The thermal fluxes introduced $q_{\alpha*}$ satisfy the boundary condition

$$q_{2*} - q_{1*} = \rho \Delta w_* l_{2,t}. \tag{31}$$

The system of Eqs. (26), (28) and (31) is closed with respect to $q_{\alpha*}$ and ℓ_2.

Stage 3 is completed by exit of the phase transtiion boundary to the layer boundary (at time $t = t_3$), after which the process is described by a single Eq. (1) for the entire phase 2 layer until the onset of any following phase transition. Incidentally, second and subsequent phase transitions are possible before the first is completed.

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ROTATIONAL RELAXATION TIME OF NITROGEN

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Rotational relaxation can be described theoretically with the help of the Wang-Chang—Uhlenbeck equations for the one-particle distribution function $f_j(\mathbf{v}, \mathbf{r}, t)$ [1], where j is the rotational quantum number, $\mathbf{v}$ is the velocity, $\mathbf{r}$ is the position, and t is the time. When the charcteristic rotational relaxation time is much larger than the time to establish an equilibrium distribution of the translational energies of the molecules, the distribution function fj can be written in the form of a product fj = $f(\mathbf{r}, \mathbf{v}, t)N_j(\mathbf{r}, t)$. In this case relaxation of $N_j(\mathbf{r}, t)$ occurs for the equilibrium translational energy distribution and is described by the system of kinetic equations of [2]. Reliable information on the rate constants of the rotational transitions is not available at the present time, and this makes analysis of the rotational kinetics on the basis of these equations difficult.

A less detailed description can be obtained by using the relaxation equation

$$dE_R/dt = -(E_R - E_t)/\tau_R, \tag{1}$$

which is only valid when the deviation from equilibrium is small (E_R and E_t are the actual and equilibrium values of the rotational energy and τ_R is the rotational relaxation time).

The rotational relaxation time has been determined from various types of experiments (ultrasound, shock waves, thermal transpiration, and so on), including measurements of the parameters in a free jet [3]. It is usually assumed in analyzing the experimental results that (1) is valid over the entire flow field, although in free jets significant deviations from equilibrium can be reached, which makes the validity of this equation doubtful over the entire region of the parameters. A second deficiency of the analysis of existing experimental data is that the characteristic collision number $Z_R = \tau_R/\tau_t$ is assumed to be a constant over the entire flow field and is determined from certain quantities measured in the jet (τ_t is the translational relaxation time). However, it is well known that Z_R depends on the temperature of the gas, which varies significantly in the jet.

In the present paper the rotational relaxation time in molecular nitrogen is found in supersonic free jets using electron beam diagnostics. The population densities of the rota-

tional levels of the N_2 molecules and the rotational energy E_R are calculated from the measured intensities of the rotational lines of the first negative band system of nitrogen, which are excited by the electron beam. For small deviations from equilibrium the quantity τ_R is determined from the measured value of E_R with the help of (1), as in [4] for the relaxation of nitrogen in argon. Our method allows one to determine the temperature dependence of the relaxation time and to extend the determination of τ_R down to 10 K, which was inaccessible in earlier studies. A more detailed analysis of the theoretical calculations of the rotational relaxation rate becomes possible when the temperature interval increases.

The density and temperature of the gas decrease rapidly for supersonic expansion of a gas from a nozzle into a vacuum. The decrease in the collision frequency leads to a gradual loss of equilibrium between the internal and translational degrees of freedom of the molecules. The vibrational degrees of freedom are not excited at fairly low temperatures and energy exchange in collisions occurs only between the rotational and translational degrees of freedom. One can obtain information on the rate of the relaxation process by following experimentally · the gradual deviation from equilibrium of the rotational degrees of freedom.

However, the gas escapes into a vacuum chamber with a low, but nonzero pressure. The flow parameters in the supersonic core of a free jet, bounded by the peripheral shock wave and by the Mach disk, are the same as for escape into a vacuum. The core dimensions depend on the parameters of the source and on the pressure in the vacuum chamber. At low pressures the braking of the jet becomes diffusive and the gas molecules surrounding the core and the compression layers behind the shock waves penetrate into the core, breaking up the flow. The use of jets of this kind to study relaxation processes is complicated by the fact that besides the gasdynamical expansion one must take into account diffusion, scattering, and so on. We were able to decrease the penetration of gas from the surrounding space into the jet core by high evacuation of the vacuum chamber with the help of oil diffusion and helium cryogenic pumps. In those cases when the effect of penetration was possible, it was controlled by changing the gas pressure in the vacuum chamber [5].

Another undesirable process, leading to a more complicated mechanism of energy exchange in the flow, is condensation. The rapid temperature drop in the expanding gas leads to the possibility of condensation. It is possible to limit the effect of condensation on rotational relaxation by using small gas densities, i.e., small braking pressures. However, this condition is incompatible with the condition for limiting the effect of the background gas. Therefore we used the largest possible nozzles consistent with minimizing the braking pressure. The question of obtaining data free from the effects of condensation of the background gas is considered in more detail in [5, 6].

The experiments were conducted using steady free nozzle jets in the Institute of Thermal Physics, Academy of Sciences of the USSR, Siberian Branch (bench No. VS-4 [5]). The electronic-vibrational-rotational spectra of the first negative band system of N_2^+ were recorded at different distances from the nozzle exit plane along the axis of the jet. These spectra were excited by an electron beam with electron energies 10-15 keV. The volume from which the radiation was sampled was bounded by the diameter of the electron beam (~2 mm) and the dimensions of the slits (0.1 × 3 mm). The distance from the nozzle exit plane was varied by moving the source of gas with respect to the stationary electron beam. We used the interval of braking temperatures T_0 from 295-1000 K. The braking temperature was measured by the discharge method and was controlled by thermocouples. The determination of T_0 is discussed in more detail in [7], and [8] contains a detailed discussion of the apparatus and errors in the raw measurements.

In order to compute the relaxation time from (1), it is necessary to find the energy of the rotational degrees of freedom in the ground state $N_2 X^1\Sigma_g^+$, v = 0 as a function of the coordinate x (x = 0 corresponds to the nozzle exit plane). In the experiments we measured the rotational spectra of the first negative system of bands (the process $N_2^+ B^2\Sigma_u^+$, v' = 0 → $N_2^+ X^2\Sigma_g^+$, v'' = 0 + hν) excited by direct electron collision [8] (the process $N_2 X^1\Sigma_g^+$, v = 0 + e⁻ → $N_2^+ B^2\Sigma_u^+$, v' = 0 + 2e⁻). The intensity of the rotational lines in the spectrum is related to the energy of the rotational degrees of freedom in the state $N_2 X^1\Sigma_g^+$, v = 0 by the equation [9]

$$E_R^X = \frac{\sum_{k'} I_{k'} (2k' + 1)(k' + 1)\Theta}{\sum_{k'} I_{k'} (2k' + 1)/k'} - \Delta E, \tag{2}$$

where $\Delta E = 12.7$ K is the rotational heating for electronic excitation of nitrogen molecules

TABLE 1

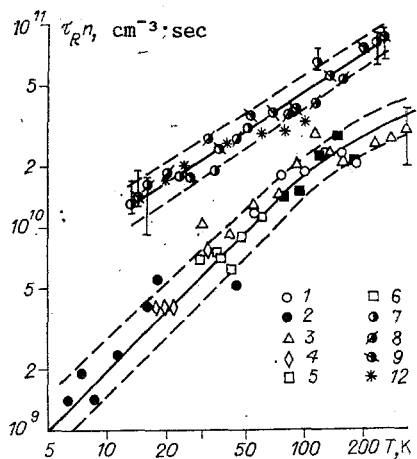| Expt. run	T_0, K	$n_0 d_* \cdot 10^{-16}$, cm^{-2}	x/d_*	Expt. run	T_0, K	$n_0 d_* \cdot 10^{-16}$, cm^{-2}	x/d_*
1	880	11	0,8—9,7	7	990	70,1	0,8—13,7
2	293	74,9	15—35	8	900	26,1	0,8—9,7
3	665	2,9	0,8—9,8	9	730	12,1	0,8—10
4	293	9,9	0,7—10	10	730	6,6	0,8—6,8
5	293	8,1	0,9—5,8	11	670	2,9	0,8—5,8
6	860	5,8	1,4—8,2				

Fig. 1

in the state $N_2^+ B^2 \Sigma_u^+$. Because transitions between ortho and para-nitrogen are forbidden the rotational energy was calculated separately for these two modifications.

For the central flow tube of a steady free jet (1) transforms to

$$\frac{u}{n d_*} \frac{dE_R}{d(x/d_*)} = -\frac{E_R - E_t}{n \tau_R}.$$

(3)

Here u and n are the velocity and density of the gas, which are calculated using the isoentropic relations for a diatomic gas with $\gamma = c_p/c_V = 1.4$; d_* is the critical nozzle diammeter. Equation (3) was used to calculate $n\tau_R$ from the measured rotational energies E_R along the coordinate x/d_*. A smoothing cubic spline [10] was used in determining the derivatives $dE_R/d(x/d_*)$. Table 1 gives data corresponding to the experimental conditions. The values of $n\tau_R$ calculated from the experimental data are shown in Fig. 1 against the translational temperature (the sets of points correspond to the number of the run listed in Table 1). The sets of points 1-6 correspond to the relaxation time in pure nitrogen. For comparison, Fig. 1 also shows data on the relaxation time of a weak impurity of nitrogen in argon, as obtained in [4] by a similar method (sets of points 7-9).

Because of the large deviations from equilibrium, runs 10 and 11 are not suitable for determining $n\tau_R$ by this method (see [4]). The results of these runs and runs 7-9 were used in [11] to compute the rate constants $K_{ij}(T)$ of rotational relaxation, from which $n\tau_R$ can be calculated [12]: $(n\tau_R)^{-1} = \sum_i \sum_{j>i} K_{ij}(T) N_i^*(T)(\varepsilon_i - \varepsilon_j)^2/(\langle \varepsilon^2 \rangle - \langle \varepsilon \rangle^2)$, where ε_i is the rotational energy of level i, $\langle \varepsilon^l \rangle = \sum_i N_i^* \varepsilon_i^l$. The resulting values of $n\tau_R$ are shown in Fig. 1 (set of points 12). The solid and dashed curves approximate the data within a 95% confidence interval. For relaxation of N_2 in Ar [4] $n\tau_R = 2.3 \cdot 10^9 \cdot T^{0.66}$ (cm$^{-3} \cdot$sec) and for relaxation in pure nitrogen in the region T < 200 K

$$n\tau_R = 2.0 \cdot 10^8 \cdot T^{1.0} \text{ (cm}^{-3} \cdot \text{sec)}.$$

(4)

It is important to note that for the system N_2 + Ar two essentially different methods of determining the relaxation times from data both close and far from equilibrium lead to the identical results (Fig. 1).

We begin our analysis of the results with a comparison of the energy of the rotational degrees of freedom measured in the present paper and in earlier publications. The molecular beam method was used in [3, 13-16]. The molecular beam was extracted from a supersonic jet

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with the help of a skimmer. Moments of the velocity distribution function of the molecules (the mean flow velocity and the translational temperature) were measured in this beam. Then the energy of the rotational degrees of freedom was found from conservation of energy. The molecular beam was extracted at a large enough distance from the nozzle exit plane so that rotational relaxation was frozen. The method requires high accuracy (in particular, the error in determining the velocity should not exceed 0.1%) and also requires the validity of the conservation of energy equation for the axial flow tube (this requirement may not be satisfied for highly rarefied jets).

The rotational energies measured in [3, 13-16] (sets of points 1-5) and in the present paper (set of points 6) at a distance $x/d_* = 38$ from the nozzle are shown in Fig. 2. In this case the rotational energy E_R is rather close to its limiting frozen value; this can be verified by comparing with other experiments in which E_R was determined for $x/d_* > 100$. The observed satisfactory agreement of the data indicates the absence of deviations in the raw experimental results. The solid curves show the calculated values of E_R using (3) with the relaxation time given by (4) (curve 7) and with relaxation times lying within the confidence interval (curves 8 and 9). The calculated and experimental curves approach one another only in the limit of large values of the parameter $n_0 d_*$. One can obtain agreement between experiment and calculation over the entire region of $n_0 d_*$ by varying Z_R, but of course only at one particular distance from the nozzle exit plane [3, 13-16]. However, this method of finding Z_R gives erroneous results for the following reasons. The value of Z_R depends on the temperature of the gas. The temperature of the gas on the axis of the jet varies over a wide range: from the braking temperature (300 K and higher) to fractions of a degree Kelvin at the point of extraction of the molecular beam. The value of Z_R found by fitting to experiment is an average and cannot be regarded as corresponding to any particular value of the temperature. In addition, the E_R data from [3, 13-16] were obtained for fairly low values of $n_0 d_*$ and large distances from the nozzle exit plane, i.e., under conditions when deviations from equilibrium are large and the applicability of (1) is doubtful. It is evident from the result of [4] that the quantity $n\tau_R$ depends on $n_0 d_*$ when determined from experiments with large deviations from equilibrium and hence it is not a true physical characteristic.

The values of Z_R found here are compared in Fig. 3 with previous experimental (a) and theoretical (b) data. In the calculation of τ_t the gas-kinetic cross section was computed using the formula $\sigma_t = \sigma_0(1 + C/T)$, where $\sigma_0 = 32.56 \cdot 10^{-16}$ cm^2 and $C = 105$ K were taken from [17].

The results of the ultrasonic measurements are shown in Fig. 3a by the sets of points 1-3 (corresponding to [18-20], respectively). A large amount of ultrasonic data obtained at room temperature and systematized in [19] are represented in Fig. 3a in the form of the interval 4 bounding limiting values of Z_R. The values of Z_R calculated from the thermal transpiration experiments are shown by the sets of points 5-11 (corresponding to [21-27], respectively). The data obtained from shock-wave experiments are shown by the sets of points 12-14 ([28-30]), and our results are shown by the set of points 15. The results for Z_R calculated from the transport coefficients are not compared here, since they are less accurate. However, they agree fairly well in order of magnitude with the data shown in Fig. 3a. There is satisfactory overall agreement between the results obtained by the different methods; however we note that the data from the acoustic and thermal transpiration measurements lie somewhat below the rest of the data. At low temperatures $Z_R \lesssim 1$. However this may not correspond with reality since there is no reliable information on the gas-kinetic cross section for $T \lesssim 80$ K.

Note that the data of the ultrasonic measurements (the sets of points 2 and 16 in Fig. 3a) closely agree with our results, whereas such agreement was not obtained for N_2 + Ar mixtures [4]. Since the number of recorded rotational lines and the method of analyzing the data were identical for pure N_2 and for the N_2 + Ar mixture, this suggests that the application of a weak N_2 impurity in Ar leads to a weak sensitivty of the ultrasonic absorption coefficient measured in [31].

The experimental data are compared with various theoretical dependences in Fig. 3b. The region of values of Z_R obtained in the present paper is cross hatched; the corresponding region is shaded for the other papers included in the study. The results obtained by calculations in the framework of classical mechanics are represented by curve 1 [32], 2 [33], 3 [28], 4 [34], 5 [35]. The results of [36] are shown by the set of curves 6 for different values of the anisotropic part of the interaction potential. Recently [37] the same authors carried through calculations based on more complicated expressions for the molecular interaction potential [38, 39]; these results correspond to curves 9 and 10 in Fig. 3b. The calculations
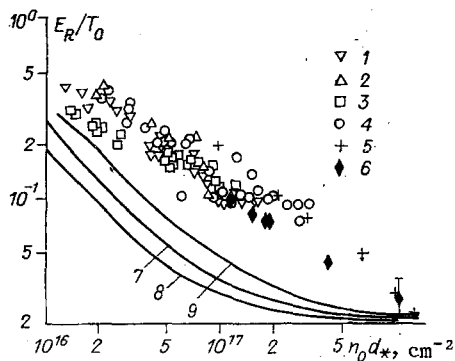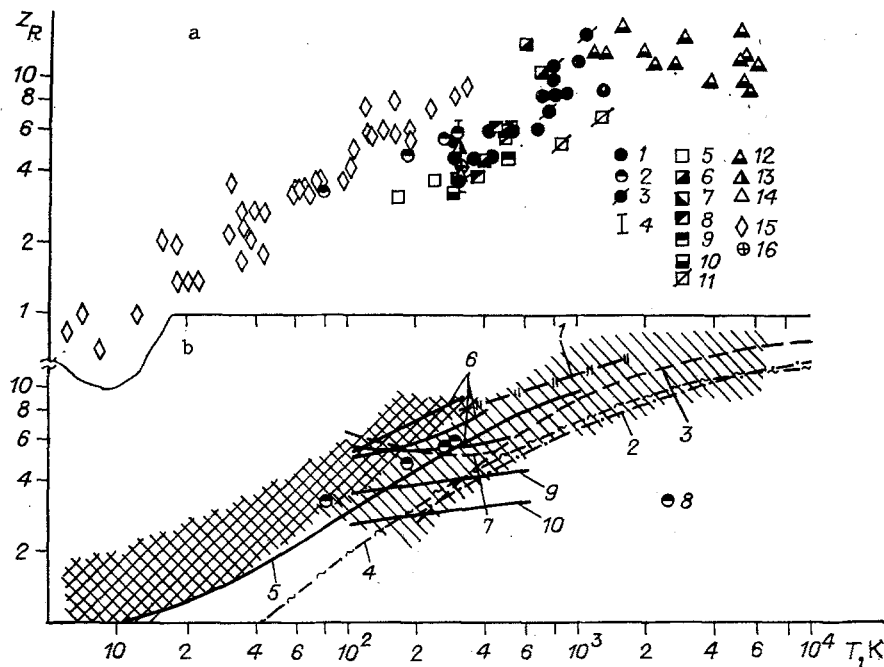
Fig. 2

Fig. 3

of [40] were carried out by a similar method for the experimental conditions of [19] and they closely agree with the experimental results; these calculated results are represented by the set of points 8. Curve 7 corresponds to the results of [41].

We note that the differences between the theoretical curves $Z_R(T)$ in the system $N_2 + N_2$ are roughly the same as the scatter of the experimental results. The three-dimensional classical theory [32, 35] gives results which are closest to the experimental data. All of the classical theories [29, 33-36], as well as experiment, predict a growth in Z_R with increasing temperature. The values of Z_R obtained from the trajectory calculations [36] depend strongly on the anisotropy parameters of the interaction potential, which suggests that these parameters could be determined by comparison with the experimental results. For example, a comparison was carried out in [36] with the data of [13, 18, 19] (see Fig. 3a) over a narrow temperature region and the values of the parameters $a = 0.1$ and $b = 0.7$ were obtained (notation of [36]); this corresponds to the lowest of the curves labeled 6 in Fig. 3b. However, the upper curve, for which $a = 0.13$ and $b = 0.5$, better corresponds to the data overall.

Finally we point out the main advantages of our method of determining the rotational relaxation time over the earlier methods. The spectroscopic method of determining the energy of the rotational degrees of freedom gives the capability of controlling the contribution of each rotational level to the calculated relaxation time of the entire system. This capability does not exist in the ultrasonic and thermal transpiration methods, as well as methods based on measurements of the transport coefficients.

The relaxation times and quantities directly measurable in different types of experiments are related through the kinetic theory of the given phenomenon (examples are propaga-

tion of ultrasound, rarefied gas flow in a capillary, shock-wave structure, and so on). It is obvious that the interpretation of the experimental data must include analysis of the theoretical relations corresponding to the experimental conditions. In particular, one of the fundamental assumptions of all of these theories is the smallness of the deviation from equilibrium. In the analysis of the results obtained in free jets there exists the possibility of choosing the conditions such that the deviation from equilibrium can be considered small and where the relaxation equation is applicable [4]. One expects that in thermo-molecular flows large deviations from equilibrium will not occur; however the kinetic theory of this phenomenon is complicated and is still being developed. As follows from [42], analysis of the thermal transpiration data using the new theories gives higher values of Z_R than those shown in Fig. 3a.

For propagation of ultrasound one cannot guarantee that the deviation from equilibrium will always be small and we concur with the authors of [32] that in ultrasonic measurements "the deviation from equilibrium, particularly at low temperatures (near 300 K), can be significant." In strong shock waves it is meaningless to speak of a small deviation from equilibrium between the rotational and translational degrees of freedom.

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