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ALLOWING FOR INTERMOLECULAR ENERGY EXCHANGE IN THE DESCRIPTION OF RELAXATION PROCESSES IN TERMS OF ADIABATIC VARIABLES

V. M. Strel'chenya

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The solution of the complete system of gasdynamic equations, supplemented by kinetic equations describing vibrational-rotational relaxation or the kinetics of phase transformations [1-4], is an exceedingly complex problem. The difficulty of the problem makes it important to find simpler methods of describing transformation kinetics by means of approximate solutions of relaxation equations [1, 5]. One such method was developed in [6-9] for rate coefficients of arbitrary form. It is based on the introduction of adiabatic variables which diagonalize (in the case of distributions which are smooth with respect to quantum number) the initial system of kinetic equations to within a small parameter.

In the present study – a continuation of [6-9] – we examine two methods of describing the contribution of intermolecular energy exchange to the relaxation of the populations of individual levels in terms of adiabatic variables. The methods make it possible to obtain approximate analytic solutions of the kinetic equations for different relaxation regimes.

1. System of Relaxation Equations and Adiabatic Variables. Let us examine the process of relaxation in a mixture of molecules of species s. We will assume that the internal state of a molecule is characterized by a single quantum number ν (such as in the case of delayed vibrational relaxation in a mixture of diatomic molecules [10]). The equations for the populations of individual energy levels $n_s(\nu) \equiv n_s(\nu; \mathbf{r}, t)$ have the form [10]

$$\begin{gathered}\frac{\partial n_s(\nu)}{\partial t} + \nabla\cdot[\mathbf{u}n_s(\nu)] + \nabla\cdot[\mathbf{u}_s(\nu)n_s(\nu)] = I_\nu(s\,|\,\mathbf{n}),\\ I_\nu(s\,|\,\mathbf{n}) = \sum_{i=1,2} I_\nu^{(i)}(s\,|\,\mathbf{n}) = \sum_{i=1,2}\sum_{s_1} I_\nu^{(i)}(s, s_1\,|\,\mathbf{n}),\end{gathered} \tag{1.1}$$

where $\mathbf{u}$ is the hydrodynamic velocity; $\mathbf{u}_s(\nu)$ is the rate of diffusion of molecules of species s in the state ν; $I_\nu^{(1)}$ is the linear part of the collision integral, describing the transfer of energy between the internal and translational degrees of freedom of the gas; and $I_\nu^{(2)}$ is the quadratic part of the collision integral, responsible for intermolecular energy transfer.

For the concentrations

$$x_s(\nu) \equiv n_s(\nu)/n,\ \ n \equiv \sum_{s,\nu} n_s(\nu) \tag{1.2}$$

Eqs. (1.1) take the form

$$\dot{x}_s(\nu) + n^{-1}\nabla\cdot[\mathbf{u}_s(\nu)n_s(\nu)] = I_\nu(s|\mathbf{x}) \tag{1.3}$$

at

$$\begin{gathered}I_\nu^{(1)}(s, s_1\,|\,\mathbf{x}) = \sum_\mu [P_{\mu\nu}(s, s_1)\,x_s(\mu) - P_{\nu\mu}(s, s_1)\,x_s(\nu)],\\ I_\nu^{(2)}(s, s_1\,|\,\mathbf{x}) = \sum_{\varkappa,\lambda,\mu}\left[P_{\mu\nu}^{\varkappa\lambda}(s, s_1)\,x_s(\mu)\,x_{s_1}(\varkappa) - P_{\nu\mu}^{\lambda\varkappa}(s, s_1)\,x_s(\nu)\,x_{s_1}(\lambda)\right]\end{gathered} \tag{1.4}$$

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(the dot denotes the total derivative with respect to time). In (1.4), $P_{\mu\nu}(s, s_1)$ and $P_{\mu\nu}{}^{\kappa\lambda}(s, s_1)$ are the probabilities of the inelastic transitions $\mu \to \nu$ and $\mu, \kappa \to \nu, \lambda$ in the collision of molecules s and s_1 per unit of time.

After introduction of the probabilities $P_{\mu\nu}(s) \equiv \sum_{s_1} P_{\mu\nu}(s, s_1)$ of $\mu \to \nu$ transitions in a molecule s accompanying a collision with any other particle, we find it convenient to write the linear part of the collision integral as a sum taken over the number of transferred quanta γ:

$$\begin{gathered} I_\nu^{(1)}(s \mid \mathbf{x}) = \sum_\gamma I_{\nu;\gamma}^{(1)}(s \mid \mathbf{x}), \\ I_{\nu;\gamma}^{(1)}(s \mid \mathbf{x}) \equiv P_{\nu+\gamma\nu}(s)\, x_s(\nu+\gamma) - [P_{\nu\nu+\gamma}(s) + P_{\nu\nu-\gamma}(s)]\, x_s(\nu) + \\ + P_{\nu-\gamma\nu}(s)\, x_s(\nu-\gamma). \end{gathered} \tag{1.5}$$

We change over from the concentrations $x_s(\nu)$ ($\nu \geq 0$) to the adiabatic variables $x_s(0)$, $f_s(\nu)$ ($\nu \geq 1$) for $I_\nu{}^{(1)}$ by means of the nonlinear substitution of variables [7]

$$a_\nu(s) f_s(\nu) = x_s(\nu)/x_s(\nu-1), \quad a_\nu(s) = P_{\nu-1\nu}(s)/P_{\nu\nu-1}(s), \tag{1.6}$$

It follows from (1.6) that

$$x_s(\nu+\gamma) = x_s(\nu) \prod_{\delta=1}^{\gamma} a_{\nu+\delta}(s)\, f_s(\nu+\delta). \tag{1.7}$$

Insertion of (1.6)-(1.7) into (1.3)-(1.5) leads to the following systems of equations [7-9] (we are not yet examining the quadratic part of the collision integral and, for simplicity, we ignore diffusion):

$$\begin{gathered} \frac{d}{dt} \ln f_s(\nu) = R_s(f_s(\nu)) + E_s(\mathbf{f}_s) + \Gamma_\nu(s), \\ \frac{d}{dt} \ln x_s(0) = \sum_\gamma \left[\Pi_{0;\gamma}^{+}(s) \prod_{\delta=1}^{\gamma} f_s(\delta) - P_{0\gamma}(s) \right], \end{gathered} \tag{1.8}$$

where

$$R_s = \sum_\gamma R_s^{(\gamma)} \equiv \sum_\gamma \left[A_{\nu;\gamma}(s) f_s^\gamma(\nu) + B_{\nu;\gamma}(s) + C_{\nu;\gamma}(s) f_s^{-\gamma}(\nu) \right]; \tag{1.9}$$

$$\begin{gathered} E_s = \sum_\gamma E_s^{(\gamma)} \equiv \sum_\gamma \{ f_s^\gamma(\nu) \left[\Pi_{\nu;\gamma}^{+}(s) (\pi_s(\nu, \gamma; 0) - 1) - \right. \\ \left. - \Pi_{\nu-1;\gamma}^{+}(s) (\pi_s(\nu-1, \gamma; \nu) - 1) \right] + f_s^{-\gamma}(\nu) \left[\Pi_{\nu;\gamma}^{-}(s) \times \right. \\ \left. \times \left(\pi_s^{-1}(\nu-\gamma, \gamma; \nu) - 1 \right) - \Pi_{\nu-1;\gamma}^{-}(s) \left(\pi_s^{-1}(\nu-1-\gamma, \gamma; \nu) - 1 \right) \right] \}; \end{gathered} \tag{1.10}$$

$$\Gamma_\nu(s) = -\frac{d}{dt} \ln a_\nu(s). \tag{1.11}$$

In (1.9)-(1.10) we introduced the notation

$$\begin{gathered} A_{\nu;\gamma}(s) = \Pi_{\nu;\gamma}^{+}(s) - \Pi_{\nu-1;\gamma}^{+}(s); \quad C_{\nu;\gamma}(s) = \Pi_{\nu;\gamma}^{-}(s) - \Pi_{\nu-1;\gamma}^{-}(s); \\ B_{\nu;\gamma}(s) = -\left[(P_{\nu\nu+\gamma}(s) - P_{\nu-1\nu-1+\gamma}(s)) + (P_{\nu\nu-\gamma}(s) - P_{\nu-1\nu-1-\gamma}(s)) \right]; \\ \Pi_{\nu;\gamma}^{+}(s) = P_{\nu+\gamma\nu}(s) \prod_{\delta=1}^{\gamma} a_{\nu+\delta}(s); \quad \Pi_{\nu;\gamma}^{-}(s) = P_{\nu-\gamma\nu}(s) \prod_{\delta=1}^{\gamma} a_{\nu-\gamma+\delta}^{-1}(s); \\ \pi_s(\nu, \gamma; \mu) = \prod_{\delta=1}^{\gamma} f_s(\nu+\delta)/f_s(\mu). \end{gathered}$$

In the case of smooth distributions for which $f_s(\nu)$ depends weakly on ν, the nondiagonal (with respect to ν) term $E_s(\mathbf{f}_s)$ in (1.8) is small [since $\pi_s(\nu, \gamma; \mu) \sim 1$] and in the zeroth approximation with respect to E_s we can solve Eqs. (1.8) independently of one another. Thus, transformation (1.6) will be regarded as an analog of the transition from phase to adiabatic variables in Hamiltonian mechanics, and the variables $f_s(\nu)$ themselves will be referred as adiabatic [11].

2. Transition to Adiabatic Variables in the Quadratic Part of the Collision Integral. There are two methods of accounting for the contribution of intermolecular exchange in Eqs. (1.8).

A. The first method involves the introduction of the "effective" probabilities of inelastic transitions $P_{\mu\nu}^*(s)$. In terms of the latter, the right side of system (1.3) can be written in a form analogous to (1.5) [7, 9] with

$$\begin{aligned} P^*_{\nu+\gamma\nu}(s) &\equiv \sum_{s_1}\Big[P_{\nu+\gamma\nu}(s, s_1) + \sum_{\delta} M^{(\delta)}_{\nu+\gamma\nu}(s, s_1)\Big], \\ P^*_{\nu\nu+\gamma}(s) &\equiv \sum_{s_1}\Big[P_{\nu\nu+\gamma}(s, s_1) + \sum_{\delta} M^{(\delta)}_{\nu\nu+\gamma}(s, s_1)\Big]. \end{aligned} \tag{2.1}$$

Here, the moments $M_{\mu\nu}^{(\delta)}$ are determined by the equalities

$$\begin{aligned} M^{(\delta)}_{\nu+\gamma\gamma}(s, s_1) &\equiv \sum_{\varkappa} P^{\varkappa-\delta\varkappa}_{\nu+\gamma\nu}(s, s_1)\, x_{s_1}(\varkappa-\delta), \\ M^{(\delta)}_{\nu\nu+\gamma}(s, s_1) &\equiv \sum_{\varkappa} P^{\varkappa\varkappa-\delta}_{\nu\nu+\gamma}(s, s_1)\, x_{s_1}(\varkappa). \end{aligned} \tag{2.2}$$

Thus, in this approach, allowance for intermolecular energy transfer does not alter the structure of Eqs. (1.8), and in Eqs. (1.6)-(1.7) it is sufficient to make the substitution

$$a_\nu(s) \to a^*_\nu(s) \equiv P^*_{\nu-1\nu}(s)/P^*_{\nu\nu-1}(s).$$

Here, however, system (1.8) must be supplemented by a system of equations describing the evolution of the moments $M_{\mu\nu}^{(\delta)}$ over time. The derivation of this system depends on the character of the dependence of the rate coefficients in (2.2) on the quantum numbers. For example, the form of this system was discussed in [6, 7, 9] for the vibrational relaxation of harmonic oscillators in the presence of sources of vibrationally excited particles and weakly harmonic oscillators with small deviations from equilibrium. A separate article will examine the derivation of a closed system of equations for the moments $M_{\mu\nu}^{(\delta)}$ in the case of vibrational relaxation in a mixture of weakly harmonic oscillators with arbitrary deviations from equilibrium. Nevertheless, the approach described above can be used to obtain approximate analytic solutions for system (1.8) in those situations when the moments $M_{\mu\nu}^{(\delta)}$ are slow variables compared to $f_S(\nu)$.

B. The second method involves "symmetrization" of system (1.3), which is "asymmetric" in the sense that its left side and the first term in the right side are linear with respect to x_S, while the second term is quadratic. Recalling that in the derivation of (1.3) it was assumed that the probabilities of $\mu \to \nu$ transitions in a molecule s are independent of the internal state of the partner in the collision s_1, if we drop this assumption we can write $I_\nu^{(1)}(s, s_1|\mathbf{x})$ in the form

$$I^{(1)}_\nu(s, s_1\,|\,\mathbf{x}) = \sum_{\varkappa,\mu}\big[P^{\varkappa\varkappa}_{\mu\nu}(s, s_1)\, x_s(\mu)\, x_{s_1}(\varkappa) - P^{\varkappa\varkappa}_{\nu\mu}(s, s_1)\, x_s(\nu)\, x_{s_1}(\varkappa)\big]. \tag{2.3}$$

Here, $\sum\limits_{\varkappa} P^{\varkappa\varkappa}_{\mu\nu}(s, s_1)\, x_{s_1}(\varkappa) = P_{\mu\nu}(s, s_1)$ i.e., $P^{\varkappa\varkappa}_{\mu\nu}(s, s_1) \approx P_{\mu\nu}(s, s_1)/x_{s_1}$, $x_{s_1} = \sum\limits_{\varkappa} x_{s_1}(\varkappa)$. To obtain the "symmetric" (quadratic with respect to x_S) analog of kinetic equations (1.3), with allowance for (2.3) we write the following system for the product of the concentrations

$$y_{ss_1}(\nu, \lambda) \equiv x_s(\nu)\, x_{s_1}(\lambda), \tag{2.4}$$

for simplicity ignoring diffusion processes:

$$\begin{aligned} \frac{d}{dt} y_{ss_1}(\nu, \lambda) = {} & \sum_{\mu}\big[P^{\lambda\lambda}_{\mu\nu}(s, s_1)\, y_{ss_1}(\mu, \lambda) - P^{\lambda\lambda}_{\nu\mu}(s, s_1)\, y_{ss_1}(\nu, \lambda)\big] + \sum_{\varkappa}\big[P^{\varkappa\lambda}_{\nu\nu}(s, s_1) y_{ss_1}(\nu, \\ & \varkappa) - P^{\lambda\varkappa}_{\nu\nu}(s, s_1)\, y_{ss_1}(\nu, \lambda)\big] + \sum_{\varkappa,\mu}\big[P^{\varkappa\lambda}_{\mu\nu}(s, s_1)\, y_{ss_1}(\mu, \varkappa) - P^{\lambda\varkappa}_{\nu\mu}(s, s_1)\, y_{ss_1}(\nu, \lambda)\big]. \end{aligned}$$

As before, it is convenient to rewrite the collision integral in the form of a sum taken over the number of transmitted quanta. To abbreviate the notation [taking into account the symmetry properties of the transition probabilities $P_{\mu\nu}^{\kappa\kappa}(s, s_1) = P_{\kappa\kappa}^{\mu\nu}(s_1, s)$, $P_{\mu\nu}^{\kappa\lambda}(s, s_1) = P_{\kappa\lambda}^{\mu\nu}(s_1, s)$], we perform the summation $SF_{\nu,\lambda}(s, s_1) \equiv F_{\nu,\lambda}(s, s_1) + F_{\lambda,\nu}(s_1, s)$, where $F_{\nu,\lambda}(s, s_1)$ is any quantity dependent on the quantum numbers ν and λ of molecules s and s_1, respectively:

$$\dot{y}_{ss_1}(\nu, \lambda) = \sum_{\gamma} \sum_{i=1,2} \mathrm{S} I^{(i)}_{\nu,\lambda;\gamma}(s, s_1 \,|\, \mathbf{y}). \tag{2.5}$$

Here, $I_{\nu,\lambda;\gamma}{}^{(1)}$ has the same structure as $I_{\nu;\gamma}{}^{(1)}$ in (1.5), while with allowance for only the most likely isoquantum transitions $I_{\nu;\lambda;\gamma}{}^{(2)}$ has the form

$$I^{(2)}_{\nu,\lambda;\gamma} = P^{\lambda-\gamma\lambda}_{\nu+\gamma\nu}(s, s_1)\, y_{ss_1}(\nu + \gamma, \lambda - \gamma) - P^{\lambda\lambda-\gamma}_{\nu\nu+\gamma}(s, s_1)\, y_{ss_1}(\nu, \lambda).$$

We introduce variables $z_{ss_1}(\nu, \lambda)$ normalized with respect to the equilibrium (Boltzmann) values:

$$z_{ss_1}(\nu, \lambda) = y_{ss_1}(\nu,\lambda)/\left[x_s^{\mathrm{B}}(\nu)\, x_{s_1}^{\mathrm{B}}(\lambda)\right],$$
$$x_s{}^{\mathrm{B}}(\nu) \equiv Q_s^{-1} \exp\left[-\varepsilon_s(\nu)\right];\ \varepsilon_s(\nu) \equiv E_s(\nu)/k_{\mathrm{B}}T. \tag{2.6}$$

In terms of these variables, (2.5) appears as

$$\frac{d}{dt} \ln z_{ss_1}(\nu, \lambda) + \left[\varepsilon_s(\nu) - \langle\varepsilon_s\rangle + \varepsilon_{s_1}(\lambda) - \langle\varepsilon_{s_1}\rangle\right] \frac{d}{dt} \ln T = \sum_{\gamma} \sum_{i=1,2} \mathrm{S} I^{(i)}_{\nu,\lambda;\gamma}(s, s_1 \,|\, \mathbf{z})$$
$$\left(\langle\alpha_s\rangle \equiv \left[\sum_{\nu} \alpha_s(\nu) \exp(-\varepsilon_s(\nu))\right] \Big/ \left[\sum_{\nu} \exp(-\varepsilon_s(\nu))\right]\right). \tag{2.7}$$

In system (2.7), we change over to the adiabatic variables $z_{ss_1}(0, \lambda)$, $f_{ss_1}(\nu, \lambda)$ $(\nu \geq 1)$, determined by analogy with (1.6) from the equalities

$$b_{\nu,\lambda}(s, s_1)\, f_{ss_1}(\nu, \lambda) = \frac{z_{ss_1}(\nu, \lambda)}{z_{ss_1}(\nu - 1, \lambda)},\quad b_{\nu,\lambda}(s, s_1) = \frac{P^{\lambda\lambda}_{\nu-1\nu}(s, s_1)\, x_s^{\mathrm{B}}(\nu - 1)}{P^{\lambda\lambda}_{\nu\nu-1}(s, s_1)\, x_s^{\mathrm{B}}(\nu)}. \tag{2.8}$$

Differentiating (2.8), we find

$$\frac{d}{dt} \ln f_{ss_1}(\nu, \lambda) = \frac{d}{dt}\left[\ln z_{ss_1}(\nu, \lambda) - \ln z_{ss_1}(\nu - 1, \lambda) - \ln b_{\nu,\lambda}(s, s_1)\right].$$

The derivatives of the variables $z_{ss_1}(\nu - 1, \lambda)$ and $z_{ss_1}(\nu, \lambda)$ can be calculated using system (2.7), while the coefficient can be calculated on the basis of its definition in (2.8). Then

$$\frac{d}{dt} \ln f_{ss_1}(\nu, \lambda) = \sum_{\gamma} \sum_{i=1,2} \mathrm{S}\left[I^{(i)}_{\nu,\lambda;\gamma} - I^{(i)}_{\nu-1,\lambda;\gamma}\right] + \dot{T} \frac{\partial}{\partial T} \ln \frac{P^{\lambda\lambda}_{\nu\nu-1}(s, s_1)}{P^{\lambda\lambda}_{\nu-1\nu}(s, s_1)}. \tag{2.9}$$

If we use the relation

$$z_{ss_1}(\nu + \gamma, \lambda) = z_{ss_1}(\nu, \lambda) \prod_{\delta=1}^{\gamma} b_{\nu+\delta,\lambda}(s, s_1) f_{ss_1}(\nu + \delta, \lambda)$$

which follows from (2.8), we can readily express the γ-quantum collision integrals in the right side of (2.9) through the adiabatic variables:

$$I^{(1)}_{\nu,\lambda;\gamma}(s, s_1 \,|\, \mathbf{f}) = \left[\Pi^{+}_{\nu,\lambda;\gamma}(s, s_1)\, \pi_{ss_1}(\nu, \lambda, \gamma; \nu)\, f^{\gamma}_{ss_1}(\nu, \lambda) - P^{\lambda\lambda}_{\nu\nu+\gamma}(s, s_1)\right] -$$
$$- \left[P^{\lambda\lambda}_{\nu\nu-\gamma}(s, s_1) - \Pi^{-}_{\nu,\lambda;\gamma}(s, s_1)\, \pi^{-}_{ss_1}(\nu - \gamma, \lambda, \gamma; \nu)\, f^{-\gamma}_{ss_1}(\nu, \lambda)\right],$$
$$I^{(2)}_{\nu,\lambda;\gamma}(s, s_1 \,|\, \mathbf{f}) = P^{\lambda\lambda-\gamma}_{\nu\nu+\gamma}(s, s_1)\left[\Phi_{\nu,\lambda;\gamma}(s, s_1) \frac{\pi_{ss_1}(\nu, \lambda, \gamma; \nu)}{\pi_{s_1s}(\lambda - \gamma, \nu + \gamma, \gamma; \lambda)} \times \frac{f^{\gamma}_{ss_1}(\nu, \lambda)}{f^{\gamma}_{s_1s}(\lambda, \nu + \gamma)} - 1\right],$$

where

$$\Phi_{\nu,\lambda;\gamma}(s, s_1) = \frac{P^{\lambda-\gamma\lambda}_{\nu+\gamma\nu}(s, s_1)}{P^{\lambda\lambda-\gamma}_{\nu\nu+\gamma}(s, s_1)} \frac{\Pi^{+}_{\nu,\lambda;\gamma}(s, s_1)\, \Pi^{-}_{\lambda,\nu+\gamma;\gamma}(s_1, s)}{P^{\lambda\lambda}_{\nu+\gamma\nu}(s, s_1)\, P^{\lambda-\gamma\lambda}_{\nu+\gamma\nu+\gamma}(s, s_1)};$$
$$\pi_{ss_1}(\nu, \lambda, \gamma; \mu) = \prod_{\delta=1}^{\gamma} f_{ss_1}(\nu + \delta, \lambda)/f_{ss_1}(\mu, \lambda),$$

The subscript λ in the coefficients $\Pi_{\nu,\lambda;\gamma}^{\pm}(s, s_1)$, which are determined as they were in system (1.8) denote that the coefficients are expressed through the probabilities of one-quantum transitions. Considering that these probabilities depend weakly on the superscript, we find from (2.9) that

$$\begin{aligned} &\frac{d}{dt}\ln f_{ss_1}(\nu, \lambda) = J_{\nu,\lambda}^{(1)}(s, s_1) + J_{\nu,\lambda}^{(2)}(s, s_1) + \Gamma_{\nu,\lambda}(s, s_1), \\ &J_{\nu,\lambda}^{(1)} = \sum_{\gamma}\left[I_{\nu,\lambda;\gamma}^{(1)}(s, s_1 \mid \mathbf{f}) - I_{\nu-1,\lambda;\gamma}^{(1)}(s, s_1 \mid \mathbf{f})\right], \\ &J_{\nu,\lambda}^{(2)} = \sum_{\gamma}\mathbf{S}\left[I_{\nu,\lambda;\gamma}^{(2)}(s, s_1 \mid \mathbf{f}) - I_{\nu-1,\lambda;\gamma}^{(2)}(s, s_1 \mid \mathbf{f})\right], \\ &\Gamma_{\nu,\lambda}(s, s_1) = \dot{T}\frac{\partial}{\partial T}\ln\left[P_{\nu\nu-1}^{\lambda\lambda}(s, s_1)/P_{\nu-1\nu}^{\lambda\lambda}(s, s_1)\right]. \end{aligned} \tag{2.10}$$

The first two terms in the right side of (2.10) describe the degree to which inelastic collisions of molecules s and s_1 in states ν and λ contribute to changes in the variable $f_{ss_1}(\nu, \lambda)$ when there is a change in the state of only the molecule $s(J_{\nu,\lambda}^{(1)})$ or both molecules $(J_{\nu,\lambda}^{(2)})$. The last term plays the role of "hydrodynamic source."

By analogy with (1.8), $J_{\nu,\lambda}^{(1)}$ can be written in the form of the sum of the diagonal $R_{ss_1}(f_{ss_1}(\nu, \lambda))$ and the nondiagonal $E_{ss_1}(\mathbf{f}_{ss_1})$ (with respect to ν) parts having the structures (1.9) and (1.10), respectively. Here, the coefficients in the expressions are determined through the quantity $\Pi_{\nu,\lambda;\gamma}^{\pm}(s, s_1)$. The term $J_{\nu,\lambda}^{(2)}$ contributes only to the part which is diagonal with respect to $\mathbf{f}$.

Thus, in the given approach, allowance for intermolecular exchange does not alter the structure of Eqs. (1.8). It gives only the contribution to the component in their right side which is nondiagonal with respect to $\mathbf{f}$. For smooth initial distributions ($f_{ss_1}(\nu, \lambda)$ ~ f), the effect of the nondiagonal component on the relaxation process can be considered only within the framework of perturbation theory or by the iteration method. Here, as the zeroth approximation it is natural to use the solution of the system which is diagonal with respect to $\mathbf{f}$

$$\frac{d}{dt}\ln f_{ss_1}(\nu, \lambda) = R_{ss_1}\left(f_{ss_1}(\nu, \lambda)\right) + \Gamma_{\nu,\lambda}(s, s_1). \tag{2.11}$$

In a number of cases, this solution can be found analytically [7, 8].

In contrast to the first approach, here Eqs. (2.10)-(2.11) need not be supplemented by a system of equations for the moments of a distribution function. First of all, this opens up the possibility of analyzing general laws governing relaxation processes with an arbitrary dependence of the probabilities of inelastic collisions on the quantum numbers. Secondly, it significantly simplifies both analytical and numerical integration of system (2.10) for specific relaxation problems [11]. Now the latter do not have to be linked with integration of a system for moments (for example, for the store of vibrational quanta in the case of vibrational relaxation). However, this approach requires a much smoother distribution function compared to the first approach [due to the presence of ratios of the form $f_{ss_1}(\nu, \lambda)/f_{s_1s}(\lambda, \nu)$ in $J_{\nu,\lambda}^{(2)}$], and it appreciably increases the size of the initial system of equations.

System (2.10) must be supplemented by equations for $z_{ss_1}(0, \lambda)$. With $\nu = 0$, it follows from (2.5) that

$$\dot{y}_{ss_1}(0, \lambda) = \sum_{\gamma}\sum_{i=1,2}\mathbf{S}I_{0,\lambda;\gamma}^{(i)}(s, s_1 \mid \mathbf{y}).$$

Proceeding successively to variables (2.6) and the adiabatic variables, we obtain

$$\begin{gathered} \frac{d}{dt}\ln z_{ss_1}(0, \lambda) + \left[\varepsilon_s(0) - \langle\varepsilon_s\rangle + \varepsilon_{s_1}(\lambda) - \langle\varepsilon_{s_1}\rangle\right]\frac{d}{dt}\ln T = \\ = \sum_{\gamma}\Big\{\Pi_{0,\lambda;\gamma}^{+}(s, s_1)\prod_{\delta=1}^{\gamma} f_{ss_1}(\delta, \lambda) - P_{0\gamma}^{\lambda\lambda}(s, s_1) + \Pi_{\lambda,0;\gamma}^{+}(s_1, s)\,\pi_{s_1s}(\lambda, 0, \gamma; \lambda)\, f_{s_1s}^{\gamma}(\lambda, 0) - \\ - P_{\lambda\lambda+\gamma}^{00}(s_1, s) - P_{\lambda\lambda-\gamma}^{00}(s_1, s) + \Pi_{\lambda,0;\gamma}^{-}(s_1, s)\,\pi_{s_1s}^{-\gamma}(\lambda-\gamma, 0, \gamma; \lambda)\, f_{s_1s}^{-\gamma}(\lambda, 0) + \\ + P_{0\gamma}^{\lambda\lambda-\gamma}(s, s_1)\left[\Phi_{0,\lambda;\gamma}(s, s_1)\prod_{\delta=1}^{\gamma} f_{ss_1}(\delta, \lambda)/f_{s_1s}(\lambda-\gamma+\delta, \gamma) - 1\right]\Big\}. \end{gathered} \tag{2.12}$$

Finally, to make the reverse transition from adiabatic variables to concentrations, we need to successively use definitions (2.8), (2.6), (2.4)

$$y_{ss_1}(\nu,\lambda)=x_s(\nu)\,x_{s_1}(\lambda)=\overset{B}{x}_s(\nu)\,\overset{B}{x}_{s_1}(\lambda)\,z_{ss_1}(0,\lambda)\prod_{\delta=1}^{\nu}b_{\delta,\lambda}(s,s_1)\,f_{ss_1}(\delta,\lambda),$$

From this, with allowance for normalization of the concentrations [by virtue of (1.2)], we have

$$x_s(\nu)=\sum_{\lambda,s_1}y_{ss_1}(\nu,\lambda)=\overset{B}{x}_s(\nu)\sum_{\lambda,s_1}\overset{B}{x}_{s_1}(\lambda)\,z_{ss_1}(0,\lambda)\prod_{\delta=1}^{\nu}b_{\delta,\lambda}(s,s_1)\,f_{ss_1}(\delta,\lambda). \tag{2.13}$$

3. Relaxation in a Boltzmann Thermostat. One-Quantum Approximation. In the above discussion, we did not presume satisfaction of the principle of a detailed balance between the probabilities of inelastic transitions. This makes it possible to use the above-described methods to study isolated modes of polyatomic molecules or the relaxation of gas mixtures in a non-Boltzmann thermostat (such as in the presence of sources of excited particles [7, 9]). If the principle of detailed balance is satisfied (Boltzmann thermostat), i.e.,

$$P_{\mu\nu}^{\varkappa\lambda}(s,s_1)\exp\left[-\left(\varepsilon_s(\mu)+\varepsilon_{s_1}(\varkappa)\right)\right]=P_{\nu\mu}^{\lambda\varkappa}(s,s_1)\exp\left[-\left(\varepsilon_s(\nu)+\varepsilon_{s_1}(\lambda)\right)\right], \tag{3.1}$$

then the structure of the equations obtained in Part 2 is simplified greatly because in this case $b_{\nu,\lambda}(s,\ s_1)=1$, $\Pi_{\nu,\lambda;\gamma}{}^{+}(s,\ s_1)=P_{\nu\nu+\gamma}{}^{\lambda\lambda}(s,\ s_1)$, $\Pi_{\nu,\lambda;\gamma}{}^{-}(s,\ s_1)=P_{\nu\nu-\gamma}{}^{\lambda\lambda}(s,\ s_1)$, $\Phi_{\nu,\lambda;\gamma}(s,\ s_1)=1$. Here, Eq. (2.13) for the reverse transition from adiabatic variables to concentrations takes the form

$$x_s(\nu)=\overset{B}{x}_s(\nu)\sum_{\lambda,s_1}z_{ss_1}(0,\lambda)\,\overset{B}{x}_{s_1}(\lambda)\prod_{\delta=1}^{\nu}f_{ss_1}(\delta,\lambda). \tag{3.2}$$

Considering that the probabilities of transitions generally decrease rapidly with an increase in the number of transmitted quanta, a solution to the equations of vibrational kinetics in the zeroth approximation can be found within the framework of the model of one-quantum transitions. Then with satisfaction of Eq. (3.1) in the first approach, we obtain a system of the form (1.8) with $\gamma=1$ for the functions $f_s(\nu)$. In this system, the effective probabilities (2.1) are determined by the equalities

$$\begin{gathered}P_{\nu+1\nu}^{*}(s)=\sum_{s_1}\left[P_{\nu+1\nu}(s,s_1)+\sum_{\varkappa}P_{\nu+1\nu}^{\varkappa-1\varkappa}(s,s_1)\,x_{s_1}(\varkappa-1)\right],\\ P_{\nu\nu+1}^{*}(s)=\sum_{s_1}\left[P_{\nu\nu+1}(s,s_1)+\sum_{\varkappa}P_{\nu\nu+1}^{\varkappa\varkappa-1}(s,s_1)\,x_{s_1}(\varkappa)\right],\end{gathered} \tag{3.3}$$

while in the second approach system (2.10)-(2.13) reduces to equations for $f_{ss_1}(\nu,\ \lambda)$ (the index $\gamma=1$ will henceforth be omitted):

$$\begin{gathered}\frac{d}{dt}\ln f_{ss_1}(\nu,\lambda)=A_{\nu,\lambda}(s,s_1)\,f_{ss_1}(\nu,\lambda)+B_{\nu,\lambda}(s,s_1)+\\ +C_{\nu,\lambda}(s,s_1)\,f_{ss_1}^{-1}(\nu,\lambda)+\sum_{i=1,2}E_{\nu,\lambda}^{(i)}(s,s_1)+\Gamma_{\nu,\lambda}(s,s_1),\end{gathered} \tag{3.4}$$

where

$$\begin{gathered}A_{\nu,\lambda}(s,s_1)=P_{\nu\nu+1}^{\lambda\lambda}(s,s_1)-P_{\nu-1\nu}^{\lambda\lambda}(s,s_1);\ C_{\nu,\lambda}(s,s_1)=P_{\nu\nu-1}^{\lambda\lambda}(s,s_1)-P_{\nu-1\nu-2}^{\lambda\lambda}(s,s_1);\\ B_{\nu,\lambda}(s,s_1)=-\left[A_{\nu,\lambda}(s,s_1)+C_{\nu,\lambda}(s,s_1)\right];\ \Gamma_{\nu,\lambda}(s,s_1)=\\ =-\left[\varepsilon_s(\nu)-\varepsilon_s(\nu-1)\right]\frac{d}{dt}\ln T;\end{gathered}$$

$$\begin{gathered}E_{\nu,\lambda}^{(1)}(s,s_1)=f_{ss_1}(\nu,\lambda)\,P_{\nu\nu+1}^{\lambda\lambda}(s,s_1)\left[f_{ss_1}(\nu+1,\lambda)/f_{ss_1}(\nu,\lambda)-1\right]-\\ -f_{ss_1}^{-1}(\nu,\lambda)\,P_{\nu-1\nu-2}^{\lambda\lambda}(s,s_1)\left[f_{ss_1}(\nu,\lambda)/f_{ss_1}(\nu-1,\lambda)-1\right];\\ E_{\nu,\lambda}^{(2)}(s,s_1)=P_{\nu\nu+1}^{\lambda\lambda-1}(s,s_1)\left[f_{ss_1}(\nu+1,\lambda)/f_{s_1s}(\lambda,\nu+1)-1\right]-\\ -P_{\nu-1\nu}^{\lambda\lambda-1}(s,s_1)\left[f_{ss_1}(\nu,\lambda)/f_{s_1s}(\lambda,\nu)-1\right]+P_{\nu\nu-1}^{\lambda\lambda+1}(s,s_1)\left[f_{s_1s}(\lambda+1,\nu)/f_{ss_1}(\nu,\right.\\ \left.\lambda+1)-1\right]-P_{\nu-1\nu-2}^{\lambda\lambda+1}(s,s_1)\left[f_{s_1s}(\lambda+1,\ \nu-1)/f_{ss_1}(\nu-1,\lambda+1)-1\right],\end{gathered}$$

and to equations for $z_{ss_1}(0, \lambda)$:

$$\frac{d}{dt}\ln z_{ss_1}(0,\lambda)+\left[\varepsilon_s(0)-\langle\varepsilon_s\rangle+\varepsilon_{s_1}(\lambda)-\langle\varepsilon_{s_1}\rangle\right]\frac{d}{dt}\ln T=$$
$$=P_{01}^{\lambda\lambda}(s,s_1)\left[f_{ss_1}(1,\lambda)-1\right]+P_{\lambda\lambda+1}^{00}(s_1,s)\left[f_{s_1s}(\lambda+1,0)-1\right]+$$
$$+P_{\lambda\lambda-1}^{00}(s_1,s)\left[f_{s_1s}^{-1}(\lambda,0)-1\right]+P_{01}^{\lambda\lambda-1}(s,s_1)\left[f_{ss_1}(1,\lambda)/f_{s_1s}(\lambda,1)-1\right]. \tag{3.5}$$

In the zeroth approximation with respect to $E_{\nu,\lambda}$ and $\Gamma_{\nu,\lambda}$, we can use (3.4) to obtain Riccati's general equation with a zero coefficient sum. The general solution of this equation can be written in quadratures with arbitrary transition probabilities [7].

4. Example: One-Component Gas, Harmonic Approximation. To compare the methods described in Part 2 for changing over to adiabatic variables in the quadratic part of the collision integral, we will examine the following model problem: thermalization of a vibrationally excited (with $T_V \neq T$) gas of diatomic molecules modeled by harmonic oscillators, assuming satisfaction of condition (3.1). Since only one-quantum transitions with the probabilities [1]

$$P_{\nu+1\nu}=(\nu+1)P_{10},\ P_{\nu\nu+1}=(\nu+1)P_{01},$$
$$P_{\nu+1\nu}^{\varkappa-1\varkappa}=\varkappa(\nu+1)Q_{10},\ P_{\nu\nu+1}^{\varkappa\varkappa-1}=\varkappa(\nu+1)Q_{10}, \tag{4.1}$$

are allowed in a harmonic approximation, we will use the simplified equations in Part 3 as the initial equations.

In the first approach (Part 1A), if we consider that, due to (3.3) and (4.1), the effective probabilities in the given situation have the form

$$P_{\nu+1\nu}^{*}=(\nu+1)[P_{10}+Q_{10}(\alpha+1)],\ P_{\nu\nu+1}^{*}=(\nu+1)[P_{01}+Q_{10}\alpha]$$

$\left(\alpha\equiv\sum_{\varkappa}\varkappa x(\varkappa)\right.$ is the store of vibrational quanta), for the variables $x(0)$, $f(\nu)$ $(\nu \geq 1)$ we obtain

$$\frac{d}{dt}\ln f(\nu)=Af(\nu)+B+Cf^{-1}(\nu)+E_\nu(\mathbf{f})+\Gamma,\ \frac{d}{dt}\ln x(0)=A(f(1)-1), \tag{4.2}$$

where

$$A=P_{01}+\alpha Q_{10};\ C=P_{10}+(\alpha+1)Q_{10};\ B=-(A+C);\ \Gamma=-\frac{d}{dt}\ln\frac{A}{C};$$
$$E_\nu(f)=A(\nu+1)f(\nu)[f(\nu+1)/f(\nu)-1]-C(\nu-1)f^{-1}(\nu)[f(\nu)/f(\nu-1)-1].$$

If the initial distribution is a Boltzmann distribution with a nonequilibrium vibrational temperature $T_V{}^0$, i.e., if

$$x(\nu\,|\,t=0)=x^{\mathrm{B}}(\nu\,|\,T_V^0)=\left[1-\exp(-\Theta_V^0)\right]\exp(-\nu\Theta_V^0) \tag{4.3}$$

($\Theta_V = \hbar\omega_e/k_BT_V$), then we need to supplement this system with the initial conditions

$$f(\nu\,|\,t=0)=f^0(\nu)=(C^0/A^0)\exp(-\Theta_V^0),\ x(0\,|\,t=0)=x^0(0)=1-\exp(-\Theta_V^0). \tag{4.4}$$

It is not hard to show that for distributions of the form (4.3) the nondiagonal term $E_\nu(\mathbf{f})$ in the first equation is identically equal to zero. Thus, this equation (with allowance for the form of the "source term" Γ) can be rewritten in the form

$$\frac{d}{dt}\ln g(\nu)=Cg(\nu)+B+Ag^{-1}(\nu) \tag{4.5}$$

($g(\nu) \equiv Af(\nu)/C$, $g^0(\nu) = \exp(-\Theta_V{}^0)$). Since the coefficients in (4.5) satisfy the condition $A + B + C \equiv 0$, the solution of Cauchy problem (4.2), (4.4) is readily calculated in quadratures (see [7, 8]). Omitting this solution here, we note only that since neither the initial data nor the coefficients in (4.5) depend on ν, the form of initial distribution (4.4) remains unchanged in the course of the relaxation process in this case (this is the well-known principle of canonical invariance). The only changes are in the vibrational T_V and translational T temperatures of the gas. By virtue of the energy conservation law, these quantities are connected by the relation

$$\Theta^{-1} + \frac{2}{5}(\exp\Theta_V - 1)^{-1} = \text{const}, \; \Theta = \hbar\omega_e/k_B T, \tag{4.6}$$

while the Cauchy solution that was found is the integral equation for $\Theta_V(t)$ [since $\Theta_V(t) = -\ln g(t)$]. This equation can easily be used to obtain the familiar equation for the store of vibrational quanta

$$\dot{\alpha} = P_{10}(\Theta(\alpha))[(1+\alpha)\exp(-\Theta(\alpha)) - \alpha], \; \alpha = (\exp\Theta_V - 1)^{-1}, \tag{4.7}$$

where the relation $\Theta(\alpha)$ is determined by Eq. (4.6).

Within the framework of the second approach (Part B), we obtain the below Cauchy problem for functions f from (3.4)-(3.5), probabilities of the form (4.1), and initial conditions (4.3) in the case of a one-component gas

$$\frac{d}{dt}\ln f(v) = A_\lambda f(v) + B_\lambda + C_\lambda f^{-1}(v) + \frac{d}{dt}\ln\frac{C}{A}; \; f^0(v) = \exp(\Theta^0 - \Theta_V^0), \tag{4.8}$$

where $A_\lambda = P_{01}{}^{\lambda\lambda}$; $C_\lambda = P_{10}{}^{\lambda\lambda}$; $B_\lambda = -(A_\lambda + C_\lambda)$ and we considered that with satisfaction of the principle of detailed balance

$$f(v, \lambda) = \frac{C_\lambda}{A_\lambda}\frac{x(v)}{x(v-1)} = e^{\Theta}\frac{x(v)}{x(v-1)} = f(v).$$

As above, Eq. (4.8) is easily integrated in quadratures, while the absence of a dependence on v in the coefficients and initial conditions again leads to the principle of canonical invariance. Here, however, since the coefficients in (4.8) are not explicitly dependent on α, the solution obtained in the Cauchy problem determines the time dependence $\Theta_V(t)$ if $\Theta(t)$ is given by external conditions. If $\Theta(t)$ and $\Theta_V(t)$ are connected by law (4.6), then the two approaches prove to be equivalent.

Thus, two methods have been proposed for describing relaxation processes in mixtures of molecular gases in terms of adiabatic variables. The advantages and disadvantages of each were discussed. The changeover to adiabatic variables in the kinetic equation makes it possible to establish more general relaxation laws for arbitrary models of rate coefficients [8, 9], obtain approximate analytic solutions to these equations [7, 8], and construct effective algorithms for their numerical integration [11]. Using the one-quantum transition approximation, we presented the simplest form of the relaxation equations in adiabatic variables for relaxation of a gas mixture in a Boltzmann thermostat. The use of these approaches was illustrated by means of a simple model problem on the vibrational relaxation of diatomic harmonic molecules in a Boltzmann thermostat.

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OSCILLATIONS IN THE PARALLEL DISCHARGE OF TWO SUPERSONIC NONISOBARIC JETS

S. G. Mironov

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Oscillating flows in systems of parallel supersonic nonisobaric jets discharged into a submerged space [1-12] are of considerable interest from the viewpoint of practical applications of this phenomenon and the development of models of transients in supersonic jets. Worth noting among the different studies in this area is [2], where for a system of parallel jets and jets parallel to a shield the author obtained the first data on the possible modes of vibration, their relative intensities, and the region of Mach numbers in which they exist. The authors of [4-12] presented the most complete empirical results on the modal composition of acoustic vibrations, their intensities, and the types of oscillations of the wave structure of the jets in relation to the discharge parameters and the interlaminar distance in two-jet systems. This information was compared with the analogous characteristics of single jets. However, no physical model has yet been presented to describe these characteristics in systems of jets.

In the present study, we obtain new empirical results for two parallel jets and propose models to describe the directionality of the acoustic radiation and the conditions for the excitation of oscillations.

1. Two parallel jets were created by means of supersonic conical nozzles with an outlet section having a diameter $d_a = 1.4 \cdot 10^{-2}$ m. We used nozzles with the Mach numbers M_a = 1, 1.5, 2, and 3.7 and the cone angle 9°. Compressed air from the prechamber was directed to smoothly change the distance between nozzles $\bar{S} = S/d_a$ from 1.8 to 7 and to displace the nozzles longitudinally relative to each other. We could remove one nozzle and in the middle of the inter-nozzle gap install a flat metal shield parallel to the jet axis. The shield measured 0.15 × 0.3 m.

The acoustic pressure pulsations were measured with two piezoelectric sensors. The diameter of their receiving part was $3 \cdot 10^{-3}$ m, while the limiting measurement frequency was 60 kHz. One sensor was placed on the line of centers of the nozzles midway between the latter. This sensor was positioned 3 diameters downflow of the nozzle edges. The constancy of the middle location of the sensor was assured by installing it at the center of a strip of rubber attached to the nozzles. To determine the directionality of the acoustic radiation in the plane perpendicular to the jets, the second sensor was positioned so that it could turn about its axis. This axis was parallel to the jets and passed through the center of the nozzle spacing at the level of the first sensor. The radius of rotation was 7 nozzle diameters.

Information on vibrations of the wave structure of the jets was obtained with IAB-451 shadowgraph equipped with an ISSh-15 stroboscopic lamp that was synchronized with the acoustic pressure pulsations. This made it possible to obtain images of the vibration phases with a high degree of averaging over random fluctuations of the flow field.

2. The first three schlieren photographs in Fig. 1 show the three main types of flows that were recorded experimentally. These flows exist during oscillations of two parallel jets in the system. The photographs correspond to the following discharge conditions: 1) $M_a = 1$, $n = 2.37$, $\bar{S} = 2.5$; 2) $M_a = 1$, $n = 1.83$, $\bar{S} = 5.15$; 3) $M_a = 1$, $n = 1.32$, $\bar{S} = 1.8$. It is evident from the first photograph that the wave structure of the jets undergoes flexural

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