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ROTATIONAL RELAXATION IN THE TRANSITION REGIME OF FREE NITROGEN JETS

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The development of laser technology, the study of nonequilibrium chemical reactions, and the spectroscopy of a supercooled gas require a quantitative description of the kinetics of the population densities of rotational levels. Currently, inelastic collisions with molecular rotational-energy transfer are being studied intensely. Progress in the theory is reported in [1]. Considerable successes have also been achieved in experiments on determining the cross sections of rotationally inelastic collisions [2]. However, information obtainable from work along these lines is not sufficient to describe such processes as rotational relaxation in free jets, shock waves, and nonequilibrium chemical reactions, i.e., for those cases when information on rate constants, the integrated cross sections of inelastic collisions, is necessary to describe the phenomenon. In order to determine the rate constants, different approximations [1], whose validity is not always obvious, are used. For this reason, it makes sense to carry out experimental investigations over a wide range of variation of the parameters being determined and to construct empirical generalizations based on them.

It has been established experimentally (cf. [3, 4]) that the deviation from equilibrium between rotational and translational degrees of freedom occurs with a breakdown in the Boltzmann distribution of the population densities. In order to describe the nonequilibrium distribution that arises, the concept of a rotational temperature is not adequate; it is necessary to introduce the concept of population-density temperatures for separate rotational levels, determined from the equation $N_k = N_0(2k + 1)\exp[-k(k + 1)\Theta/T_k]$ substituting the measured values of the population densities of the k-th N_k and zeroth N_0 rotational levels (Θ is the characteristic temperature). It has been found [3] that the temperatures of the populations of the lower rotational levels are closer to the translational temperature than the upper levels, which indicates the stronger coupling of these levels to the translational degrees of freedom. Here, it is not clear whether or not it is possible to examine translational and rotational relaxation separately, as done at the present time in most theoretical papers [5, 6]. There are as yet no experimental studies in which combined measurements of the population density distributions of the rotational levels and the translational velocity distribution function are carried out.

The purpose of the present work is to study experimentally the translational—rotational relaxation in a free nitrogen jet in the transition regime from continuous to free-molecular motion. The free nitrogen jet was chosen as the object of the investigation for the following basic reasons. In such a jet, it is possible to obtain and maintain for a long time the necessary degree of departure from equilibrium with respect to the rotational and internal degrees of freedom of the molecules. The flow along the axis of the jet is quite well described as a spherical expansion into a vacuum, which greatly simplifies the geometry of the problem [7]. The low supersonic flux density permits using a wide range of diagnostic tools, which give detailed information on molecular rotational and translational degrees of freedom.

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1. The investigation was carried out on the gasdynamic low-density setup at the Institute of Thermophysics of the Siberian Branch of the Academy of Sciences of the USSR (stand VS-4), equipped with electron-beam and molecular-beam diagnostics for measuring the gas density, population density of the rotational levels, and the translational velocity distribution function. The technique of electron-beam diagnostics is described in detail in [8] and that of molecular-beam diagnostics in [9].

Axisymmetrical sonic nozzles served as a source of gas. The diameter of the nozzles d_* varied from 0.54 to 50 mm. In order to avoid a large effect of viscosity in the subsonic part of the nozzle, the internal angle of the nozzle was chosen to be quite large, while the thickness of the rim in the critical section was made as small as possible. Commercial-grade nitrogen was used as the working gas. The stagnation temperature T_0 was room temperature in all experiments. It was controlled by Chromel—Alumel thermocouples, placed inside the forechamber of the nozzle. The gasdynamic setup was evacuated by a cryogenic pump, operating on gaseous helium at a temperature 7-13°K. The pressure level of the background gas in all measurements did not exceed 0.3 Pa. As a result of the low background level, its effect on the measured parameters was negligibly small [3].

A large part of the experiments in this work was carried out under conditions such that in the vicinity of the measurements, the gas was in a saturated state. In the supercooled gas, the population density kinetics are determined by simultaneous occurrence of rotational relaxation and condensation. For this reason, in order to analyze the rotational relaxation, it is necessary to separate out the effect of condensation on the level population densities. Strictly speaking, this can be done only approximately, since at the present time, even the criteria that determine this effect are not known.

The rate of rotational relaxation is determined by the number of binary collisions and at a constant stagnation temperature depends on the product of the stagnation pressure and the diameter of the nozzle section $p_0 d_*$ [6, 10]. For condensation, the dependence on $p_0 d_*$ is weaker and has the form $p_0 d_*^{\alpha}$ where $\alpha < 1$ [11]. For this reason, as also in [6, 8], in the present work, the effect of condensation was separated out by comparing the experimental data, obtained for constant $p_0 d_*$ but different p_0, with a change in the diameter of the nozzle. The values of $\log(I_{k'}/I_1 k')$ as a function of distance x/d_*, where $I_{k'}$ is the intensity of the lines in the spectrum of the first negative band system, were compared; k' is the rotational quantum number in the $B^2\Sigma$ $(v = 0)$ state of the nitrogen ion; x is the distance from the nozzle cutoff. Coincidence of the values $\log(I_{k'}/I_1 k')$ obtained for different nozzles was taken as an indication of the fact that the effect of condensation on the intensity of the rotational lines is insignificant.

2. An example of the results of the measurements of the rotational-level population densities and the translational velocity distribution function of the molecules is shown in Fig. 1 for $p_0 d_* = 6650$ Pa·mm and $T_0 = 300$°K in the form of a graph of T_i/T_0 as a function of x/d_*, where i is the variable index. The continuous line indicates the calculation of an isentropic flow for heat capacity ratio $\gamma = 1.4$ [12]. The velocity distribution function of the molecules is ellipsoidal and is characterized by two translational temperatures (parallel $T_{\parallel}$ and perpendicular $T_{\perp}$), corresponding to distributions of molecular velocities along and across the axis of the flow. The following notation is used in Fig. 1: points 1-3 denote the population temperatures of the 11th, 7th, and third levels, respectively; 4 and 5 are $T_{\parallel}$ and $T_{\perp}$. As can be seen from the graph, the results of the measurements of the translational temperatures confirm the basic results of the investigation of the distribution function in flows of a monotomic gas: $T_{\perp}$ follows for a quite long time the equilibrium isentropic temperature and then decreases with increasing x/d_* more quickly than the latter; $T_{\parallel}$ deviates much sooner from the equilibrium value and with further expansion of the gas remains constant. It should be noted that the temperatures of the populations of separate levels T_k differ from the equilibrium values at smaller distances from the nozzle, and in addition, this deviation is greater the higher the rotational quantum number.

The results of the present work are compared with data on measurements of the distribution function in a helium jet [13] (we have similar data from the literature on nitrogen) in Fig. 2, the notation in which is presented in Table 1. The results are presented in the form of dependences of measured temperatures, normalized to the corresponding computed isentropic values T_{isen}, as a function of the parameter P_B. The parameter P_B, introduced in [14] in order to generalize the results on the deviation of the translational temperature from equilibrium isentropic values with a change in the conditions under which the relaxa-

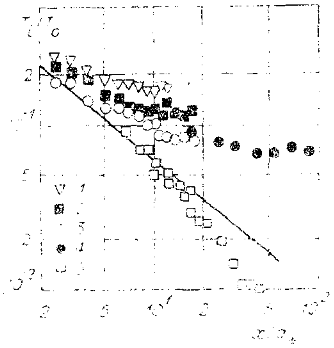

Fig. 1

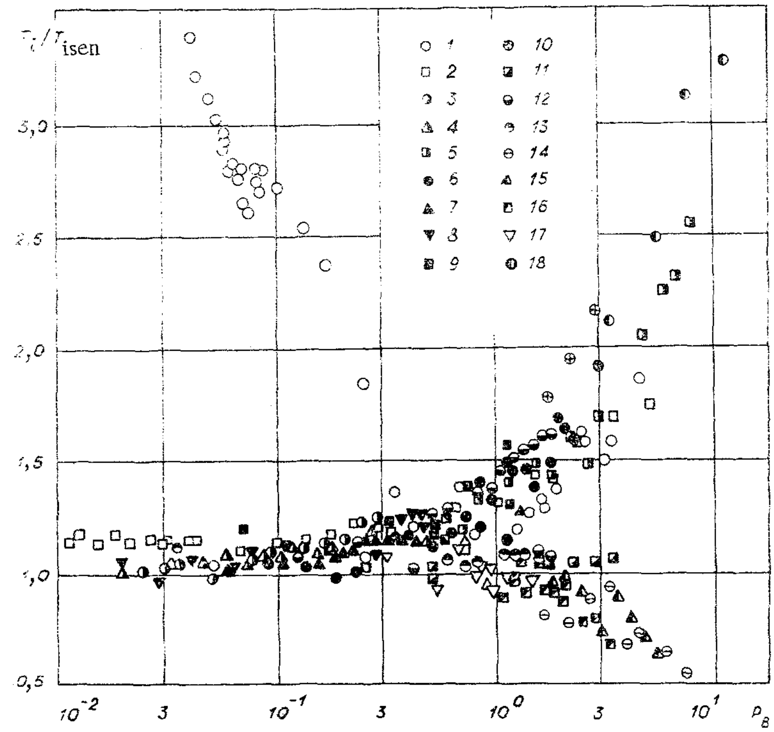

Fig. 2

tion process occurred, is actually a local Knudsen number, calculated as the ratio of the mean-free path length to the density scale length [14], and can be written in the form

$$P_B = (u/\nu\rho)|d\rho/dx|, \tag{2.1}$$

where u is the mean mass velocity of the flow; $\nu = v_s/\lambda$ is the average collision frequency; $v_s = \sqrt{8kT/\pi m}$ is the average thermal speed; λ is the mean free path; ρ is the gas density. Substituting into (2.1) approximate expressions for Mach's number and the gas density from [7], the equation used for P_B in the calculations has the form

$$P_B = c_1 (x/d_*)^\nu (p_0 d_*)^{-1},$$

where the coefficient c_1 depends on the magnitude of the ratio of heat capacities.

As follows from Fig. 2, the transverse temperature in helium [13] in the entire range of values P_B is close to the isentropic value, while in nitrogen, according to the data obtained in the present work, $T_\perp/T_{isen}$ deviates from unity for $P_B > 1$. The reason for this difference could be the effect of the background gas in the experiments [13], penetrating into the jet far from the nozzle, i.e., for large values of the parameter P_B. This conclusion follows from the analysis of the results in [13], in which the experimental values of both $T_\perp$ and $T_\parallel$ deviate from the general dependences toward higher temperatures at smaller distances from the nozzle, the more rarefied the jet. The experimental data obtained in the

TABLE 1

No. (in order)	Gas	T_i	$p_0 d_*$ Pa·mm	x/d_*
1	N_2	T_{tr}	$6\cdot10^3-2\cdot10^6$	38
2	N_2	T_{tr}	$6.7\cdot10^2-4.4\cdot10^5$	5
3	N_2	T_{tr}	$3\cdot10^4$	0.4–10
4	N_2	T_{tr}	$3.1\cdot10^4$	1.4–10
5	N_2	T_{tr}	$3.2\cdot10^4$	8–100
6	N_2	T_{tr}	$2\cdot10^3$	1–12
7	N_2	T_{tr}	$1\cdot10^4$	1–12
8	N_2	T_{tr}	$2\cdot10^4$	1–12
9	N_2	T_{tr}	$2.3\cdot10^4-3.7\cdot10^5$	20
10	He	$T_{\parallel}$	$6.81\cdot10^3$	2–9
11	He	$T_{\perp}$	$6.81\cdot10^3$	2–9
12	He	$T_{\parallel}$	$4.73\cdot10^4$	2–19
13	He	$T_{\perp}$	$4.73\cdot10^4$	2–19
14	N_2	$T_{\perp}$	$1.36\cdot10^4$	18–76
15	N_2	$T_{\perp}$	$4.1\cdot10^4$	25–92
16	N_2	$T_{\perp}$	$6.82\cdot10^4$	37–92
17	N_2	$T_{\perp}$	$1.5\cdot10^5$	45–92
18	N_2	$T_{\parallel}$	$6.1\cdot10^3$	17–41

present work are practically unaffected by the background gas, which is what permitted recording the decrease in $T_{\perp}$ compared to T_{isen}.

The result is also confirmed by measurements of $T_{\parallel}$: the data for helium [13] are somewhat higher than the results for nitrogen with high values of P_B. Unfortunately, we do not have experimental data on the parallel temperature in nitrogen for values of the parameter $P_B < 3$, while in helium the measurements are limited to the region of values $P_B < 4$. For this reason, in Fig. 2, in addition to the results of measurements of the transverse and parallel temperatures, the results of estimates of the translational temperature from the rotational spectra T_{tr} are also presented.

As follows from [3], as well as from Fig. 1 of the present work, the values of the temperatures of the level population densities T_k decrease monotonically with decreasing k, approaching the translational temperature. For this reason, the value T_k for $k\to0$ can give an estimate of the translational temperature for a Maxwellian distribution and $T_{\parallel}$ in the absence of equilibrium between translational degrees of freedom.

The results presented in Fig. 2 for the temperature T_{tr} were obtained over a wide range of values of the stagnation pressure, distances from the cutoff of the sonic nozzles, and nozzle diameters (see Table 1) and fall into the range of values of the parameter P_B from 10^{-2} to 10^1. The values of T_{tr} for small P_B coincide with the data on $T_{\parallel}$ from [13] and fall on the general dependence, deviating from unity for $P_B \sim 0.1$. Some of the separation of the results of measurements with increasing P_B, aside from the reasons indicated above, is a result of the approach of the experimental data for $P_B\to\infty$ to different limits ("freezing") of the translational temperature at different distances from the nozzle (for $p_0\to0$) and for different stagnation pressures ($x/d_*\to\infty$). This indicates the impossibility of generalizing the experimental data over the entire range of measurements of the parameter P_B in the coordinates of Fig. 2.

On the whole, the agreement between the results for nitrogen and helium can be assumed to be good, if we take into account the known uncertainty in determining the mean free paths λ for low gas temperatures (10-18°K) at which the measurements were carried out. In the present work, the mean free path was determined from the eqaution $\lambda = 1/(\sqrt{2}\pi\sigma^2 n)$, where n is the number density of the gas; σ is the gas kinetic diameter of molecules, taken as equal to $3.5\cdot10^{-8}$ cm for nitrogen and $2.8\cdot10^{-8}$ cm for helium independent of the gas temperature [15].

The experimental data obtained correspond to the region in which relaxation processes are observed and are not distorted by the effect of condensation. In order to check this

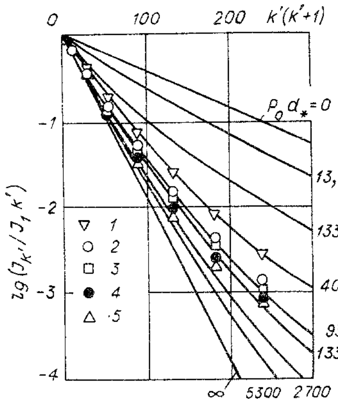

Fig. 3

result, Fig. 2 also shows the results of measurements of T_{tr}, obtained over a wide range of values of $p_0 d_*$, up to $p_0 d_* \gg 10^5$ Pa·mm (regimes 1, 2, 9), i.e., known to be affected by condensation for small values of P_B [3]. Indeed, as $p_0 d_*$ increases, i.e., with decreasing P_B for fixed distance from the nozzle, these results deviate from the overall dependence all the more strongly and for higher values of P_B, the greater x/d_*. As P_B increases, the effect of condensation decreases, so that the experimental data fall on a general curve.

Thus, we can state that for expansion from a sonic nozzle, the quantity $P_B \sim 0.1$ determines the boundary at which the Maxwellian distribution over translational velocities in flows of a diatomic gas breaks down.

3. In the region of purely rotational relaxation, the evolution of the rotational line intensity distributions, reflecting the evolution of the population density distributions, is shown in Fig. 3 over a wide range of values $p_0 d_*$ in the coordinates $\log(I_{k'}/I_1 k') \sim k'(k'+1)$ for $x/d_* = 2.45$ and $T_0 = 290°K$. In Fig. 3, the numbers 1-5 indicate the experimental data for $5.32 \cdot 10^4$, $1.24 \cdot 10^5$, $1.76 \cdot 10^5$, $3.6 \cdot 10^5$, and $7.05 \cdot 10^5$ Pa·mm, respectively. At constant stagnation temperature, the rate of rotational relaxation is determined by the magnitude of the product $p_0 d_*$. For $p_0 d_* \to 0$, the number of collisions in the flow drops and the population density distribution approaches that in the forechamber, i.e., to a Boltzmann distribution with the stagnation temperature, while for $p_0 d_* \to \infty$, the population density distribution approaches the equilibrium distribution, corresponding to isentropic expansion. The actual experimental values of the population densities, presented in Fig. 3, vary within these limits with variation of $p_0 d_*$.

In Fig. 3, the continuous lines show the results of a calculation using a model of the rotational relaxation (here and in what follows, the theoretical calculations of the rotational-level population densities are taken from [16]). The computed and experimental data do not completely agree, which is related to the inadequate choice of rate constants of rotational relaxation for the upper rotational levels in [16], as well as the effect of the initial stage of condensation. But, on the whole, the calculations and the experiments show that the deviation from isentropic flow with decreasing $p_0 d_*$ occurs through the population density distributions, not differing strongly from Boltzmann distributions, and in this case the population densities of all levels increase at the same time. The deviation from the Boltzmann distribution decreases for small $p_0 d_*$ and for such distributions it is possible to introduce a single rotational temperature, different from the translational temperature. This kind of variation in the population densities as a function of $p_0 d_*$ is also observed for other values of x/d_*, which makes it possible to generalize the results of the measurements.

We will represent the results obtained as a dependence of the ratio of the differences between logarithms of reduced intensities

$$\eta_{k'} = \frac{(\lg(I_{k'}/I_1 k'))_{T_{isen}} - (\lg(I_{k'}/I_1 k'))_{T_{k'}}}{(\lg(I_{k'}/I_1 k'))_{T_{isen}} - (\lg(I_{k'}/I_1 k'))_{T_0}} \qquad (3.1)$$

on the product $p_0 d_*$ (Fig. 4). The numerator in expression (3.1) represents the difference between the logarithms of reduced rotational level intensities in the flow at a fixed distance from the nozzle with an equilibrium isentropic outflow (with temperature T_{isen}) and under real conditions (with the population temperature $T_{k'}$), while the denominator corre-

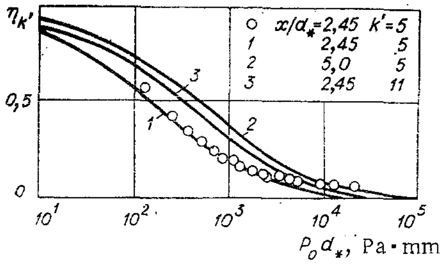

Fig. 4

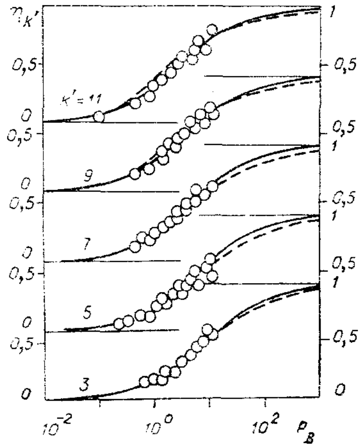

Fig. 5

sponds to the limiting difference between logarithms for isentropic and effusive (for $T_{k'} = T_0$, T_0 is the stagnation temperature) outflow. The points in Fig. 4 show the experimental data for $x/d_* = 2.45$ and $k' = 5$, and curve 1 shows the results of the theoretical calculation. Naturally, as $p_0 d_*$ increases (for $p_0 d_* \geqslant 4 \cdot 10^3$ Pa·mm), the experimental data, as in Fig. 3, deviate somewhat from the computed data. Nevertheless, as expected, both the experiment and calculation fall on a smooth curve, varying from 1.0 for $p_0 d_* \to 0$ to 0 for $p_0 d_* \to \infty$.

For a different distance from the nozzle ($x/d_* = 5$), the data fall on a similar smooth curve (curve 2), but do not coincide with the first curve. A similar result is also obtained for other distances. The observed deviation is caused by a slowing down of the relaxation process with increasing x/d_* and as a function of distance. From the analysis of data presented in Fig. 4 and other similar data, it is natural to suggest that in order to generalize the experimental data, obtained under conditions of pure rotational relaxation, it is possible to use the parameter P_B. Such generalizations for several values of k' are presented in Fig. 5, where the points are the experimental data, bounded by the region of values of P_B corresponding to conditions of purely rotational relaxation, and the dashed curves show the theoretical calculations, carried out for a larger range of values of P_B. Both the experimental and the computed data, which agree with the experimental data, lie on common curves for fixed k', and in addition, $\eta_{k'}$ varies from zero for $P_B < 10^{-2}$ to 1 for $P_B > 10^3$.

In Figs. 4 (curve 3) and 5 it is evident that the generalizing curves $\eta_{k'}$ as a function of $p_0 d_*$ and P_B for different numbers k' do not coincide, although they have the same form, which is explained by the different relaxation rate for gas molecules in different rotational levels [3]. In order to obtain a single curve, generalizing the values of $\eta_{k'}$ for different $p_0 d_*$, x/d_*, and k' the parameter P_B was multiplied by some function of k'. The form of the curves of $\eta_{k'}$ as a function of P_B for any k' can be described by the expression

$$\eta_{k'} = 1 - [1 + c_1(k') P_B^{0.75}]^{-1}, \tag{3.2}$$

which gives the correct limiting values of $\eta_{k'}$ ($\eta_{k'} = 0$ for $P_B \to 0$ and $\eta_{k'} = 1$ for $P_B \to \infty$) and in the region of values $10^{-2} < P_B < 10^3$ approximates well the experimental and computed data. The parameter c_1 depends on the level number k' and is described by the equation

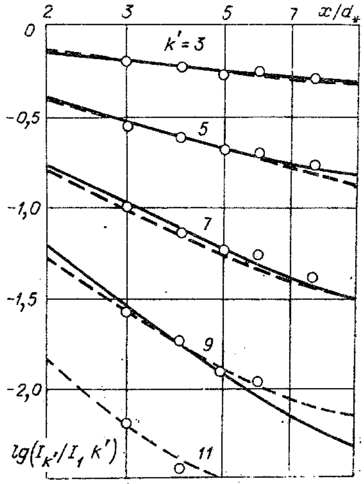

Fig. 6

$$c_1(k') = 0.1 + 0.03k'. \tag{3.3}$$

The approximating curves calculated using Eqs. (3.2) and (3.3) for corresponding level numbers k' are shown in Fig. 5 by continuous lines. As is evident from the graph, this empirical dependence can be completely generalized by the available experimental data in the range of the parameters $x/d_* = 2\text{–}10$, $p_0 d_* = 6.6 \cdot 10^1 \text{–} 4 \cdot 10^4$ Pa·mm, $k' = 3\text{–}11$, i.e., under conditions when rotational relaxation is not distorted by any additional effects. A similar generalization is also obtained for the population densities of rotational levels in the $X^1\Sigma$ (v = 0) state of the nitrogen molecule. In this case, instead of the coordinate $\eta_{k'}$, we used the coordinate

$$\eta_k = \frac{\lg(N_k/N_0(2k+1))_{T_{\text{isen}}} - \lg(N_k/N_0(2k+1))_{T_k}}{\lg(N_k/N_0(2k+1))_{T_{\text{isen}}} - \lg(N_k/N_0(2k+1))_{T_0}} = \frac{1/T_{\text{isen}} - 1/T_k}{1/T_{\text{isen}} - 1/T_0},$$

where k is the rotational quantum number in the $X^1\Sigma$ (v = 0) state.

The empirical generalization obtained makes it possible to estimate the distribution of the population densities of levels in the range of flow regimes from continuous to nearly free molecular flow with insignificant deviation of the density and velocity from the corresponding isentropic values.

An example of such an estimate for the dependences of logarithms of the reduced rotational line intensities on the distances x/d^* in one of the experimental regimes ($p_0 d^* = 1.8 \cdot 10^3$ Pa·mm, $d^* = 15$ mm, $T_0 = 295°$K) is shown in Fig. 6 for odd level numbers k' by continuous lines. Here, for comparison, the experimental (points) and theoretical results, using the rotational relaxation model (dashed lines), are also presented.

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EXPERIMENTAL INVESTIGATION OF THE CHARACTERISTICS OF SUPERSONIC EXPANSION OF ELECTRIC-ARC PLASMA JETS

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UDC 533.9.082.76

Under certain conditions, large deviations from thermodynamic equilibrium are possible with supersonic stationary plasma flow out of a high-pressure chamber into a low-pressure region. Theoretical studies of free supersonic plasma expansion [1-3] and experimental studies of supersonic underexpanded plasma jets [4-6] show that ionization and thermal equilibrium break down in the flow field at some stage of expansion. In this case, the degree of ionization exceeds the equilibrium value, while the electron temperature turns out to be higher than the temperature of heavy particles (atoms and ions). The deviation from equilibrium increases downstream.

The parameters of the plasma in a supersonic jet are closely related to the type of plasma source and flow conditions. In this paper, we attempt to establish the basic parameters that characterize the deviation from ionization and thermal equilibrium and to obtain generalizing dependences for the electron temperature T_e and concentration n_e in the region of free expansion for an argon electric-arc plasma flowing out of a sonic nozzle into a flooded low-pressure region.

1. Conditions and Range of the Investigations. The construction of the plasma source and the scheme of the initial region of the jet are shown in Fig. 1a. The electric-arc source consists of a cathode unit 1 with a tungsten cathode, housing 2, and an anode nozzle 3. We used cylindrical nozzles with diameters d = 2 and 5 mm and length d. The working gas was introduced through tangential openings in the housing. The source was placed inside a vacuum chamber with a volume of 10 m^3 on a coordinated setup with two degrees of freedom in the horizontal plane. The measuring instruments were stationary relative to the vacuum chamber.

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