variable ε_1, the reserve of oscillatory quanta in these combined modes; analogously, we introduced the quantity ε_2 for the symmetric and deformation modes of the CO_2. Then the kinetic equations have the form [9]

$$\frac{d\varepsilon_1}{dt} = (W_{3\Sigma} + E)\, x_{CO_2}; \tag{1.2}$$

$$\frac{d\varepsilon_2}{dt} = -A\,(3W_{3\Sigma} + P + 2E); \tag{1.3}$$

$$\Phi(t) = N_0 x_{CO_2} \left[A E_{010} P - (E_{001} - 3AE_{010})\, W_{3\Sigma}\right], \tag{1.4}$$

where

$$W_{3\Sigma} = \frac{p_{3\Sigma}}{8}\left[\exp\left(\frac{3E_{010} - E_{001}}{kT}\right)\varepsilon_2^3 (1 + \varepsilon_1) - \varepsilon_1 (2 + \varepsilon_2)^3\right];$$

$$E = \frac{16}{(2 + \varepsilon_2)^4} \frac{(1 + \varepsilon_2)}{(1 + \varepsilon_1)} \left[\frac{\varepsilon_2^2}{(2 + \varepsilon_2)^2} - \frac{\varepsilon_1}{1 + \varepsilon_1}\right] \frac{\sigma I}{\hbar \omega};$$

$$P = p_{20}\left(\varepsilon_2 - \varepsilon_2^0\right)\left(1 - \exp\left(-\frac{E_{010}}{kT}\right)\right); \quad A = \frac{2\,(1 + \varepsilon_2)}{2 + 6\varepsilon_2 + 3\varepsilon_2^2};$$

$p_{3\Sigma}$ is the probability of V—V' exchange between the antisymmetric and deformation modes of the CO_2 corresponding to three-quantum disintegration, p_{20}, rate of V—T relaxation of the deformation mode, σ, cross section of absorption of the radiation exciting the molecules, with intensity $I(t)$ at a frequency of $\omega = (E_{001} - E_{100})/\hbar$, N_0 number of molecules per unit volume, x_{CO_2}, partial concentration of CO_2 in the mixture, ε_2^0, equilibrium value of ε_2.

The equations of the "thermodynamic" model (1.1)-(1.4) satisfactorily describe the phenomenon of kinetic cooling of the CO_2—N_2 mixture under the action of radiation in the range $x_{CO_2} \gtrsim 0.001$ and $T \lesssim 1200°K$. Outside this range the situation no longer satisfies the model's fundamental assumption that a Boltzmann distribution of the molecules by levels is rapidly established in each of the modes of CO_2 and N_2.

The characteristic parameters of the variation of temperature with time in the effect under consideration are: ΔT_{max}, the maximum depth of cooling, and t_{max}, the time when this is attained, and t_c, the length of time the cooled state exists. In order to obtain analytic expressions for these quantities, we linearize Eqs. (1.1)-(1.4) for the conditions: ε_1, $\varepsilon_1^0 << 1$, $|\varepsilon_2 - \varepsilon_2^0| \ll \varepsilon_2^0$, $\Delta T \ll T_0$, where ΔT is the temperature variation during the time the cooling exists. Then in the time interval $t < t_c$, assuming that $p_{3\Sigma}(T)$, $p_{20}(T)$, $\sigma(T)$, $\varepsilon_2^0(T)$ have constant values taken at the equilibrium temperature T_0, and setting $\varepsilon_2^k = (\varepsilon_2^0)^{k-1}\varepsilon_2$, we obtain

$$\frac{d\varepsilon_1}{dt} = -\varepsilon_1\left(p'_{3\Sigma} + \gamma x_{CO_2}\right) + \varepsilon_2\left(\beta p'_{3\Sigma} + \gamma\delta x_{CO_2}\right); \tag{1.5}$$

$$\frac{d\varepsilon_2}{dt} = A\left[\varepsilon_1\left(\frac{3p'_{3\Sigma}}{x_{CO_2}} + 2\gamma\right) - \varepsilon_2\left(\frac{3p'_{3\Sigma}\beta}{x_{CO_2}} + 2\gamma\delta + p'_{20}\right) + p'_{20}\varepsilon_2^0\right]; \tag{1.6}$$

$$\rho c_p^{RT}\frac{dT}{dt} = N_0 x_{CO_2}\left[AE_{010}p'_{20}(\varepsilon_2 - \varepsilon_2^0) - \Delta E\frac{p'_{3\Sigma}}{x_{CO_2}}(\beta\varepsilon_2 - \varepsilon_1)\right], \tag{1.7}$$

$$p'_{3_\Sigma} = p_{3_\Sigma}\left(1 + \frac{\varepsilon_2^0}{2}\right)^3, \quad p'_{20} = p_{20}\left(1 - \exp\left(-\frac{E_{010}}{kT}\right)\right),$$

$$\gamma = \frac{\sigma I}{\hbar\omega}\frac{(1 + \varepsilon_2^0)}{\left(1 + \frac{1}{2}\varepsilon_2^0\right)^4}, \quad \Delta E = E_{001} - 3AE_{010},$$

$$\delta = \frac{\varepsilon_2^0}{(2 + \varepsilon_2^0)^2}, \quad \beta = \frac{(\varepsilon_2^0)^2}{(2 + \varepsilon_2^0)^3}\exp\left(-\frac{\Delta E}{kT}\right).$$

The comparison in [7] of the experimental data with the calculations from the initial equations (1.1)-(1.4) for room temperatures indicates that the selected model correctly describes the kinetic-cooling effect. This study considered a long light pulse ($I(t) = I_0$) over a wide temperature range T_0 = 300-1200°K; a numerical solution by the standard Kutta–Merson procedure was found for the initial system (1.1)-(1.4), and the general solution for the linearized equations (1.5)-(1.7) was tabulated.

In the numerical calculations relaxation-time values for T_0 = 300°K from [11] were used, with graphs of relaxation time versus temperature taken from [12]. The values of the radiation absorption coefficients for different temperatures were determined from the curves of [13].

2. Kinetic Cooling at Room Temperatures. At relatively low temperatures $T_0 \leqslant 350$°K the additional condition $\varepsilon_2^0 \ll 1$ is satisfied. The following relations then hold:

$$p'_{3_\Sigma} \simeq p_{3_\Sigma}, \; p'_{20} \simeq p_{20}, \; \delta \simeq \frac{\varepsilon_2^0}{4}, \; \beta \simeq \frac{(\varepsilon_2^0)^2}{8}\exp\left(-\frac{\Delta E}{kT}\right), \tag{2.1}$$
$$A \simeq 1, \; \gamma \simeq \frac{\sigma I}{\hbar\omega},$$

simplifying the linearized system of equations (1.5)-(1.7). Furthermore, for some limiting cases (with respect to T_0, I, x_{CO_2}) it is possible to introduce the small parameter $\mu \ll 1$, which describes the ratio of the probabilities of oscillatory relaxation ($p_{3\Sigma}$, p_{20}) and optical excitation (γ). Then by decomposing the general solution with respect to the parameter μ and retaining the terms of the first approximation in each of its components in the expression for $\Delta T(t) = T(t) - T_0$, we obtain simple formulas for the parameters of the kinetic cooling. Thus, in the saturation regime which is realized for $\gamma \gg p_{3\Sigma}/x_{CO_2}$, p_{20}/ε_2^0, we obtain the following equation for $\Delta T(t)$, the depth of cooling as a function of time:

$$\Delta T(t) = \frac{N_0 E_{010}\varepsilon_2^{0^2}}{2\rho c_p^{RT}}\left[-\left(1 + \frac{p_{3_\Sigma}}{2p_{20}}\right)\left(1 - \exp\left(-\frac{p_{20}t}{1 + \frac{\varepsilon_2^0}{2x_{CO_2}}}\right)\right) + \right. \tag{2.2}$$
$$\left. + \left(1 + \frac{\varepsilon_2^0}{2x_{CO_2}}\right)^{-2}\left(1 - \frac{\Delta E}{E_{010}}\frac{p_{3\Sigma}}{2p_{20}}\right)\frac{p_{20}}{\gamma x_{CO_2}}\left(1 - \exp\left(-\gamma x_{CO_2}\left(1 + \frac{\varepsilon_2^0}{2x_{CO_2}}\right)t\right)\right) + \left(1 + \frac{\Delta E}{E_{010}}\right)\frac{p_{3\Sigma}t}{2}\right],$$

and hence for the maximum depth of cooling and the time when it is attained, we have

$$\Delta T_{\max} \simeq \frac{N_0 E_{010}\varepsilon_2^{0^2}}{2\rho c_p^{RT}}\left[-\left(1 + \frac{p_{3\Sigma}}{2p_{20}}\right) + \left(1 + \frac{\Delta E}{E_{010}}\right)\left(1 + \frac{\varepsilon_2^0}{2x_{CO_2}}\right)\frac{p_{3\Sigma}}{2p_{20}}\left(1 + \ln\frac{1 + \frac{2p_{20}}{p_{3\Sigma}}}{\left(1 + \frac{\Delta E}{E_{010}}\right)\left(1 + \frac{\varepsilon_2^0}{2x_{CO_2}}\right)}\right)\right]; \tag{2.3}$$

$$t_{\max} \simeq \frac{1+\frac{\varepsilon_2^0}{2x_{CO_2}}}{p_{20}} \ln \left[\frac{1+\frac{2p_{20}}{p_{3\Sigma}}}{\left(1+\frac{\Delta E}{E_{010}}\right)\left(1+\frac{\varepsilon_2^0}{2x_{CO_2}}\right)} \right]. \tag{2.4}$$

The time the cooling exists is determined in this case from the transcendental equation

$$1-\exp\left(-\frac{p_{20}t}{1+\frac{\varepsilon_2^0}{2x_{CO_2}}}\right) \simeq \frac{1+\frac{\Delta E}{E_{010}}}{1+\frac{p_{3\Sigma}}{2p_{20}}} \frac{p_{3\Sigma}t}{2}. \tag{2.5}$$

In the low-intensity regime $\gamma \ll p_{3\Sigma}/x_{CO_2}$, p_{20}/ε_2^0, in a manner analogous to (2.2), we have

$$\Delta T(t) \simeq \frac{N_0 x_{CO_2} E_{010} \varepsilon_2^{0^2}}{2\rho c_p^{RT}} \frac{\gamma}{p_{3\Sigma}} \left[-\left(\frac{3}{2\left(1-\frac{p_{3\Sigma}}{p_{20}}\right)} + \frac{\Delta E}{2E_{010}} \right)\left(1-e^{-p_{3\Sigma}t}\right) + \right.$$

$$\left. + \frac{p_{3\Sigma}}{p_{20}} \frac{\left(1+\frac{p_{3\Sigma}}{2p_{20}}\right)}{\left(1-\frac{p_{3\Sigma}}{p_{20}}\right)} \left(1-e^{-p_{20}t}\right) + \left(1+\frac{\Delta E}{E_{010}}\right)\frac{p_{3\Sigma}t}{2} \right]. \tag{2.6}$$

The simplest formulas for the cooling parameters are obtained from (2.6) for $x_{CO_2} \leqslant 0.1$ where $p_{3\Sigma} \ll p_{20}$:

$$\Delta T_{\max} \simeq \frac{N_0 x_{CO_2} E_{010} \varepsilon_2^{0^2}}{4\rho c_p^{RT}} \frac{\gamma}{p_{3\Sigma}} \left[-\left(3+\frac{\Delta E}{E_{010}}\right) + \left(1+\frac{\Delta E}{E_{010}}\right)\left(1+\ln\frac{3+\frac{\Delta E}{E_{010}}}{1+\frac{\Delta E}{E_{010}}}\right) \right],$$

$$t_{\max} \simeq \frac{1}{p_{3\Sigma}} \ln \frac{3+\frac{\Delta E}{E_{010}}}{1+\frac{\Delta E}{E_{010}}}.$$

The time t_c is determined from the equation $1-e^{-p_{3\Sigma}t} \simeq \frac{1+\frac{\Delta E}{E_{010}}}{3+\frac{\Delta E}{E_{010}}} p_{3\Sigma}t$.

1. The variation of the cooling depth $\Delta T_{\max}$ and the time $t_{\max}$ as functions of the carbon dioxide concentration in the mixture are shown in Figs. 1 and 2. Here the curves obtained from the solution of the linearized equations (1.5)-(1.7) for the saturation intensities $I = I_s$, $I = 10^{-2} I_s$ and $I = 10^{-3} I_s$ (curves 1, 2, and 3, respectively) are compared with the curves obtained from the solution of the initial equations (1.1)-(1.4) for $I = I_s$ (curves 4) and from formulas (2.3), (2.4) (curves 5). The curves obtained from the linearized equations (1.5)-(1.7), taking account of the "low-temperature" approximation (2.1), practically coincide (to within 1-3%) with the analytic results obtained by formulas (2.3), (2.4). Thus, the principal error in the transition from the nonlinear equations (1.1)-(1.4) to formulas (2.2), (2.3) is introduced by the "low-temperature" approximation. A similar pattern was observed for all the other curves shown in Figs. 1-3.

From Fig. 1 it can be seen that as x_{CO_2} decreases, the depth of cooling increases, departing from the stationary value which depends on the radiation intensity. The reason for this is that the nitrogen molecules are effectively exchanging energy with the $CO_2(001)$, but the state $N_2(v = 1)$ has no rapid channels of deactivation. Therefore increasing the N_2 concentration in the mixture leads to a slowing-down of the three-quantum disintegration ($p_{3\Sigma}$) which determines the heating of the gas. The rate of cooling is determined by the lower of the two probabilities p_{20} and γ.

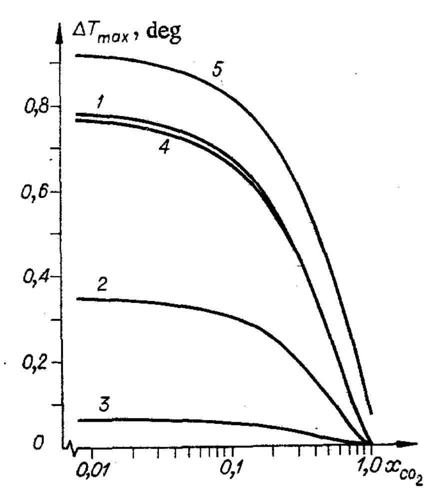

Fig. 1

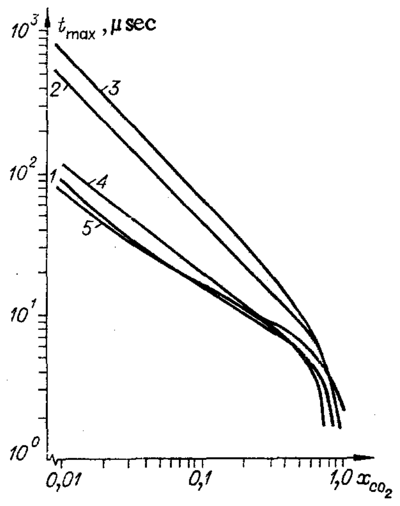

Fig. 2

The time of existence and time of attainment of maximum cooling are considerably increased as x_{CO_2} decreases. The time t_c is determined chiefly by the rate of heating of the mixture ($p_{3\Sigma}$), which is usually the slowest of the relaxation processes under consideration. For example, when $x_{CO_2} \lessapprox 0.1$, we find from Eq. (2.5) that $t_c \approx 2/p_{3\Sigma}(1 + \Delta E/E_{010})^{-1}$. The time t_{max} in the saturation regime is proportional to the time of V–T relaxation (p_{20}^{-1}) required for restoring an equilibrium population of the $CO_2(100)$ level for the same population of $CO_2(001)$ because of saturation of the transition (001–100). Further excitation of the molecules from the ground state stops, i.e., the cooling process ends. For low intensities the probability γ determines, on the one hand, the rate of cooling, since $\gamma \ll p_{20}/\varepsilon_2^0$, and on the other hand, the variation of the number of molecules in the antisymmetric mode of CO_2. Since $\gamma \ll p_{3\Sigma}/x_{CO_2}$, before the cooling process is completed, a substantial contribution to the temperature variation is made by the heating resulting from three-quantum disintegration, and therefore in the low-intensity regime $t_{max} \sim p_{3\Sigma}^{-1}$.

2. The variation of the depth of cooling as a function of the intensity of the exciting oscillation is characterized by a linear segment (2.6) when $\gamma \ll p_{3\Sigma}/x_{CO_2}$, p_{20}/ε_2^0 and by the establishment of a stationary level in the saturation regime ($\gamma \gg p_{3\Sigma}/x_{CO_2}$, p_{20}/ε_2^0). A study of the regime of very low intensities on the basis of Eqs. (1.1)-(1.4) showed that the kinetic-cooling phenomenon has no threshold with respect to I.

As the intensity of the exciting radiation increases, the characteristic times of kinetic cooling decrease, but the changes in t_c are small, since in all regimes we have $t_c \sim p_{3\Sigma}^{-1}$, and the decrease of t_{max} is more noticeable (see Fig. 2) especially for small values of x_{CO_2}. Since mixtures with a low CO_2 content are of great interest, we found an analytic solution for $x_{CO_2} \approx 0.01$ for an arbitrary value of radiation intensity:

$$\Delta T(t) \simeq \frac{N_0 E_{010} \varepsilon_2^{0^2}}{2\rho c_p^{RT}} \left(1 + \frac{p_{3\Sigma}}{\gamma x_{CO_2}}\right)^{-1} \left\{ -\left[\frac{3 + \frac{\Delta E}{E_{010}}}{2\left(1 + \frac{\gamma x_{CO_2}}{p_{3\Sigma}}\right)} + \frac{1}{1 + \frac{p_{3\Sigma}}{\gamma x_{CO_2}}} \right] \times \right.$$

$$\left. \times \left(1 - \exp\left(-p_{3\Sigma} t \frac{1 + \frac{\gamma x_{CO_2}}{p_{3\Sigma}}}{1 + \frac{\gamma \varepsilon_2^0}{2p_{20}}}\right)\right) + \frac{p_{3\Sigma}}{p_{20}} \frac{1 + \frac{\gamma x_{CO_2}}{p_{3\Sigma}}}{\left(1 + \frac{\gamma \varepsilon_2^0}{2p_{20}}\right)^2} \left(1 - \exp\left(-p_{20} t \left(1 + \frac{\gamma \varepsilon_2^0}{2p_{20}}\right)\right)\right) + \left(1 + \frac{\Delta E}{E_{010}}\right) \frac{p_{3\Sigma} t}{2} \right\}. \tag{2.7}$$

This formula was obtained by joining together the solutions for four intensity regions: $\gamma \ll p_{3\Sigma}/x_{CO_2} \sim p_{20}$, $\gamma \sim p_{3\Sigma}/x_{CO_2} \sim p_{20}$, $\gamma \gg \frac{p_{3\Sigma}}{x_{CO_2}} \sim p_{20}$, $\gamma \gg \frac{p_{20}}{\varepsilon_2^0} \gg \frac{p_{3\Sigma}}{x_{CO_2}}$. Figure 3 shows the variation of the depth of cooling as a function of intensity, obtained for $x_{CO_2} = 0.01$ from Eqs. (1.1)-(1.4).

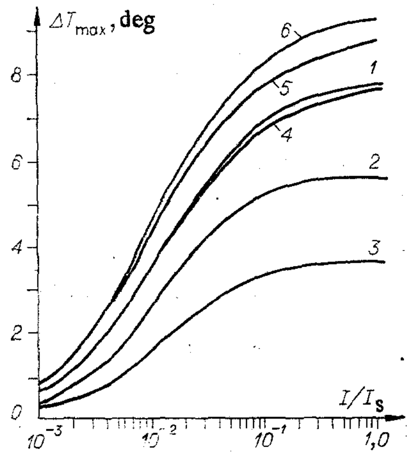

Fig. 3

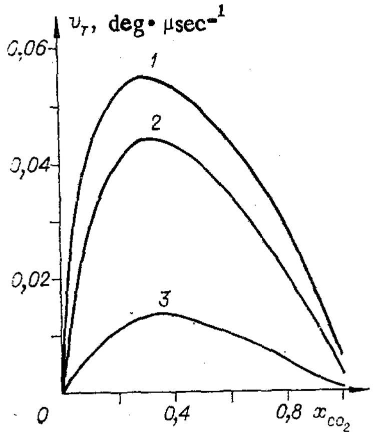

Fig. 4

(curve 4) and formula (2.7) (curve 5), and also from a solution from the linearized equations for x_{CO_2} = 0.01, 0.2, 0.4 (curves 1, 2, and 3), and for the same equations but taking account of the approximation (2.1) (curve 6) for x_{CO_2} = 0.01.

A special feature of mixtures with low CO_2 content is the considerable difference between the values of the rates of the relaxation processes $p_3 \ll p_{20}$, and therefore there may be an "intermediate" saturation regime, considered in detail for the atmosphere in [3]. In this case $(2p_{20}/\varepsilon_2^0 \gg \gamma \gg p_3\Sigma/x_{CO_2}$, which corresponds to $I \simeq 2 \cdot 10^5$ W/cm^2), it follows from (2.7) that

$$\Delta T(t) \simeq \frac{N_0 E_{010} \varepsilon_2^{0^2}}{2\rho c_p^{RT}} \left[-\left(1 - e^{-\gamma x_{CO_2} t}\right) + \frac{\gamma x_{CO_2}}{p_{20}} \left(1 - e^{-p_{20} t}\right) + \left(1 + \frac{\Delta E}{E_{010}}\right) \frac{p_{3\Sigma} t}{2}\right], \tag{2.8}$$

$$\Delta T_{\max} \simeq \frac{N_0 E_{010} \varepsilon_2^{0^2}}{2\rho c_p^{RT}} \left[-1 + \frac{p_{3\Sigma}}{2\gamma x_{CO_2}} \left(1 + \frac{\Delta E}{E_{010}}\right) \left(1 + \ln \frac{2\gamma x_{CO_2}}{p_{3\Sigma}\left(1 + \frac{\Delta E}{E_{010}}\right)}\right)\right].$$

Formula (2.8) is transformed into the corresponding formula of [3] if we make the substitutions $p_3\Sigma \to P_{30} + \delta P_{20}$, $p_{20} \to (2g/(1+g))P_{10}$, $x_{CO_2} \to \delta$, $2E_{010} = E_1$, $E_{101} + \Delta E = E_2 - E_1$, where P_{10}, P_{20}, P_{30} are the probabilities of collision deactivation of the levels $CO_2(010)$, $CO_2(001)$ and $N_2(v = 1)$, respectively, δ is the ratio of the partial concentrations of CO_2 and N_2, g is the statistical weight of the level $CO_2(020)$ [3]. Thus, the "thermodynamic" model on the boundary of its domain of applicability yields results which agree with the level-by-level consideration of the kinetic-cooling effect. We note that as a result of the greater increase in the probability of three-quantum disintegration ($p_3\Sigma$) with gas temperature in comparison with the rate of the V–T relaxation (p_{20}), the condition $2p_{20}/\varepsilon_2^0 \gg \gamma \gg p_3\Sigma/x_{CO_2}$ is violated, and for $T_0 \gtrsim 600$°K the "intermediate saturation" regime ceases to exist.

A study of the variation of the cooling parameters as functions of x_{CO_2} and I showed that the greatest depth of cooling of the CO_2–N_2 mixture at T_0 = 300°K is is attained in the saturation regime in mixtures with a low content of $CO_2(x_{CO_2} \leqslant 0.01)$ and amounts to $\Delta T_{\max} \simeq$ 0.8 °K where the durations of the exciting light pulse must be no less than $t_{\max}(x_{CO_2} = 0.01, I = I_s) \sim 100$ μsec.

3. The effect of the duration of the light pulse on the cooling of the gas may be considered next. All of the above results were obtained for $I(t) = I_0 \begin{cases} 1, & t > 0, \\ 0, & t < 0. \end{cases}$ calculations for a light pulse of finite duration $\left(I(t) = I_0 \begin{cases} 1, & 0 < t < \tau_p, \\ 0, & t < 0 > \tau_p \end{cases}\right)$ showed that the part for $\tau_p > t_{\max}(x_{CO_2}, I)$ (where $t_{\max}$ is the time required for attaining maximum depth in a long pulse) does not lead to additional cooling, and for $\tau_p < t_{\max}$ the depth of cooling is less than in a long radiation pulse. Therefore, in order to obtain maximum cooling for a given mixture without unnecessary energy losses, we must have $\tau_p \simeq t_{\max}$.

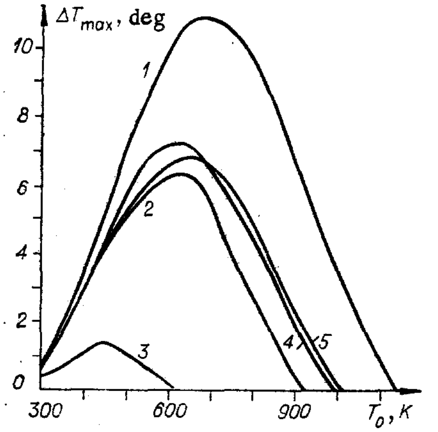

Fig. 5

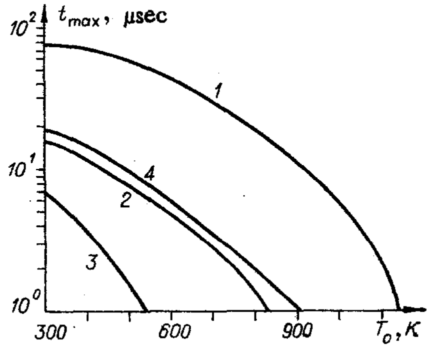

Fig. 6

Figure 4 shows the variation of $v_T = \Delta T_{max}/t_{max}$ as a function of the CO_2 concentration for various intensities of the exciting radiation $I = I_s$, $10^{-1}I_s$, and 10^{-2} I_s (curves 1, 2, and 3, respectively). In the first place, the quantity v_T can be treated as the average rate of cooling of the mixture by a radiation pulse of duration $\tau_p < t_{max}$ (x_{CO_2}, I). Then from the behavior of the curves in Fig. 4 we can conclude that the variation of the depth of cooling as a function of x_{CO_2} for a short light pulse of duration $\tau_p \leqslant t_{max}(x^*_{CO_2}, I_s)$ ($x^*_{CO_2} \simeq 0.3$ corresponds to the abscissas of the maxima of the curves for $v_T(x_{CO_2})$) and arbitrary shape as a function of time will have a form analogous to the function $v_T(x_{CO_2})$. This is confirmed by the results of the experiments in [7] and the numerical calculations for $I(t) = I_0 e^{-t/\tau_p}$ and $I(t) = \begin{cases} 1, & 0<t<\tau_p, \\ 0, & t<0,\ t>\tau_p, \end{cases}$ where τ_p satisfies the inequality $\tau_p \leqslant t_{max}$ ($x^*_{CO_2}$, I_s). As the duration of the exciting square pulse increases ($\tau_p > t_{max}(x_{CO_2}, I_s)$, the maximum depth of cooling will be attained in the mixture with lower x_{CO_2}, and the value of this concentration can be found from the equation $\tau_p \simeq t_{max}(x_{CO_2})$. In the second place, the value of v_T can be interpreted as the coefficient of utilization of the light energy of a square pulse of duration $\tau_p = t_{max}(x_{CO_2})$ for cooling the mixture. Then it follows from Fig. 4 that the energy losses to obtain a fixed amount of kinetic cooling are minimal in a mixture with $x_{CO_2} = x^*_{CO_2}$ and low radiation intensities ($I \leqslant 10^4$ W/cm²). It should be borne in mind that the greatest depth of cooling is obtained in a mixture with $x_{CO_2} \leqslant 0.01$ in the saturation regime, but because of the value of $I_s t_{max}(I_s\ x_{CO_2} = 0.01)$ the energy of the light pulse is high, and because there is little absorption of the radiation by a mixture with low CO_2 concentrations, this energy is not utilized effectively. A compromise case in which we obtain significant depths of cooling, for example 50% of $\Delta T_{max}(I_s, x_{CO_2} = 0.01) \simeq 0.8$K, for low energies in a light pulse of duration $\tau_p = t_{max}(x_{CO_2})$ is realized when $x_{CO_2} \sim 0.1$ and $I \sim 10^5$ W/cm².

3. Effect of Initial Temperature of the CO_2–N_2 Mixture on Kinetic Cooling. A study of the effect for $T_0 = 300$°K showed that the depth of cooling is proportional to $\varepsilon_2^{0\,2}$, which, to within a constant coefficient, is equal to the population of the low $CO_2(100)$ level of the transition absorbing a 10.6-μm radiation. Therefore, an increase in the population of this level when the temperature of the gas mixture increases must lead to an increase in the depth of cooling under the action of the radiation.

On the basis of the initial equations (1.1)-(1.4) and the linearized equations (1.5)-(1.7), we conducted a numerical study of the variation of the kinetic-cooling parameters as functions of temperature. For a saturation regime ($\gamma \gg p_3'\Sigma/x_{CO_2}$, p_{20}'/ε_2^0) we obtained an analytic solution

$$\Delta T(t) = \frac{N_0 x_{CO_2} E_{010} \varepsilon_2^0}{\rho c_p^{RT}} \left[-\frac{(1+gf)(1+n)f}{(1-f)^2 n} \left(1 - e^{-Ap'_{20}\frac{1-f}{n+f}nt}\right) + \right. \\ \left. + \frac{fn(1-gn)Ap'_{20}}{(f+n)^2 \gamma x_{CO_2}} \left(1 - e^{-\gamma x_{CO_2}\frac{n+f}{n}t}\right) + \frac{(1+g)f}{1-f} Ap'_{20} t \right]; \tag{3.1}$$

$$\Delta T_{\max} \simeq \frac{N_0 x_{CO_2} E_{010} \varepsilon_2^0}{\rho c_p^{RT}} \frac{(n+f)}{n(1-f)^2} \left\{ -\frac{(1+gf)(1+n)f}{n+f} + (1+g)f\left[1 + \ln\frac{(1+gf)(1+n)}{(1+g)(f+n)}\right]\right\}; \tag{3.2}$$

$$t_{\max} \simeq \frac{(n+f)}{n(1-f)Ap'_{20}} \ln\frac{(1+gf)(1+n)}{(1+g)(f+n)}, \tag{3.3}$$

where

$$f = \frac{p'_{3\Sigma}\delta}{p'_{20}x_{CO_2}}; \quad n = \frac{p'_{3\Sigma}}{2Ap'_{20}}; \quad g = \frac{\Delta E}{\Delta E_{010}}.$$

Figures 5 and 6 show the variation with temperature of the maximum depth of cooling and the time required to attain this depth in the saturation regime, curves obtained for $x_{CO_2} = 0.1$ from the initial equations (1.1)-(1.4) (curve 4) and from formulas (3.2), (3.3) (curve 5), and from the linearized equations (1.5)-(1.7) for $x_{CO_2} = 0.01$, 0.1, and 0.4 (curves, 1, 2, and 3, respectively).

This behavior of the depth of cooling can be explained by the fact that as the temperature increases, on the one hand, there is an increase in the reserve of oscillatory quanta ε_2^0, which leads to an increase in the cooling, and on the other hand, the rate of the V–T relaxation increases more slowly than the rate of three-quantum disintegration, which causes heating. As a result of this, the kinetic cooling has a maximum in its variation with temperature, and there is limit temperature above which the effect does not occur. Necessary conditions for the existence of the kinetic-cooling phenomenon can be obtained from the fact that the expression (3.3) is positive for $t_{\max}$ and the exponent of the first term in (3.1) is negative. These conditions take the form

$$\left(\frac{p'_{3\Sigma}}{p'_{20}} - \frac{2\Delta E_{010}}{\Delta E}\right) < 0, \quad \left(\frac{p'_{3\Sigma}}{p'_{20}} - \frac{x_{CO_2}}{\delta}\right) < 0. \tag{3.4}$$

For mixtures with high CO_2 content, with an upper limit temperature, the first of the formulas (3.4) changes sign; this is the formula that characterizes the ratio of the energy fluxes that lead to the heating and cooling of the gases. The temperature dependence of the probabilities of relaxation $p_{3\Sigma}$ and p_{20} shows that there is a limit to the effect on the low-temperature side as well. The values of the time required to attain maximum cooling and the duration of this cooling decrease monotonically as temperature increases, as do the oscillatory relaxation times $p_{3\Sigma}^{-1}$ and p_{20}^{-1}, which increases the effectiveness of utilization of the light energy for kinetic cooling.

It should also be noted that for temperatures T ≥ 700°K the probability p_{32} of two-quantum disintegration of the state $CO_2(001)$ in pure carbon dioxide becomes commensurable with the probability $p_{3\Sigma}$ [14]. However, at present there appear to be no direct experiments on the separate measurement of the probabilities p_{32}, $p_{3\Sigma}$ in CO_2–N_2 mixtures, so that we cannot rigorously take account of the effect of two-quantum disintegration in the problem under consideration. Therefore, confining our attention to the situation in which $p_{3\Sigma} = p_{32}$, we calculated the cooling parameters. It was found that when the depth of cooling varied with temperature (see Fig. 5), the value of the maxima decreased by no more than 10%. In this case the quantity ΔE in formulas (3.1)-(3.4) must be replaced by the expression

$$\Delta E\left(1 + \frac{E_{001} - 2\Delta E_{010}}{\Delta E}\frac{p_{32}}{p_{3\Sigma}}\right).$$

As a result of our calculations of the variation of the effect with temperature, we found that in principle the greatest depth of cooling in a CO_2–N_2 mixture at a total pressure of 1 atm is attained in a saturation regime with $x_{CO_2} \leqslant 0.01$ and temperature to ≈ 700°K and has a value of $\Delta T_{\max} \simeq 10°K$. Furthermore, we showed that the phenomenon of kinetic cooling can exist only in a finite temperature range.

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