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RESONANCE RADIATION COOLING OF A DIATOMIC
MOLECULE GAS FLUX

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The possibility of cooling a molecular gas of diatomic dipole molecules during resonance radiation absorption in the P-branch of the vibrational-rotational band was first examined in [1]. In this case the diminution in temperature of the medium is due to the appearance of an energy flux from the molecule translational degrees of freedom to the rotational because of R–T processes, and then to the vibrational. The lifetime of the effect under the action of a radiation pulse on a fixed gas is determined by the vibrational—translational (V–T) relaxation time or by the time of the intramodal vibrational—vibrational (V–V) exchange [1, 2].

The action of continuous resonance radiation on a medium consisting of diatomic dipole molecules and moving at a given velocity can, as will be shown below, also result in a change in the translational temperature and other macroscopic flux parameters. Their change will here be observed during the whole time of exposure. This paper is devoted to an investigation of the flow features of a mixture containing a diatomic dipole molecule gas in a resonance radiation field.

Let us consider the motion of an inviscid, non-heat-conducting gas in a constant-section channel. At a certain part of this channel let continuous radiation at the frequency $\nu_I = (E_{V''} + E_{j''} - E_{V'} - E_{j'})/h$ act on the gas, where h is Planck's constant, while $E_{V''}$, $E_{j''}$ and $E_{V'}$, $E_{j'}$ are the vibrational V and rotational j energies of the upper and lower levels of the absorbing transition $[(V', j') \rightarrow (V'', j'')]$, respectively.

The equations describing changes in the macroscopic parameters of such a medium have the form

$$\frac{d}{dx}(\rho u) = 0, \quad \frac{d}{dx}(\rho u^2 + p) = 0, \quad p = \frac{\rho R T}{\mu},$$

$$\frac{d}{dx}\left[\frac{5}{2}\frac{R}{\mu}T + \frac{u^2}{2}\right] = \frac{k_\nu I}{\rho u} - \sum_{i=1}^{m}\frac{de_V^i}{dx} - \sum_{i=1}^{m}\frac{de_r^i}{dx},$$

$(1)$

where $\rho$, $p$, $T$, $u$ are the gas density, pressure, translational temperature, and velocity, $\mu$ is the mixture molecular weight, $k_\nu$ is the spectral absorption index, I is the intensity of the acting radiation, R is the universal gas constant, and $e_V^i$ and $e_r^i$ are the vibrational and rotational energies per unit mass of the i-th mixture component.

Let the time of the stimulated radiation transition be $\tau_I \gg \tau_{R-T}$, $\tau_{V-V}$, where $\tau_{R-T}$ and $\tau_{V-V}$ are the characteristic times of the rotational-translational relaxation and the intramodal vibrational—vibration exchange, respectively. In this case the kinetics of the processes can be examined by using the harmonic oscillator approximation and the local vibrational temperatures model, while the molecule distribution over the rotational levels can be considered Boltzmannian with the translational temperature.

We represent the equations for the changes in the vibrational and rotational energy here in the form

$$\frac{de_V^i}{dx} = \left[\gamma_i \frac{R}{\mu}\frac{h\nu_i}{K_B}f_i + l_i\frac{k_\nu I}{\rho \nu_I}\nu_i\right]\frac{1}{u}, \quad \frac{de_r^i}{dx} = \frac{R}{u\mu}\frac{dT}{dx}\gamma_i,$$

$(2)$

where $\gamma_i$ is the molar fraction of the i-th component in the mixture, $\nu_i$ is the normal vibration frequency for the i-th component (it is assumed that the gas consists of only diatomic molecules), $l_i$ is the number of vibrational quanta acquired by the mode under the stimulated radiational transition, $K_B$ is Boltzmann's constant, m is the number of molecular components in the mixture, $f_i$ is the term responsible for the V–T relaxation and intermodal V–V' exchange. In this case the equations of the system (1) and (2) can be reduced to the form

$$\frac{dT}{dx} = \frac{1 - \varkappa M^2}{(1 - M^2)C_p}J, \quad \frac{du}{dx} = \frac{(\varkappa - 1)M^2}{(1 - M^2)u}J,$$

$(3)$

$$\frac{dp}{dx} = -\frac{\rho(\varkappa - 1)M^2}{(1 - M^2)}J, \quad \frac{d\rho}{dx} = -\frac{\rho(\varkappa - 1)}{a^2(1 - M^2)}J,$$

$$J = \frac{k_\nu I}{\rho u}\left(1 - \frac{\nu_k}{\nu_I}l_k\right) - \frac{Rh}{\mu K_B u}\sum_{i=1}^{m}\nu_i f_i \gamma_i,$$

$$M = \frac{u}{a}, \quad a = \sqrt{\varkappa\frac{R}{\mu}T}, \quad \varkappa = 1 + \left(\frac{3}{2} + \sum_{i=1}^{m}\gamma_i\right)^{-1},$$

$$C_p = \left(\frac{5}{2} + \sum_{i=1}^{m}\gamma_i\right)\frac{R}{\mu}.$$

Here i = 1, ..., k, ..., m and all $l_i = 0$ with the exception of $l_k$.

It is seen from the relationships represented that if J < 0 then for $0 < M < 1/\sqrt{\varkappa}$ the gas will be cooled and retarded, for $1/\sqrt{\varkappa} < M < 1$ it will be heated and retarded, while for M > 1 it will be cooled and accelerated. If J > 0 then, conversely, for $0 < M < 1/\sqrt{\varkappa}$ the gas will be heated and accelerated, for $1/\sqrt{\varkappa} < M < 1$ it will be cooled and accelerated, and for M > 1 heated and retarded.

For J = 0 the gasdynamic parameters will not change in the absorption zone. To realize the first case it is necessary that

$$\frac{\nu_k}{\nu_I}l_k > 1 - \frac{\rho Rh}{\mu K_B k_\nu I}\sum_{i=1}^{m}\nu_i f_i \gamma_i,$$

$(4)$

while for realization of the second

$$\frac{\nu_k}{\nu_I} l_k < 1 - \frac{\rho R h}{\mu K_b k_\nu I} \sum_{i=1}^{m} \nu_i f_i \gamma_i. \tag{5}$$

To make the analysis more graphic, we first assume that

$$\frac{\rho R h}{\mu K_b k_\nu I} \sum_{i=1}^{m} \nu_i f_i \gamma_i = 0. \tag{6}$$

Such a situation is ordinarily realized when the absorbing transition is far from saturation or the rates of the V–T and V–V' processes are small ($\tau_I \ll \tau_{V-T} \leqslant \tau_{V-V'}$, $\tau_{V-T}$ and $\tau_{V-V'}$ are the characteristic times of these processes). It is seen from (4) and (5) that the possibility of cooling (or heating) the gas under the effect of resonance radiation is determined by the relationship between the radiation frequency and the product of the normal vibrations frequency by the number of vibrational quanta acquired by the mode under the induced transitions.

Let us recall that if a molecule with dissimilar nuclei is in the ground electronic state (the projection of the orbital moment on the internuclear axis is $\Lambda = 0$) then only the transitions of the P- and R- branches are allowed in the dipole approximation. If $\Lambda \neq 0$ (for instance, in the $\Pi$, $\Delta$, $\Phi$... states), then the Q-branch transitions [3] are also possible. To calculate $\nu_I$ we use the approximation of a fluctuating rotator for which the rotational energy $E_j$ is given by the expression [3]

$$E_j = B_V j(j+1) - D_V j^2 (j+1)^2, \tag{7}$$
$$B_V = B_e - \alpha_e \left( V + \frac{1}{2} \right), \quad D_V = D_e + \beta_e \left( V + \frac{1}{2} \right),$$

where $B_e$ and $D_e$ are the rotational constants determined in the absence of nuclei vibrations, $\alpha_e$ and $\beta_e$ are certain small quantities ($\alpha_e > 0$), $j$ is the rotational, and V the vibrational quantum number. In a first approximation we can set $D_V \approx 0$.

Let us consider absorption in the P-, Q-, and R-branches successively.

For the P-branch ($j'' = j' - 1 \equiv j - 1$)

$$\nu_I = \nu_k l_k - [j (B_{V''} + B_{V'}) - j^2 \alpha_e l_k]/h.$$

The condition $\nu_k l_k - \nu_I > 0$ ($J < 0$) is satisfied for any $j$.

For the Q-branch ($j'' = j' \equiv j$)

$$\nu_I = \nu_k l_k - [j^2 \alpha_e l_k - j \alpha_e l_k]/h$$

and $J < 0$ also for any $j$.

For the R-branch ($j'' = j' + 1 \equiv j + 1$)

$$\nu_I = \nu_k l_k - [j^2 \alpha_e l_k + j (3B_{V''} - B_{V'}) + 2B_{V''}]/h.$$

Here $J < 0$ only for $j > j_0$,

$$j_0 = \frac{3B_{V''} - B_{V'} + \sqrt{(3B_{V''} - B_{V'})^2 + 8B_{V''} \alpha_e l_k}}{2\alpha_e l_k}. \tag{8}$$

It should be noted that for diatomic dipole molecules, for instance, hydrogen halide molecules, the boundary value $j_0$, at which the condition $J < 0$ starts to be satisfied, is sufficiently large (for instance, $j_0 = 67$ for hydrogen chloride).

Consequently (8) yields elevated values of $j_0$, and the complete formula (7) must be utilized for the calculation of $\nu_I$. The quantity $j_0$ will here be less than that calculated by means of (8).

Let us note that for transitions with $l_k > 1$ gas cooling during radiation absorption in the R-branch will occur for smaller $j_0$ than for transitions with $l_k = 1$.

Therefore, for $0 < M < 1/\sqrt{\varkappa}$ and $M > 1$, if the molecules are in the ground electronic state, gas cooling is possible not only during radiation absorption in the P-branch, as was noted earlier in [1], but also for large rotational numbers ($j > j_0$) in the R-branch. If the molecules are in the excited electronic states ($\Pi$, $\Delta$,...), then for the same M values gas cooling will also hold even for absorption in the Q-branch.

For $1/\sqrt{} < M < 1$ the gas will be cooled independently of whether its molecules are in the ground or the excited electronic states, only for radiation absorption in the R-branch with $j < j_0$.

We linearize the equations of the system (3) for a quantitative estimate of the effect of diminishing the translational temperature. The following relationships [4]

$$\frac{T-T_0}{T_0} = \frac{1-\varkappa M_0^2}{C_p T_0 \left(1 - M_0^2\right)} J', \quad \frac{u-u_0}{u_0} = \frac{\varkappa - 1}{a_0^2 \left(1 - M_0^2\right)} J', \tag{9}$$

$$\frac{p-p_0}{p_0} = \frac{\varkappa(\varkappa - 1)}{a_0^2 \left(1 - M_0^2\right)} J', \quad \frac{\rho - \rho_0}{\rho_0} = -\frac{(\varkappa - 1)}{\left(1 - M_0^2\right) a_0^2} J',$$

$$J' = \frac{\nu_I - \nu_h l_h}{\nu_h l_h} \int\limits_0^x \sum_{i=1}^m de_V^i$$

are obtained for the changes in the macroscopic gas parameters (T, p, $\rho$, u), where $T_0$, $p_0$, $u_0$, $\rho_0$ are the unperturbed stream parameters. Furthermore, we use the notation $\Delta T = T - T_0$ and $\delta T = (T - T_0)/T_0$.

For $x = L_I$ ($L_I = u\tau_I$) (we denote the parameters in this section with the subscript I), let the transition $(V', j') \to (V'', j'')$ be saturated completely, i.e., $k_\nu(L_I) = 0$. The vibrational energy of the absorbing component in this section is determined by the relationship

$$e_V^h (L_I) = \frac{R}{\mu}\, \gamma_h\, \frac{h\nu_h}{K_B} \left\{ \frac{B_{V''}}{B_{V'}} \exp\left[\frac{E_{j''} - E_{j'}}{K_B T_I}\right] - 1 \right\}^{-1}.$$

Taking into account that for $x = 0$

$$e_V^h (x = 0) = \frac{R}{\mu}\, \gamma_h\, \frac{h\nu_h}{K_B} \left[ \exp\left(\frac{h\nu_h}{K_B T_0}\right) - 1 \right]^{-1},$$

it is possible to obtain

$$J' = \frac{E_{j''} - E_{j'}}{l_h K_B}\, \frac{R}{\mu}\, \gamma_h \left\{ \left[ \frac{B_{V''}}{B_{V'}} \exp\left(\frac{E_{j'} - E_{j''}}{K_B T_I}\right) - 1 \right]^{-1} - \left[ \exp\left(\frac{h\nu_h}{K_B T_0}\right) - 1 \right]^{-1} \right\}; \tag{10}$$

$$T_I = T_0 + \frac{1 - \varkappa M_0^2}{C_p \left(1 - M_0^2\right)} J'. \tag{11}$$

It is seen from (10) that the values of $\Delta T_I$, $\Delta p_I$, $\Delta \rho_I$, and $\Delta u_I$ diminish as $\left| E_{j'} - E_{j''} \right|$ increases (i.e., as j increases) and as $l_k$ increases.

The relationships (9) and (10) permit estimation of the maximal change, for given $M_0$ in the gas parameters in the resonance radiation absorption zone. It is also seen from the relationships mentioned that the change in T, p, $\rho$ can be greater in a moving than in a fixed gas, i.e., a gasdynamic magnification of the effect of the kinetic action holds. The change in gas parameters in the kinetic cooling regime ($J < 0$) is represented in Fig. 1 as a function of the unperturbed stream M [here 1) $\Delta T$, 2) $\Delta p$, 3) $\Delta \rho$, 4) $\Delta u$]. For $J > 0$ the change in T, p, $\rho$, and u as a function of $M_0$ has a shape analogous to the dependences in Fig. 1, and the mirror image of the corresponding curves is obtained with respect to the abscissa axis. The dashed lines in Fig. 1 display the curve branches where the linear approximation (8) can yield considerable error. Detailed dependences were obtained in [5] for the analysis of polyatomic molecule flows in a radiation field whose frequency equals the intermodal transition, for a different kinetic cooling mechanism.
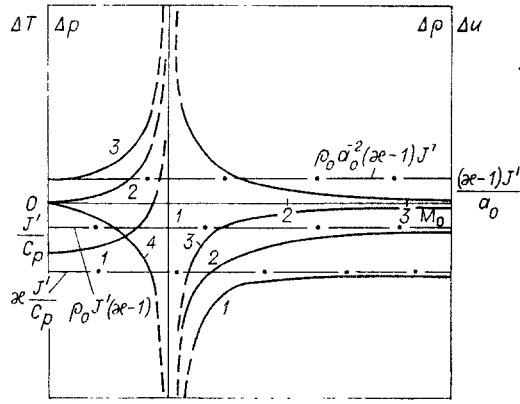
Fig. 1

If $\left|E_{j'} - E_{j''}\right| \ll K_B T_I$, then for $M_0 = 0$ with $E_{j'} - E_{j''} > 0$ it is possible to obtain

$$T_I = \frac{T_0 + \dfrac{E_{j'} - E_{j''}}{l_k K_B C_p} \dfrac{R}{\mu} \gamma_k \left[\exp\left(\dfrac{h\nu_k}{K_B T_0}\right) - 1\right]^{-1}}{1 + \dfrac{R}{\mu} \dfrac{\gamma_k}{C_p l_k}}. \tag{12}$$

from (10) and (11).

For a gas containing only molecular components $\left(\sum\limits_i \gamma_i = 1\right)$, a simple formula is obtained that determines the maximal value of the temperature in the radiation absorption zone

$$\frac{J'}{C_p} \equiv T_I - T_0 = \frac{(E_{j'} - E_{j''})\left[\exp\left(\dfrac{h\nu_k}{K_B T_0}\right) - 1\right]^{-1} - T_0}{\dfrac{7}{2}\dfrac{l_k}{\gamma_k} + 1}. \tag{13}$$

It follows from (13) that for $h\nu_k/T_0 < K_B$ and $E_{j'} - E_{j''} \ll T_0$ $J'/C_p = -T_0 \Big/ \left(\dfrac{7}{2}\dfrac{l_k}{\gamma_k} + 1\right)$,

i.e., independently of the species of absorbing gas molecules. This is also verified by numerical computations of the complete system of gasdynamics equations and kinetic equations under the condition $f_i = 0$, carried out for hydrogen halide molecules.

Let us again mention that the expressions (10) and (13) obtained are an upper bound for J' (or $\Delta T$). In reality, the quantity J' and, therefore, $\Delta T$ as well will be substantially less because of both the processes R–T and V–T and V–V exchange.

To analyze the influence of V–T relaxation and intermolecular V–V' exchange on the change in the macroscopic flux parameters in the absorption zone, we now examine the case when

$$\frac{\rho R h}{\mu K_B k_\nu I} \sum_{i=1}^{m} \nu_i f_i \gamma_i \neq 0, \quad E_{j''} < E_{j'}.$$

Let the gas consist of two components with vibrations k and s with dissimilar relaxation times not connected by resonance, while the radiation is absorbed by the k-th component. In this case the expression for J' can be represented in the form

$$J' = \int_0^x \left\{ \frac{k_\nu I}{\rho u} \frac{E_{j''} - E_{j'}}{h\nu_I} - \frac{pR}{\mu K_B T u} \Theta_k \gamma_k W_{ks} \gamma_s \left[ L_{ks}\left(\frac{l_k}{g_k} - \frac{\Theta_s}{\Theta_k} \frac{l_s}{g_s}\right) - \right. \right. \tag{14}$$

$$\left. \left. - (\varepsilon_k - \varepsilon_{k0}) \frac{\sum\limits_{i=1}^{m} W_{k0}^i \gamma_i}{W_{ks}\gamma_s} - (\varepsilon_s - \varepsilon_{s0}) \frac{\Theta_s}{\Theta_k} \frac{\gamma_s}{\gamma_k} \frac{\sum\limits_{i=1}^{m} W_{s0}^i \gamma_i}{W_{ks}\gamma_s} \right] \right\} dx,$$

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$$L_{ks} = \left[ \varepsilon_k^{l_k} (\varepsilon_s + 1)^{l_s} \exp\left( \frac{\Theta_k - \Theta_s}{I} \right) - (\varepsilon_k + 1)^{l_k} \varepsilon_s^{l_s} \right],$$

$$\Theta_i = \frac{h\nu_i}{K_B}, \quad \varepsilon_i = \left[ \exp\left( \frac{\Theta_i}{T_i} \right) - 1 \right], \quad \varepsilon_{i0} = \varepsilon_i(T),$$

where $W_{ks}$ is the rate constant for the intermolecular V–V' exchange between the k and s components, $W_{j_0}^i$ is the rate constant of V–T relaxation during a collision with the i-th partner (j = k, s), $T_i$ is the vibrational temperature of the i-th mode, $l_k$ and $l_s$ are the quantities of vibrational quanta acquired (or lost) by the k and s components, respectively, during a V–V' exchange, and $g_k$ and $g_s$ are multiplicities of the degeneration in their vibrations.

We shall assume that $\sum_i W_{k0}^i \gamma_i \gg W_{ks}\gamma_s$, i.e., V–T relaxation occurs more rapidly in the absorbing component than does V–V' exchange. In this case, in the interval $[0, L_{V-T}]$ where $L_{V-T} = u\tau_{V-T}$,

$$J' = \int_0^x \left\{ \frac{k_\nu I}{\rho u} \frac{E_{j''} - E_{j'}}{h\nu_I} + \frac{pR}{\mu K_B T u} \gamma_k \Theta_k \sum_i W_{k0}^i \gamma_i (\varepsilon_k - \varepsilon_{k0}) \right\} dx. \tag{15}$$

If $\tau_I \sim \tau_{V-T}$, then a situation can occur when the equality

$$\frac{k_\nu I}{\rho u} \frac{|E_{j''} - E_{j'}|}{h\nu_I} = \frac{Rp}{\mu K_B T u} \gamma_k \Theta_k \sum_i W_{k0}^i \gamma_i (\varepsilon_k - \varepsilon_{k0})$$

will be satisfied even before saturation of the absorbing transition, and changes in the macroscopic flux parameters will not occur in the absorption zone. The values of $\delta T$, $\delta p$, $\delta u$, $\delta\rho$ will here evidently be less than for $f_i = 0$. The essential feature of the action of V–T processes in a moving absorbing gas as compared with a fixed gas is the fact that energy liberation from the vibrational degrees of freedom into translational will occur for another value of the Mach number than that for which resonance radiation resulted in the appearance of energy flux from the translational into the vibrational degrees of freedom. This feature can result in a smaller rise in gas temperature because of V–T relaxation as compared with a fixed gas.

To be graphic, we examine the case when $\tau_I < \tau_{V-T}$. For $x = L_I$ the parameters in the absorption zone are determined by (9) and (10). Here $M_I = u_I \left( \varkappa \frac{R}{\mu} T_I \right)^{-1/2} \equiv M_0$. For $x = L_{V-T}$ the upper bound for $J'$ and the temperature $T_\tau$ yields

$$J' = \frac{\Theta_k R}{\mu} \gamma_k \left\{ \left[ \frac{B_{V''}}{B_{V'}} \exp\left( \frac{E_{j'} - E_{j''}}{K_B T_I} \right) - 1 \right]^{-1} - \left[ \exp\left( \frac{\Theta_k}{T_\tau} \right) - 1 \right]^{-1} \right\}; \tag{16}$$

$$T_\tau = T_I + \frac{1 - \varkappa M_I^2}{1 - M_I^2} \frac{J'}{C_p}. \tag{17}$$

Then we will have from (17) and (11)

$$\frac{T_I - T_0}{T_\tau - T_I} \approx \frac{1 - \varkappa M_0^2}{1 - \varkappa M_I^2} \frac{1 - M_I^2}{1 - M_0^2} \frac{E_{j''} - E_{j'}}{l_k h\nu_k}. \tag{18}$$

In a fixed gas (M → 0)

$$\frac{T_I - T_0}{T_\tau - T_I} = \frac{E_{j''} - E_{j'}}{l_k h\nu_k}. \tag{19}$$

It follows from a comparison of (18) and (19) that since $M_I > M_0$, the quantity $\left| \frac{T_0 - T_I}{T_\tau - T_I} \right|$ is greater in a moving than in a fixed gas for $M_0 > 1$ and $M_I > 1$. This condition is

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satisfied if $T_T < T_T^F$ ($T_T^F$ is the value of $T_T$ in the fixed gas). An interesting situation occurs if $1/\sqrt{\varkappa} < M_I < 1$. In this case the V–T relaxation will also result in gas cooling.

We now assume that intermolecular V–V' exchange occurs more rapidly than V–T relaxation, i.e., $\sum_i W_{k0}^i \gamma_i \ll W_{ks}\gamma_s$, $\sum_i W_{s0}^i \gamma_i \ll W_{ks}\gamma_s$.

Then in the interval $[0, L_{V-V'}]$, where

$$J' = \int_0^x \left\{ \frac{k_v I}{\rho u} \frac{E_{j''} - E_{j'}}{h\nu_I} - \frac{pR}{\mu K_B T u} \gamma_k \Theta_h W_{ks}\gamma_s L_{ks} \left( \frac{l_h}{g_k} - \frac{l_s}{g_s} \frac{\Theta_s}{\Theta_h} \right) \right\} dx. \tag{20}$$

Let $\Theta_s l_s/g_s > \Theta_h l_h/g_h$. If $T_S < T_k$ here, then the quantity $|J'|$ will be greater than for $f_i = 0$ and the change in the macroscopic stream parameters in the absorption zone can be stronger. If $T_S > T_k$, then $|J'|$ will be less than for $f_i = 0$ and for

$$\frac{k_v I}{\rho u} \frac{|E_{j''} - E_{j'}|}{h\nu_I} < \frac{Rp}{\mu K_B T u} \Theta_h \gamma_h W_{ks}\gamma_s L_{ks} \left( \frac{l_h}{g_h} - \frac{l_s}{g_s} \frac{\Theta_s}{\Theta_h} \right) \tag{21}$$

the sign of the action $(J')$ generally changes to the opposite. For $\Theta_h l_h/g_h > \Theta_s l_s/g_s$, on the other hand, an increase in $|J'|$ will hold for $T_S > T_k$. If $T_k > T_S$, then the quantity $|J'|$ will be diminished while the sign of $J'$ will generally change when (21) is satisfied. A change in the magnitude of the energy flux from the translational to the vibrational degrees of freedom upon insertion of the impurity molecules (s) in the resonantly absorbing gas is explained by the fact that in a V–V' exchange the part of the energy, equal to $(\Theta_s - \Theta_k)K_B$, being transmitted from one type of vibration to another is selected from the translational degrees of freedom (or is liberated in the translational degrees of freedom). It should be noted that the possibility of cooling a gas of diatomic molecules during V–V' exchange was first discussed in [6].

To estimate the effect of changes in the macroscopic flux parameters because of V–V' exchange between molecules of appropriate mixture components, we assume that $\tau_I < \tau_{V-V'}$ and saturation of the absorbing transition occurs more rapidly than the V–V' exchange starts. Let quasiequilibrium also be achieved between the k and s vibrations in the section $x_V = L_{V-V'}$ ($x_V > L_I$). In this case we can obtain for the upper bound of $J'$

$$J' = \gamma_h (\Theta_s - \Theta_h) \frac{R}{\mu} \left\{ \left[ \frac{B_{V''}}{B_{V'}} \exp\left( \frac{E_{j'} - E_{j''}}{K_B T_I} \right) - 1 \right]^{-1} - \left[ \exp\left( \frac{\Theta_h}{T_h} \right) - 1 \right]^{-1} \right\}. \tag{22}$$

The quantity $T_k$ is here determined from the equations

$$\frac{B_{V''}}{B_{V'}} \exp\left( \frac{E_{j'} - E_{j''}}{K_B T_I} \right) - 1 \bigg]^{-1} \gamma_h + \left[ \exp\left( \frac{\Theta_s}{T_I} \right) - 1 \right]^{-1} \gamma_s = \tag{23}$$

$$= \left[ \exp\left( \frac{\Theta_h}{T_h} \right) - 1 \right]^{-1} \gamma_h + \left[ \exp\left( \frac{\Theta_h}{T_h} \right) \exp\left( \frac{\Theta_s - \Theta_h}{T} \right) - 1 \right]^{-1} \gamma_s;$$

$$T = T_I + \frac{1 - \varkappa M_I^2}{(1 - M_I^2) C_p} J'. \tag{24}$$

The relationships (22)–(24) permit estimation of the maximal gas temperature change for a given $M_I$ due to V–V' exchange. In conformity with (8), the other parameters (p, $\rho$, u) will also change.

Let us illustrate the results obtained in an example of specific diatomic molecule gas flows, namely, hydrogen chloride (HCl) and the mixture HCl–$H_2$. The frequency of the radiation acting on the flux is in resonance with the frequency of the center of the vibrational–rotational transition line $(0, j') \to (v, j'')$, where V = 1; 2. Numerical methods are used to solve the problem. Integration of the complete system of gasdynamics and vibrational kinetics equations was performed here exactly as in [4, 5]. The scheme of the relaxational processes and its corresponding kinetic equations were taken exactly as in [7].
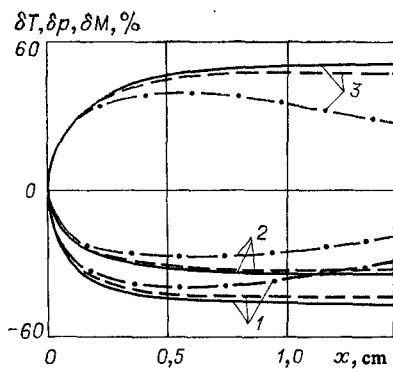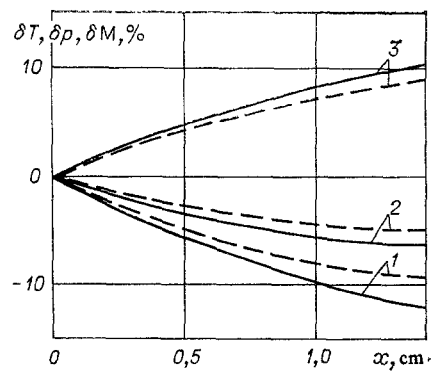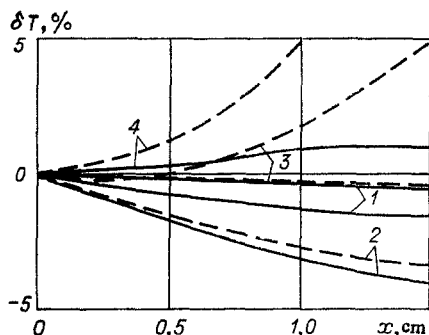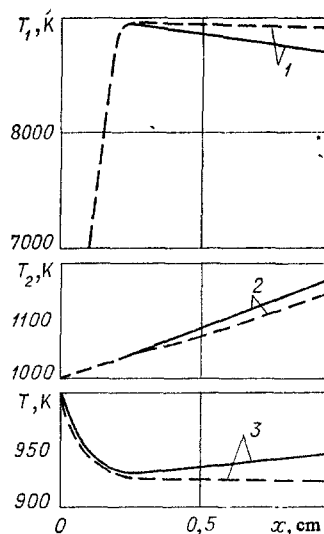
Fig. 2



Fig. 3



Fig. 4



Fig. 5

The absorption indices in the HCl molecule transitions under investigation were calculated by means of standard relationships [2] under the assumption of joint action by the Doppler and collisional spectrum line broadening mechanisms. The Einstein coefficients and shock broadening sections were selected exactly as in [7]. The values of $E_j''$ and $E_j'$ were computed by means of (7) for the determination of the relative populations of the upper and lower absorbing transition levels.

Results of computing the change in macroscopic flux parameters in the zone of action of radiation with $I_0 = 0.4$ MW/m$^2$ are represented in Fig. 2 for a hydrogen chloride flow with the initial parameters $T_0 = 1000°$K, $M_0 = 1.2$.

The frequency of the acting radiation is resonant with the frequency of the center of the vibrational—rotational transition $(0,11) \to (1,10)$ line. The numbers 1-3 here denote the relative change in the pressure $(\delta p)$, temperature $(\delta T)$, and Mach number $(\delta M)$. The change in the macroscopic parameters in a flux with $p_0 = 10^{-4}$ MPa in the absence of V—T processes is shown by the solid lines, the change in p, T, M when these processes act is shown by the dashed lines, and in a medium with elevated V—T relaxation rate $(p_0 = 5 \cdot 10^{-4}$ MPa) by the dash-dot lines. It is seen that an increase in the rate of the V—T processes (its increase is simulated by an increase in the gas pressure) will result in both diminution of the degree of kinetic cooling $(\delta T)$ and a change in the other macroscopic parameters $(\delta p, \delta M)$.

The change in the macroscopic flux parameters is weaker in radiation absorption at the P-branch overtones $(0, j) \to (2, j - 1)$ than for absorption at the ground transition. This is seen well from a comparison of Figs. 2 and 3 (the change in the macroscopic parameters is shown in Fig. 3 for absorption of radiation with $I_0 = 0.4$ MW/m$^2$ at the transition $(0,11) \to (2,10)$ for $T_0 = 1000°$K, $p_0 = 10^{-4}$ MPa, $M_0 = 1.2$, where the notation here is the same as in Fig. 2). However, even in this case the magnitude of the change in p, T, and M is significant and can reach 5-15%.

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The possibility of cooling a molecular gas during absorption of radiation in the R-branch is illustrated in Fig. 4. Here the relative change in the translational gas temperature ($\delta T$) is represented under the action of radiation with $I_0 = 10$ MW/m$^2$ whose frequency is in resonance with the frequency of the transitions $(0, j) \to (2, j + 1)$ with $j = 39, 34, 32, 29$ (curves 1-4, respectively). The unperturbed parameters of the medium are here the same as in the preceding case. As before, the change in $\delta T$ in the absence of V–T processes is shown by solid lines, and when these processes act, by dashes. The existence of the boundary $j_0$ from which value $(j > j_0)$ gas cooling starts to be observed is seen clearly. The strong influence of the V–T processes for small $j$ and the weaker influence for large $j$ ($j = 39$) is explained by the substantially high values of the absorption index for the transitions with $j = 29$, and therefore, also by the large amount of energy being liberated in the translational degrees of freedom. Let us note that numerical calculations also demonstrated the possibility of cooling a supersonic gas flow even for radiation absorption in the R-branch of the ground transition. However, the magnitude of the $j_0$ is here substantially greater and to obtain significant $\delta T$ high values of the intensity of the acting radiation ($I_0 > 10^2$ MW/m$^2$) are necessary.

As mentioned above, the possibility of diminishing the translational flux temperature in a binary gas mixture can be specified also by the nonresonance V–V' exchange process. To illustrate such a cooling mechanism, a flow of a mixture of gases HCl and $H_2$ ($\gamma_{HCl} = 0.2$, $\gamma_{H_2} = 0.8$) under the action of resonant radiation with $I_0 = 10$ MW/m$^2$, whose frequency equals the frequency of the vibrational—rotational transition $(0, 11) \to (1, 10)$ to the HCl molecule, was considered. The unperturbed parameters of the medium were assumed to be equal $p_0 = 10^{-3}$ MPa, $T_0 = 1000°$K, $M_0 = 1.2$, and the zone of radiation action $L = 2 \cdot 10^{-3}$ m. The change in the vibrational $T_i$ ($i = 1$ for HCl, $i = 2$ for $H_2$, and curves 1 and 2, respectively) and the translational temperature T (curve 3) along the longitudinal coordinate x is shown in Fig. 5; there is no energy exchange between translational and vibrational degrees of freedom (dashed lines) and when the V–T exchange exists. It is seen from the distributions represented that additional flux cooling for $I_0 = 0$ is caused by the intermolecular V–V' energy exchange process between the excited HCl molecules and the unexcited $H_2$ molecules ($T_1$ diminishes while $T_2$ rises). Vibrational—translational relaxation here results in a reduction of the zone of existence of the reduced temperature range and in diminution of the quantity $\delta T$.

In conclusion, we note that the change in macroscopic flux parameters because of the considered mechanisms during resonance radiation absorption by a gas can turn out to be governing in investigations of the nonstationary self-focusing of laser beams.

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