

ROTATIONAL RELAXATION OF MOLECULAR NITROGEN IN A FREELY EXPANDING JET

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In connection with the assumption about the possibility of producing a gasdynamic laser at rotational transitions of diatomic molecules [1, 2] and a gasdynamic condense-laser by using the phenomenon of condensation to produce the population inversion [3-5], a quantitative description of the kinetics of the rotational relaxation of the simplest diatomic molecules is necessary. In contrast to the traditional approach in gasdynamics and the theory of transport processes, when one parameter (the rotational relaxation time) is used in the description of rotational relaxation, a substantially more detailed description at the population level of the individual rotational states is required in solving spectroscopy and laser physics problems. This paper is devoted to a theoretical and experimental investigation of the rotational relaxation of nitrogen in a free, low density jet under conditions when a substantial nonequilibrium holds in the rotational level populations. On the basis of representations developed earlier [1, 6], a model is constructed for a relaxing gas which yields the magnitude of the population of individual rotational levels. The selection of a molecular nitrogen free jet as a subject for investigation is explained by the fact that the gasdynamics of such a flow has been studied well [7]. Moreover, at this time diagnostic methods have been developed to determine the molecule concentrations at many ($k \leq 20$) rotational levels [8]; hence, a jet is a good object on which a detailed comparison between theory and experiment can be made, as is done in this paper. Separation of the translational and rotational relaxation processes is allowed in the theoretical description of the flow with relaxation studied in this paper on the basis of the fact that the buildup of a Maxwell molecule velocity distribution because of elastic collisions occurs more rapidly than the redistribution of molecules at the rotational levels because of inelastic collisions. Such an approach is apparently valid for molecular hydrogen with a large value of the rotational quantum [9]. A model with separation of the processes is hypothetical for nitrogen molecules whose rotational constant is $\approx 1/20$ that for hydrogen, and is confirmed in this paper by comparing computations with experiment. It is hence assumed that rotational relaxation proceeds in an N_2 molecule stream with a known temperature, density, and velocity distribution obtained from measurements.

1. Data on the rotational level populations and the gasdynamic parameter distribution have been obtained under conditions when the population kinetics is due to rotational relaxation and the influence of the background [8], the condensation [4], and the effects of freezing of the translational temperature can be neglected. The investigation is performed in the low-density gasdynamic tube of the Institute of Thermophysics, Siberian Branch of the Academy of Sciences of the USSR [10], provided with electron-beam and molecular-beam diagnostics to measure the density, population of the rotational levels [8], and the molecule velocity distribution function [11]. Sonic nozzles of 0.5-5-mm diameter were used with a ratio of the nozzle edge thickness to diameter less than 0.02. The Reynolds number according to the critical section parameters was always greater than 100, which afforded a foundation for neglecting the influence of the boundary layer in the nozzle. The stagnation temperature in the nozzle fore-chamber was room temperature ($T_0 = 292^\circ\text{K}$) in all the experiments, and the working gas was technically pure nitrogen.

Because of interaction of the gas emerging from the nozzle with the gas in the surrounding space, a background is formed from gas molecules which penetrate into the core of the jet and the molecules in the jet

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colliding with these latter and losing the velocity of directed motion [12]. The population distribution of the rotational levels of the background molecules usually corresponds to a higher temperature than the temperature in the jet. Consequently, the influence of the background turns out to be most noticeable in population measurements for the upper levels, which are destroyed by a relaxation process during expansion. As has been noted in [8], a moderate quantity ($\leq 1\%$) of hotter gas, which is negligible in the determination of the low level populations, changes the population of the upper levels significantly. Taking account of the influence of the background was experimentally accomplished. To do this, measurements were performed at the maximum distance from the nozzle exit, i.e., where the influence of the background is most substantial, for different pressures in the surrounding space p_H . Results of the measurements are presented in Fig. 1 for $p_0 d_* = 493$ mm Hg (p_0 is the stagnation pressure and d_* is the diameter of the critical nozzle section), where ρ_k is the population of the k -th rotational level normalized relative to the gas density $\sum_0^{\infty} \rho_k$ and p_H is the pressure in the surrounding space, mm Hg. As is seen, the influence of the background becomes negligible for $p_H < 5 \cdot 10^{-3}$ mm Hg for levels with $k < 8$. Let us note that the influence of the background for $p_0 d_* < 493$ reduces the size of the jet section suitable for comparison with computation.

Supersaturation was achieved at all stagnation pressures at spacings x/d_* from the nozzle exit (x is the spacing from the nozzle exit) between 2.5 and 3.5; i.e., condensation is possible in the stream in the measurement domain. The population kinetics of rotational levels in a condensing gas stream differs significantly from that when the effect of condensation is negligible [4, 5]. As is known the similarity parameter for rotational relaxation in a homogeneous gas jet at constant stagnation temperature is $p_0 d_*$, but the condensation is $p_0 d_*^n$; according to the data in [13], $n = 0.55$ for nitrogen. Hence, the change in d_* for a constant value of $p_0 d_*$ can check on the influence of condensation. The result of such a check for a maximum value of $p_0 d_*$ is shown in Fig. 2a and b where the population temperatures of the k -th level are presented:

$$T_k = -B_0 k(k+1) / \ln [g_0 \rho_k / (g_k \rho_0)],$$

B_0 is the rotational constant, ρ_k , g_k are the population and partition function of the k -th state. The measurements were performed in four nozzles, $d_* = 0.54, 2, 2.11,$ and 5 mm (points 1-4, respectively). The data obtained for identical $p_0 d_*$ and different nozzle diameters agree, indicating the negligible influence of condensation on the population kinetics for a given $p_0 d_*$ but also for values of $p_0 d_*$ less than mentioned. The tendency to an increase in the population temperature with the increase in stagnation pressure, which is apparently associated with the effect of condensation, should be noted.

Disturbance of the Maxwell distribution which takes place during expansion affects the density and velocity slightly, but results in uncertainties in giving the translational temperature. To clarify this question, the perpendicular temperature T_{\perp} , obtained by the molecular beam method [11] (Fig. 3, points 2), the translational temperature T_t estimated by means of the low level population temperatures [12] (the points 1), and the results of an isentropic computation for the ratio between the specific heats $\gamma = 1.4$ (solid line) were compared for $p_0 d_* = 493$. As is seen, for $x/d_* \leq 30$ the perpendicular temperature and the estimated temperature T_t agree within the limits of measurement error. For values of x/d_* greater than those mentioned, the data diverge, which is caused by disturbance of the Maxwell velocity distribution. The isentropic temperature for $x/d_* < 30$ lies somewhat below the experimental results in T_t . But for this $p_0 d_*$ this difference is less than 2°K , and can henceforth be neglected.

It can be expected that for low values of $p_0 d_*$ the gas will expand as a monatomic gas with corresponding values of the velocity, density, and temperature because of freezing of the rotational relaxation. As is seen, this effect is not detected for $p_0 d_* \geq 10$ from results of measuring the density ρ normalized relative to the stagnation density ρ_0 represented in Fig. 4 (values of x/d_* are given by the digits). The relative density ρ/ρ_0 at a constant distance from the nozzle exit is independent of $p_0 d_*$. It follows from the continuity equation that rotational and translational relaxations exert no noticeable influence on the stream velocity also in the $p_0 d_*$ range considered.

In addition to the experimental data, results of an isentropic computation for $\gamma = 1.4$ are superposed by solid lines in Fig. 4. The experimental data and theoretical computation do not agree. Moreover, this difference increases with the approach to the nozzle exit. As is shown in [12], the computed and experimental values of the density agree if the location of the sonic section $x/d_* = 0$ is taken shifted by 0.6 upstream and the diameter of the critical section is taken at 0.943 of the geometric diameter. The same correction is needed for agreement between the measurement data of the static pressure [14] and is caused by the fact that the location and actual shape of the sonic section differ from those given in the computation. Agreement between the computed and experimental data with the corrections mentioned yields the right to give the density isentropically.

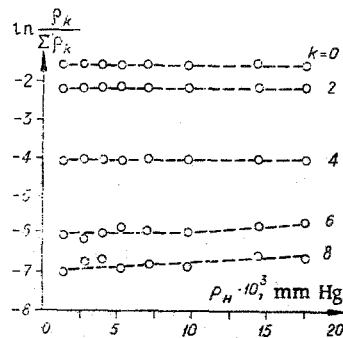


Fig. 1

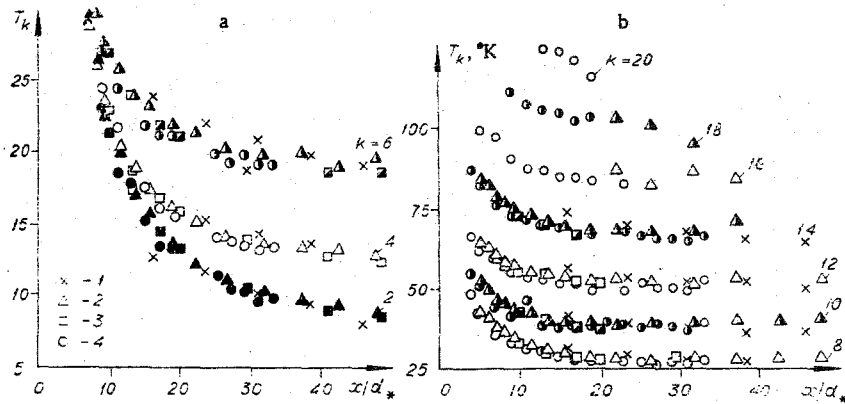


Fig. 2

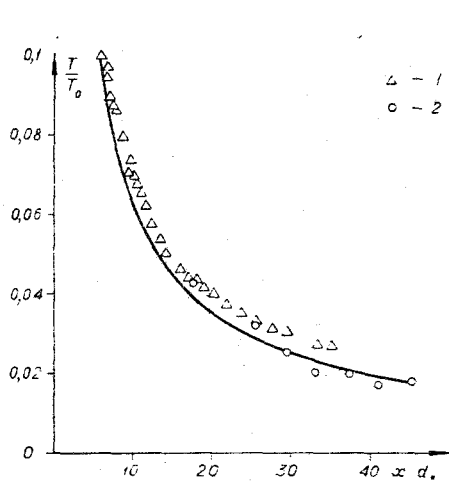


Fig. 3

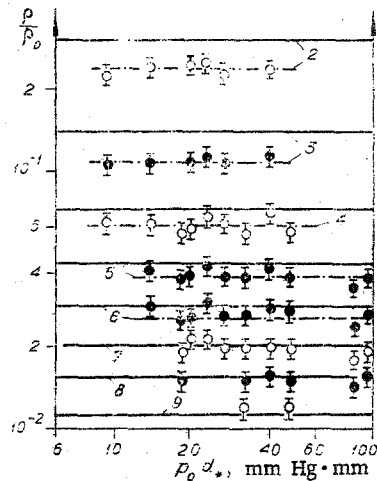
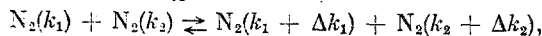


Fig. 4

For $x/d_* < 3.2$ results of an isentropic computation were approximated by a fourth power polynomial, and an approximate Sherman formula [14] for the Mach number was used with $x/d_* > 3.2$; furthermore, the density and velocity were calculated by means of known isentropic formulas. The agreement between the experimental data and the isentropic computation (with the corrections mentioned) for $p_0 d_* = 493$ up to $x/d_* \approx 30$ affords the possibility of using isentropic relationships even for the computation of the translational temperature, but only for $p_0 d_* > 493$. For values of $p_0 d_*$ less than that mentioned, the temperature obtained by extrapolating the population temperature to the zero value of the rotational quantum number is taken as the translational temperature.

2. Let us consider the collisions between molecules to be accompanied by inelastic processes



where the numbers of the rotational levels of the ground electron and vibrational states of the N_2 molecule are written in the parentheses; $\Delta k_1, \Delta k_2$ are their change during the collision act. Such inelastic collisions are

accompanied by energy exchange between the translational and rotational degrees of freedom. The quantum states of one (rt processes) or both (rr and rr' processes) of the colliding molecules hence vary. Let us note that the exchange rr' processes ($k_1 + \Delta k_1 = k_2$; $k_2 + \Delta k_2 = k_1$) do not result in a change in the state of the system of rotational levels and, hence, are not considered henceforth.

Keeping the low gas temperatures ($T < 200^\circ\text{K}$) in mind in the region of comparison between experiment and computation, we limit ourselves just to two-quantum rt processes ($k_1 = \pm 2$, $k_2 = 0$, or conversely) and two-quantum rr' processes for which $\Delta k_1 = \Delta k_2$.

The rotational transition probabilities $P_{\substack{k_1, k_1 + \Delta k_1 \\ k_2, k_2 + \Delta k_2}}$ are computed in an approximation of exponential form of the spherical part of the interaction potential [6, 15]

$$V = \exp(-\alpha R) \{ B + B_{20} [P_2(\cos \chi_1) + P_2(\cos \chi_2)] + B_{22} P_2(\cos \chi_1) P_2(\cos \chi_2) \},$$

where R is the spacing between the centers of mass of the colliding molecules; χ_i , angle between the molecular axis of the i -th molecule and the line connecting the centers of mass of the colliding molecules; $P_2(\cos \chi_i)$, Legendre polynomial of second degree; α , a parameter characterizing the form of the potential. The coefficients B , B_{20} , B_{22} appear in the expansion of the sum of the interaction potentials between the individual atoms in the molecule into series in the Legendre polynomials $P_n(\cos \chi_i)$ [15]. The coefficients B_{ij} in such an expansion are just certain proportionality factors.

The two-quantum rt and rr' transition probabilities computed in a quasiclassical approximation are, according to [15],

$$P_{\substack{k_1, k_1 - \Delta k_1 \\ k_2, k_2 + \Delta k_2}}(T) = \frac{4\pi^2 \mu^2 |\Delta \Sigma|^2 (B_{ij})^2}{\hbar^4 \alpha^4} \times \Phi(k_1 \rightarrow k_1 - \Delta k_1, k_2 \rightarrow k_2 + \Delta k_2) \sqrt{\frac{\pi}{3}} (A_{ij}/2\sqrt{kT})^{1/3} \exp[-3(A_{ij}/2\sqrt{kT})^{2/3}] (1 + e^{\Delta \Sigma/2kT})^2, \quad (2.1)$$

where $A_{ij} = \pi |\Delta \Sigma| / \hbar \alpha \sqrt{2\mu}$; $\Delta \Sigma = \sum_{i=1,2} k B_0 \Delta k_i (\Delta k_i + 2k_i + 1)$; k , Boltzmann constant; μ , apparent mass of the colliding molecules; $\Delta k_1, \Delta k_2 = 0, \pm 2$; T , gas temperature; \hbar , Planck constant;

$$\Phi(k_1 \rightarrow k_1 - 2, k_2) = \frac{9}{40} \frac{k_1(k_1 - 1)}{(2k_1 - 1)(2k_1 + 1)};$$

$$\Phi(k_1 \rightarrow k_1 - 2, k_2 \rightarrow k_2 + 2) = \frac{81}{500} 0.43 \frac{k_1(k_1 - 1)(k_2 + 1)(k_2 + 2)}{(2k_1 - 1)(2k_1 + 1)(2k_2 + 1)(2k_2 + 3)}.$$

The frequencies of the two-quantum collisional rotational rt and rr' transitions reduced (to atmospheric pressure) for the molecule N_2 are given in Fig. 5 for three values of the temperature - 30, 70, and 150°K :

$$\tilde{Z}_k^{rt} = \nu \sum_{k_2} \eta_{k_2} P_{\substack{k_1, k_1 - 2 \\ k_2, k_2}}, \quad \tilde{Z}_k^{rr'} = \nu \sum_{k_2} (\eta_{k_2} P_{\substack{k_1, k_1 - 2 \\ k_2, k_2 + 2}} + \eta_{k_2 + 2} P_{\substack{k_1, k_1 + 2 \\ k_2 + 2, k_2}}),$$

where \tilde{Z} is the total number of elastic collisions of one molecule in unit time at normal pressure and η_k is the relative number of molecules in the state k . The \tilde{Z}_k^{rt} and $\tilde{Z}_k^{rr'}$ are shown in Fig. 5 by solid and dashed lines, respectively. The gas-kinetic section was assumed constant and equal to $\sigma_g = 4\sqrt{2}\pi r^2$, where r is the effective radius of elastic collision between molecules. Values of the quantities in (2.1) are taken as follows for a computation of the probabilities:

$$B = 2 \cdot 10^{-10} (10 + \alpha^2) \text{ erg}, \quad B_{20} = 2\alpha^2 10^{-10} \text{ erg},$$

$$B_{22} = 0.2 \cdot 10^{-10} \alpha^4 \text{ egg}, \quad r = 1.5 \text{ \AA}.$$

The characteristic singularities of the graphs represented are associated with the decrease in the transition frequency as the temperature decreases, with their decrease with the growth of k for the upper levels, and an opposite dependence in the low states. Let us also note that relatively large probabilities of the exchange processes at the upper rotational levels, averaged over the relative populations of the levels η_k , yield a moderate contribution to the total probability of a change in a given state (especially for large values of α) for a Boltzmann distribution. This is due to the small fraction of particles η_k and the acute dependence of the probability on the energy defect of the process.

Unfortunately, real values of the coefficients in the anisotropic part of the potential are unknown up to this time [16]. Uncertainty also exists in the choice of the parameter α which can have, according to different experimental results [17], values within the limits $2-5 \text{ \AA}^{-1}$. In this connection, it is meaningful to speak about the dependence of the transition probabilities on k_1 and k_2 . For a given temperature this dependence will be

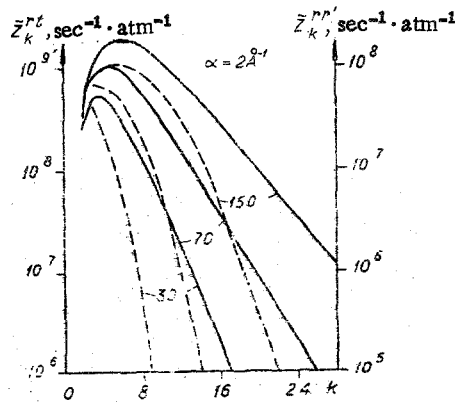


Fig. 5

determined by the quantity α . On the other hand, the mentioned uncertainties in the selection of the interaction potential can be allowed in a comparison between the results of a kinetic computation of the rotational relaxation and experimental data, whereupon values of the quantity α and the correction factor C are obtained for all the probabilities:

$$\bar{P}_{\substack{R_1, R_2 + \Delta R_1 \\ R_2, R_2 + \Delta R_2}} = CP_{\substack{R_1, R_2 + \Delta R_1 \\ R_2, R_2 + \Delta R_2}}$$

Let us note that a limitation of the kinds of inelastic processes by even values of Δk_1 and Δk_2 also corresponds to smallness of the molecule transition probabilities from one spin-modification to another. It is hence natural to expect that systems of even and odd rotational levels should vary independently. This latter assumption is verified by the results of experiments in both [8] and in this paper, where it is detected that the ratio between the intensities of the even and odd lines (2 : 1) is not disturbed under cooling during expansion.

We use the kinetic equation in the form

$$\partial \rho_k / \partial t + \text{div } \rho_k \mathbf{V}_k = St_k,$$

where $\mathbf{V}_k = \mathbf{v} + \mathbf{u}_k$; \mathbf{v} , velocity of the gas stream; \mathbf{u}_k , diffusion stream velocity (or the mean velocity of the motion of an individual component of the gas relative to the center of mass of a certain small volume); St_k , collision integral or the rate of change of the number of particles in the state k per unit volume because of inelastic collisions to describe the change in population in the rotational states. For a one-dimensional flow, the following equation can be obtained for the relative populations $\eta_k = \rho_k / \rho$:

$$\frac{d\eta_k}{dz} = \rho d/v (St_k / \rho^2) + (\rho v d_*)^{-1} \frac{d}{dz} \left(\rho D_k \frac{d\eta_k}{dz} \right), \quad (2.2)$$

where D_k is the coefficient of diffusion of molecules in the k -th state, $\bar{u}_k = -D_k \nabla \eta_k$; $z = 0.6 + 0.943x/d_*$ is the reduced coordinate along the flow axis. It follows from (2.2) that for an ideal gas with a given field of gasdynamic parameters, the quantity $\rho_0 d_* \sim \rho_0 d_*$ characterizes the dependence of the population on the coordinate $\eta_k(z)$. Furthermore, it can be concluded from a comparison of the first and second members in the right side of (2.2) that the diffusion term can be neglected for sufficiently large $\rho_0 d_*$. In this case, the equation under consideration can be converted into the following:

$$\frac{d\eta_k}{dz} = -\frac{vdC}{v} \sum_{\substack{\Delta k_1 = \pm 2 \\ \Delta k_2 = 0, \pm 2}} \sum_{h_2} \left(\eta_{h_1} \eta_{h_2} P_{\substack{R_1, R_1 + \Delta R_1 \\ R_2, R_2 + \Delta R_2}} - \eta_{h_1 + \Delta R_1} \eta_{h_2 + \Delta R_2} P_{\substack{R_1 + \Delta R_1, h \\ R_2 + \Delta R_2, h}} \right); \quad (2.3)$$

$$\sum_k \eta_k = 1, \quad (2.4)$$

where the sum in the right side of (2.3) is the model representation for the collision integral St , which satisfies the conservation laws for the numbers of particles and the energies; ν is the total number of elastic collisions between molecules per unit time. As is shown in Sec. 1, values of the density, velocity, and translational temperature can be given isentropically as functions of the Mach number $M(z)$. Results of an isentropic computation of $M(z)$ for the ratio $\gamma = 1.4$ between the specific heats were approximated by the dependences

$$M(z) = 1 + \sum_1^4 C_1(z)^i \quad (0.8 \leq z \leq 2.8),$$

$$M(z) = 3.65(z - 0.4)^{0.4} - 0.822(z - 0.4)^{0.6} (z - 0.4)^{1.2} \quad (z > 2.8),$$

where $C_1 = 0.366$, $C_2 = 2.462$, $C_3 = 1.515$, $C_4 = 0.278$.

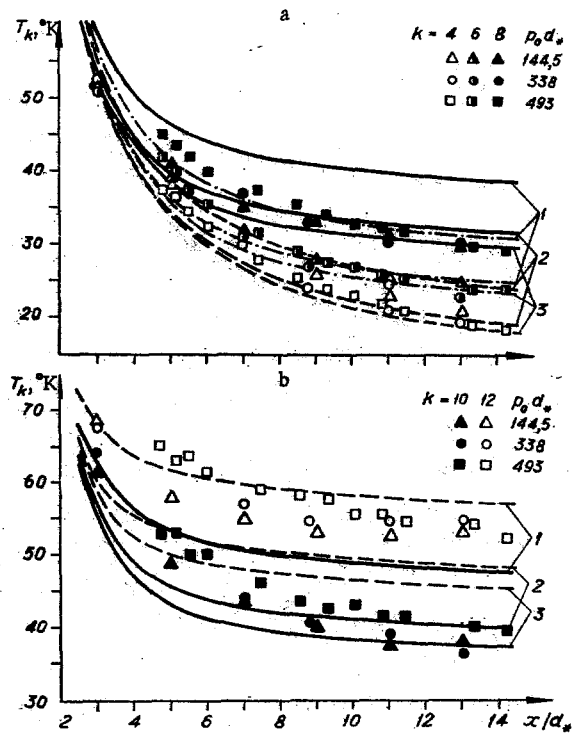


Fig. 6

The second formula is taken from [14]. The translation temperature for $p_0 d_* = 493$ and 338 was computed by isentropic relationships, while a correction taking account of the deviation of the measured from the isentropic temperature was inserted for $p_0 d_* = 144$.

The system of differential equations (2.3) under the conditions (2.4) was solved numerically (on a BÉSM-6 electronic computer) by the Runge-Kutta method with a given relative accuracy of $\varepsilon = 10^{-3}$. The number of equations was determined by the condition $3 \leq k \leq k^*$. The value of the number k in the different versions varied between 30 and 40 so as to diminish, as possible, the influence of the "boundary" of the block of equations on the result of the computation. The relative populations of the low levels were given by the temperature of the distribution T_k , equal to the translational temperature

$$\eta_k = \eta_0 g_k / g_0 \exp [-B_0 k(k+1)/T_k] \quad (k = 1; 2),$$

where the partition functions $g_k = (2+1)g_S$; $g_S = 1/3$ for odd and $g_S = 2/3$ for even values of k ; the populations of the levels $k > k^*$ were computed by means of the Boltzmann law with a temperature equal to the population temperature at the upper levels of the system k^* and $k^* - 1$. The initial distribution η_k^0 was given by the equilibrium distribution

$$\eta_k^0(z_0) = \eta_0 \frac{g_k}{g_0} \exp [-B_0 k(k+1)/T(z)]_{z=z_0}.$$

The magnitude of the initial value z_0 was selected within the range 0.8-1.8. For each version with a different value of $p_0 d_*$, the correctness of the selection of z_0 was checked by the "insensitivity" of the solution to a diminution in z_0 .

Results of the computation are represented in Fig. 6a and b in comparison with the experimental data; curves 1-3 correspond to the computed data for $p_0 d_* = 144.5, 338,$ and 493 . Let us note that similar dependences have qualitatively been obtained earlier in both a computation [1] and experiment [8]. The systems of even and odd levels relax independently, the population temperatures of the odd levels vary exactly as monotonically as the even, and their values are between adjacent values of the population temperatures of even k . The computations and experiments show that the transition from the equilibrium state in the nozzle stagnation chamber to the nonequilibrium state at remote distances from the nozzle exit occurs with a disturbance of the Boltzmann population distribution for the rotational levels. It follows from the computations that a monotonic rise in the population temperatures with the increase in k is caused by a diminution in the rotational transition probabilities with the increase in k (see Fig. 5). A better correspondence between computation and experiment holds for $p_0 d_* = 493$. It is obtained for $\alpha = 2 \text{ \AA}^{-1}$ and a correction factor of $C = 10$. As $p_0 d_*$ varies, the computation

exhibits a faster tendency to build up equilibrium at the low levels ($k < 7$) than does experiment. At the upper levels the tendencies of the experimental and theoretical results are opposite as $p_0 d_*$ changes (the data of the experiments for $p_0 d_* = 493$ lie higher than for lower values of $p_0 d_*$).

3. A certain difference between the experimental and theoretical results is related to the uncertainty in the transition probabilities at the low rotational levels due to the limits of applicability of the quasiclassical computation [6, 15]. The difference at the upper levels can be associated with both experimental error and with the influence of diffusion not taken into account, which should contribute to an increase in η_k of the upper states and, therefore, to an increase in their population temperature T_k . The difference between the experimental and computed data becomes more noticeable at low temperatures $T \approx 20^\circ\text{K}$, when the attracting part of the potential [15] can induce a contribution to the interaction, and the influence of condensation on the kinetics of rotational transitions is possible.

Let us examine the influence of diffusion in more detail. For an estimate, let us write the behavior of η_k with the change in z approximately as follows:

$$\eta_k^{(0)} = \xi + \beta_k \exp(-\alpha_k z),$$

where ξ , β_k , α_k are constants, and $\beta_k < 0$ (> 0) for the lower (upper) levels of the system. Assuming the solution in a zero approximation to be determined just by collisional processes

$$\frac{d\eta_k^{(0)}}{dz} = \frac{d}{\rho v} \text{St}_k(\eta_k^{(0)}),$$

we obtain from this same equation in a first approximation

$$\frac{d\eta_k^{(1)}}{dz} \approx \frac{d\eta_k^{(0)}}{dz} \left\{ 1 - \frac{1}{\sqrt{y}} \frac{0,019}{\rho r^2 d} \left(\frac{\langle v_g \rangle}{v} \right) \left[\frac{3}{2} \frac{\gamma-1}{z+1} + \alpha_k \right] \right\}. \quad (3.1)$$

The expression $D_k = D = \langle v_g \rangle / 12\pi \sqrt{2} \rho r^2$ [18], where $\langle v_g \rangle$ is the relative velocity of the colliding molecules, $\langle v_g \rangle = \sqrt{4kT/\pi\mu}$, is hence taken as the diffusion coefficient, while the approximate dependence $T = T_0(z+1)^{\beta(1-\gamma)}$ is taken for the temperature. It follows from (3.1) that diffusion results in an increase in the population at the upper levels ($\beta_k > 0$) and a diminution in the population at the lower levels ($\beta_k < 0$), where as the translational temperature T diminishes the influence of the diffusion term grows in comparison to the collision integral, although we note that the values of the derivatives $d\eta_k/dz$ themselves become small in absolute value in the domain of $z \gtrsim 10$ calibers and the populations in this domain vary insignificantly. The absolute value of the correction term taking account of the influence of diffusion diminishes, as is seen from (3.1), with the growth of the parameter modeling the relaxation process $p_0 d_*$; i.e., the domain of values of $p_0 d_*$ within whose limits the influence of diffusion in computations of the kinetics of the rotational system can be neglected can begin to be indicated. Thus for the modes considered in this paper, according to estimate (3.1), the contribution of the diffusion terms is a quantity not exceeding 1%.

In conclusion, let us note that the rotational relaxation model constructed is equivalent, in principle, to a hydrodynamic description with the solution of the hydrodynamics equations replaced by a field of the parameters ρ , u , T . The solution of the kinetics equations in a one-dimensional approximation is acceptable in this case since it is shown experimentally and theoretically that the deviations of the flow from one-dimensionality are felt to the greatest degree in the domain of small distances from the nozzle exit. On the other hand, the distribution over the rotational levels turned out to be equilibrium for the values of $p_0 d_*$ under consideration in this domain, and the numerical computations started with $z_0 > 0.5$.

The authors note that the molecular-beam measurement data were obtained by R. G. Sharafutdinov and A. E. Zarvin and are used with the kind agreement of the latter.

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