the thermal expansion coefficient) at relatively low values of laser beam energy density.

In conclusion, the authors consider it their pleasant duty to thank G. A. Askar'yan for evaluating the study and S. V. Luk'yanov for assistance in measuring the particle size distribution.

LITERATURE CITED

1. G. A. Asker'yan, A. M. Prokhorov, et al., "Laser beam in a liquid," Zh. Eksp. Teor. Fiz., 44, No. 6 (1963).
2. G. A. Askar'yan, "Decrease in penetrating power of intense light due to scattering on refracting aureoles of an optically perturbed medium near inhomogeneities," Zh. Eksp. Teor. Fiz., 45, No. 3 (9) (1963).
3. V. M. Izgorodin, S. B. Kormer, et al., "Change in index of refraction of a liquid containing absorbing particles and irradiated by a high power light flux," Kvantovaya Elektron., 6, No. 6 (1979).
4. V. M. Izgorodin, A. V. Pinegin, and B. A. Poklontsev, "Effect of laser radiation on index of refraction of transparent liquids containing absorbing particles," Kvantovaya Elektron., 7, No. 4 (1980).
5. G. A. Askar'yan and B. A. Dolgoshein, "Acoustical propagation of a neutrino at great depths," Preprint FIAN Akad. Nauk SSSR, No. 160 (1976).
6. K. A. Naugol'nykh and N. A. Roi, Electrical Discharges in Water [in Russian], Nauka, Moscow (1971).
7. P. I. Golubnichii, G. S. Kalyuzhnyi, and V. I. Yakovlev, "Study of acoustical radiation generation mechanism initiated by a laser beam in a liquid," Preprint FIAN Akad. Nauk SSSR, No. 167 (1978).
8. S. D. Hunter, W. V. Jones, and D. J. Malbrough, "Nonthermal acoustic signals from absorption of a cylindrical laser beam in water," J. Acoust. Soc. Amer., 69, No. 6 (1981).

DISTURBANCE OF THE BOLTZMANN POPULATION DISTRIBUTION OF ROTATIONAL LEVELS IN A FREE NITROGEN JET

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Inelastic collisions with exchange of the energy of rotational motion of molecules are being intensively investigated theoretically and experimentally (see [1, 2], for example). Supersonic expansion of gas in nozzles or in free jets is one of the most convenient subjects for the investigation of rotational relaxation, which is due to the large amount of experimental work performed in this field in recent years. In particular, in free jets one can attain controlled values of the translational temperature in the range from fractions of a degree Kelvin (at a high gas density such low temperatures cannot be obtained by other means) to several thousand degrees.

In experiments on rotational relaxation in jets various diagnostic methods are used, yielding information not only on the macroscopic parameters but also on the population distribution of rotational levels. Despite the evident progress in research, however, a number of fundamental problems still remain unsolved.

In the interpretation of results, certain authors [3, 4] state that the transition from the equilibrium state in rotational degrees of freedom in a gasdynamic source to a nonequilibrium state at a certain distance from it takes place through a succession of Boltzmann population distributions of rotational levels, whereas others [5, 6] find a disturbance of the Boltzmann distribution. The question of the form of the population distribution is important in a theoretical description of rotational relaxation. In the case of a Boltzmann distribution one can introduce a rotational temperature for which the relaxation equation

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$$dT_r/dt = -(T_r - T_t)/\tau_r$$

is valid, where T_r and T_t are the rotational and translational temperatures; τ_r is the characteristic relaxation time; t is time. In the absence of a Boltzmann distribution one must use a more complicated system of equations of level-by-level kinetics to describe rotational relaxation. Up to now the most detailed information about rotational relaxation has been obtained using electron-beam diagnostics.

The model of processes of excitation of the $N_2X^1\Sigma_g^+$ molecule to the $N_2^+B^2\Sigma_u^+$ state with subsequent spontaneous emission $N_2^+B^2\Sigma_u^+ \rightarrow N_2^+X^2\Sigma_g^+$, developed by Muntz [7] for nitrogen, by connecting the intensities of rotational lines of the 00 band of the first negative system (1 NS) with the populations of energy levels in the ground state of the molecule, made it possible to measure the distribution of rotational energies of nitrogen molecules and, in equilibrium, the rotational temperature.

In the first careful research [8], however, it was found that under certain conditions the population distribution of rotational levels in the $N_2^+B^2\Sigma_u^+$ state (v = 0) differs considerably from a Boltzmann distribution. This fact raised doubts as to the validity of Muntz's model, particularly the two assumptions made by the author in developing it: 1) The excitation $N_2X^1\Sigma_g^+ \rightarrow N_2^+B^2\Sigma_u^+$ occurs only with primary electrons of the beam; 2) the excitation by fast primary electrons occurs with dipole selection rules for rotational quantum numbers, i.e., $\Delta k = k' - k = \pm 1$, where k' and k correspond to the $N_2^+B^2\Sigma_u^+$ and $N_2X^1\Sigma_g^+$ states. Various modified excitation models have appeared [9, 10], the authors of which reject one or another of Muntz's assumptions.

At the same time, hypotheses have also been advanced that the detected difference from equilibrium is not connected either with the diagnostic method or with rotational relaxation but is caused by other processes occurring in the gas jet: by the influence of the "warm" background gas penetrating into the "cool" jet [11] and, under the conditions of strong supercooling of the gas, by the formation of clusters and liberation of the heat of condensation in the stream [12].

The aim of the present work is an experimental demonstration of the fact that under certain conditions it is just the process of rotational relaxation that results in a significantly non-Boltzmann population distribution of rotational levels in the molecular ground state.

EXPERIMENTAL

The experiments were carried out on the vacuum bench of the Institute of Heat Physics, Siberian Branch, Academy of Sciences of the USSR, equipped with a vacuum-pump system including cryogenic pumps, electron-beam diagnostics, and a spectral apparatus permitting the recording of optical and x-ray emission. The experimental scheme is described in detail in [13].

Gas from the stagnation chamber, located on a three-component positioner, expanded through a sonic nozzle with a diameter d_* of from 0.5 to 15 mm into a vacuum space with a residual pressure $p_c = 10^{-2}$–10^{-1} Pa. The stagnation pressure p_0 varied from 10^2 to 10^6 Pa and the stagnation temperature T_0 from 300 to 1000°K. The gas in the forechamber was heated with a Nichrome coil mounted in the annular gap between two quartz tubes through which the working gas was blown. The quantity T_0 was determined by the flow-rate method from the ratio of the densities of the heated gas and the gas at room temperature, from the temperatures calculated from spectrograms in the equilibrium section of jet expansion, and from readings of thermocouples located near the nozzle (at high Reynolds numbers Re_*, calculated from the critical parameters). All the methods yielded satisfactory agreement at high Re_*, while at low Re_* preference was given to the values of T_0 determined from the spectra.

An electron beam ∿1 mm in diameter with a current i_b = 1-30 mA and an electron energy E = 10-15 keV was created with an electron gun. Emission of the 00 band of the 1 NS of N_2^+, passing through a monochromator to a photomultiplier, was taken from the region of intersection of the electron beam with the axis of the jet in a direction perpendicular to it. The narrow slit of the monochromator, parallel to the electron beam, permitted the recording of radiation only from the central region of the beam according to [14], assuring that the contribution of secondary electrons from the beam aureole was small.

The influence of the background gas in the present measurements was eliminated both by pumping down to a deep vacuum using helium cryogenic pumps and by approximately equalizing

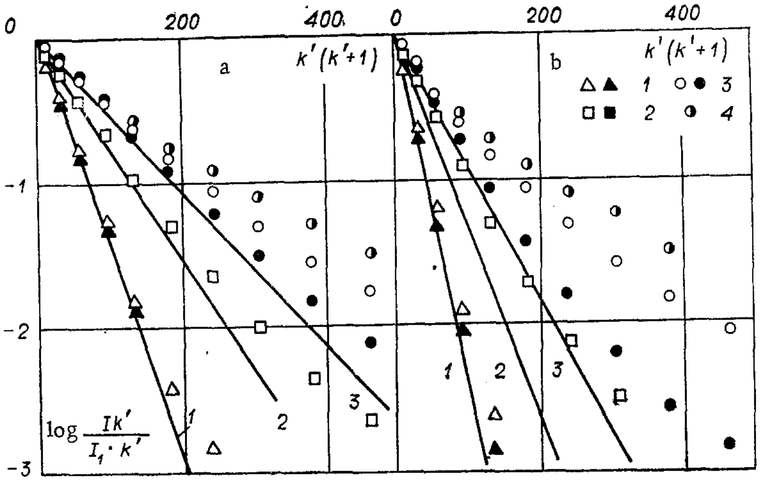

Fig. 1

the temperatures of the gas jet and the background.

A pronounced increase in the stagnation temperature T_0 of the gas allowed us to carry out experiments in the region of high local temperatures, i.e., in the absence of cluster formation.

The investigations of [14] allow us to state that the excitation of the gas of the jet by secondary electrons in the central region of the electron beam has insignificant influence on the distribution in the $N_2^+B^2\Sigma_u^+$ state. At the same time, it was shown in [4, 14] that dipole selection rules for rotational transitions are not satisfied in $N_2X^1\Sigma_g^+ \rightarrow N_2^+B^2\Sigma_u^+$ excitation by primary electrons, i.e., the rotational quantum number can change by $\Delta k = \pm 1, \pm 3, \pm 5, \ldots$.

Thus, performing the experiments on hot gas limited to two the number of factors resulting in the nonequilibrium of spectra of the 00 band of the first negative system of nitrogen obtained in free jets: disturbance of the Boltzmann distribution of rotational energy in the $N_2X^1\Sigma_g^+$ ground state due to the process of rotational relaxation and disturbance of this distribution in excitation to the $N_2^+B^2\Sigma_u^+$ state due to transitions with $\Delta k = \pm 1, \pm 3, \pm 5, \ldots$. To allow for the influence of distortions introduced by the electron beam we used the "multiquantum" model of excitation [15], verified experimentally in a wide range of parameters.

Examples of measured distributions of line intensities in the R branch of the 00 band of the first negative system of N^+ are presented in Fig. 1 in the form of dependences of log $(I_{k'}/I_1 k')$ on $k'(k' + 1)$, where $I_{k'}$ and I_1 are the intensities of the k'-th and first lines and k' is the rotational quantum number in $N_2^+B^2\Sigma_u^+$ (v' = 0). It is well known that $I_{k'} \sim k' \times N_{k'}/(2k' + 1)$ ($N_{k'}$ is the population of rotational levels in the $N_2^+B^2\Sigma_u^+$ state), and therefore straight lines on the graph correspond to a Boltzmann law of distribution of $N_{k'}$. Here x is the distance from the cut of the nozzle of diameter d_* to the observation point. The measurements were made for two distances $x/d_* = 2$ and 4 (Fig. 1a and b, respectively), stagnation temperatures $T_0 = 300$, 600, and 900°K (points 1-3, respectively), and stagnation pressures p_0: $p_0 d_* = 20.6$ Pa·m (dark symbols), 3.7 Pa·m (light symbols), and 1 Pa·m (points 4). Values of log $(I_{k'}/I_1 k')$ calculated from the formula

$$I_{k'}/P_{k'k''} = \sum_k P_{kk'} N_k, \quad k = k' \pm \Delta k, \quad \Delta k = 1, 3, 5\ldots \tag{1}$$

are plotted with solid lines 1 and 3 ($T_0 = 300$, 600, and 900°K, respectively). Here $P_{k'k''}$ are the Hanle–London factors for the emission ($B^2\Sigma \rightarrow X^2\Sigma$); $P_{kk'}$ are the probabilities of rotational transitions from the k to the k' state in the $X^1\Sigma \rightarrow B^2\Sigma$ electronic transition, induced by primary electrons of the beam. In accordance with [15],

$$P_{kk'} = (2k' + 1)\,\Sigma\, P_{0l} \begin{pmatrix} k & l & k' \\ 0 & 0 & 0 \end{pmatrix}^2,$$

where $\begin{pmatrix} k & l & k' \\ 0 & 0 & 0 \end{pmatrix}$ are Wigner three-j symbols. The probabilities of transitions from the zeroth to the l-th state were taken in the form

TABLE 1

No.	T, °K	T_0, °K	x/d_*	$n\cdot 10^{-15}$, cm^{-3}	$p_0 d^*$, Pa·m	No.	T, °K	T_0, °K	x/d_*	$n\cdot 10^{-15}$, cm^{-3}	$P_0 d^*$, Pa·m
1	240	860	1,95	1,9	3,6	4	88	906	9,8	0,4	25
2	242	906	2,03	1,2	25	5	89	293	1,95	0,5	1
3	232	—	—	1,9	—	6	88	—	—	0,8	—

$$P_{0l} = (2l + 1)\alpha^l \Big/ \sum_{l=1,3,5\ldots} (2l + 1)\alpha^l, \quad \alpha = 0{,}28.$$

The values of the relative populations of rotational levels in the $X^1\Sigma$ state in Eq. (1), $N_k = n_k \Big/ \sum_0^\infty n_k$, were assumed to be Boltzmann values with an isentropic temperature T_{is} for equilibrium isentropic expansion of a diatomic gas with $\gamma = c_p/c_V = 1.4$.

DISCUSSION OF RESULTS

It is seen from Fig. 1 that the recorded line intensities differ from their equilibrium isentropic values; at $T_0 > 300$°K the qualitative behavior of the dependences is the same as at room temperature; the detected departure from equilibrium values grows with an increase in rotational level and distance from the nozzle, as well as with a decrease in $p_0 d_*$.

Here we also note the following facts, discovered in analyzing the results of Fig. 1: The logarithms of the relative intensities of upper rotational lines for fixed T_0 lie on straight lines having the same slope for any distances x/d_* (i.e., the "population temperatures" [12] for upper levels are equal to each other and do not coincide with the isentropic temperature), and the number of lines lying on these straight lines grows with a decrease in $p_0 d_*$ and does not depend on x/d_*. These facts are not explained in the article and require further analysis.

Thus, the data presented in Fig. 1 show that the Boltzmann population distribution of rational levels in the $N_2X^1\Sigma_g^+$ state is disturbed in an expanding gas stream.

An even more obvious disturbance of the equilibrium distribution is demonstrated in Fig. 2, where we compare the intensity distributions obtained in a quiescent gas (curves 3 and 6) at the same values of the temperatures T and densities n as in the jet (curves 1, 2, 4, and 5). The conditions under which the experiments represented in Fig. 2 were performed are summarized in Table 1. At about the same densities under thermostatic conditions the influence of secondary electrons is greater than in a free jet [14]. The agreement obtained in the relative intensities of lower levels indicates that the contribution of multiquantum transitions to the recorded spectra is about the same for all the distributions at a fixed temperature. Consequently, for a given gas temperature and density the maximum nonequilibrium caused by the electron beam is reached in the distributions obtained under thermostatic conditions. Therefore, the entire difference between the results obtained for a jet and under thermostatic conditions at upper levels and low p_0 is connected with the process of rotational relaxation taking place with a disturbance of the equilibrium distribution of line intensities.

Because of the difficulties of inverting the matrix for the excitation probabilities with allowance for multiquantum transitions, in Figs. 1 and 2 we present experimental data on the distributions of line intensities in the excited state. However, analogous results were also obtained in the present work for the level populations of the ground state of nitrogen; an example of their variation for a distance $x/d_* = 5$ and a stagnation temperature $T_0 = 293$°K is presented in Fig. 3. Here the population distribution in the $N_2X^1\Sigma_g^+$ state is represented in the usual coordinates of the logarithms of the normalized populations as a function of $k(k + 1)$ for $p_0 d_* = 0.75$, 2.4, 4, and 8 Pa·m (regime numbers from 1 to 4, respectively). The populations N_k were calculated from the experimental data for intensities $I_{k'}$ using the model of multiquantum transitions under excitation by inverse selection, which required large expenditures of machine time in the calculations. As seen from Fig. 3, the population distribution differs from a Boltzmann distribution and as $p_0 d_*$ increases it approaches an equilibrium distribution, which is given by a solid line in Fig. 3, i.e., there is a similarity with the results on intensity.

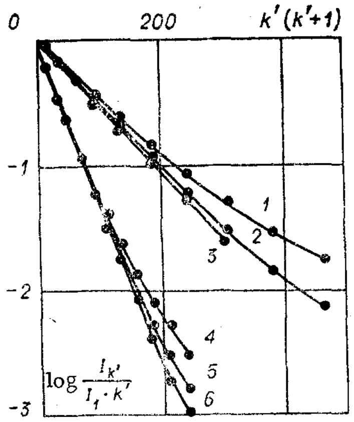

Fig. 2

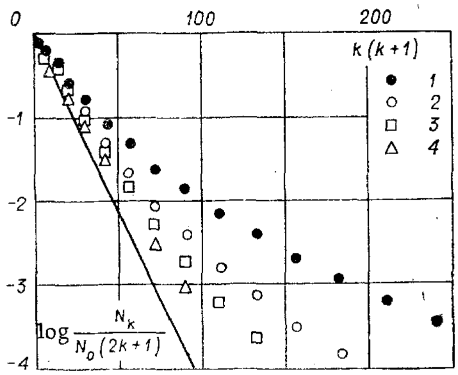

Fig. 3

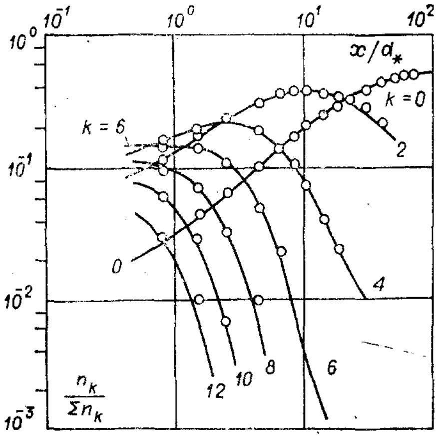

Fig. 4

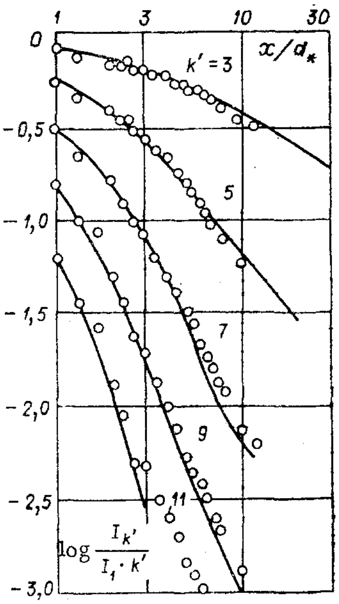

Fig. 5

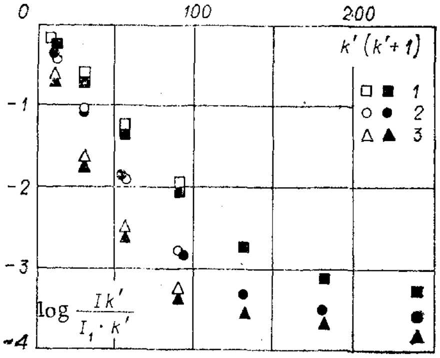

Fig. 6

Let us consider [3, 4], the results of which are at variance with those presented above at first glance. In the first of them a free jet was investigated using Raman scattering. This method permits the direct measurement of the populations of rational levels in the $X^1\Sigma$ ground state. The results of the article are presented in Fig. 4 in the form of $N_k/\Sigma N_k$ as a function of x/d_*. The populations corresponding to an isentropic calculation [16] with $\gamma = 1.4$ made for the regimes of [3] are shown by solid lines. It is seen that there is good agreement between the experimental data of [3], plotted with points, and the calculation ($p_0 d_* = 133$ Pa·m, $T_0 = 293$°K), indicating that the flow at the jet axis is close to equilibrium flow in the investigated regimes.

Because of the above-indicated difficulties in converting the experimentally obtained intensities $I_{k'}$ to populations N_k, we did not make a direct comparison of the experimental data of the present work with the results of [3]. In Fig. 5 we present the logarithms of the relative intensities of rotational lines as a function of x/d_*. The values corresponding to the same isentropic calculation [16] of N_k as in Fig. 4 with a subsequent calculation of $I_{k'}$ from the multiquantum model are shown by solid lines, while the experimental results are shown by points. As follows from the graph, the data obtained from electron-beam measurements reproducing the regimes of [3] also agree well with the isentropic calculation, which is evidence of the correctness of both methods.

Thus, the equilibrium Boltzmann population distributions obtained in [3] do not place the procedure of electron-beam diagnostics under doubt but confirm the latter, since conditions when the departure from equilibrium was negligibly small were satisfied. A similar comparison of results under the conditions of highly nonequilibrium flows with the non-Boltzmann population distribution of rotational levels discovered by the electron-beam method would be of great interest. However, laser methods do not presently possess the required sensitivity in the region of low gas densities, where this nonequilibrium arises.

On the basis of results obtained by the method of electron-beam diagnostics, the authors of [4] state that in a free jet the population distribution of rotational levels is always a Boltzmann distribution, and hence one can introduce the rotational temperature T_r, which varies with variation of p_0d_* from the stagnation temperature as $p_0d_* \to 0$ to the isentropic temperature as $p_0d_* \to \infty$.

In Fig. 6 we give the logarithms of the normalized intensities of rotational lines as a function of $k'(k' + 1)$ for $x/d_* = 4$, 8, and 32 (points 1-3, respectively), obtained under the same conditions in the present work (dark symbols) and in [4] (light symbols). The intensities of the lower lines ($k' \leqslant 9$) agree well with each other, indicating the reproducibility of the data. However, intensities for large k' were not recorded in [4], whereas it is just the upper levels which primarily depart from equilibrium. And this evidently allows the authors of [4] to seek the distribution of rotational levels N_k in the Boltzmann form, although it is evident that, with allowance for the data for $k' > 9$, the population distribution of rotational levels is essentially nonequilibrium.

Thus, in hypersonic jets of polyatomic gases, where large density and temperature gradients arise while the absolute density is low, the finite rate of rotational relaxation and the increasing difference between the energy levels with an increase in the rotational quantum number result not only in a difference between the rotational and translational temperatures but also in a relative overpopulation of the upper rotational levels. Such flows make a good subject for research into the kinetics of nonequilibrium rotational relaxation.

LITERATURE CITED

1. A. P. Clark, A. S. Dickinson, and D. Richardo, "The correspondence principle in heavy-particle collisions," Adv. Chem. Phys., 36, 63 (1977).
2. J. P. Toennies, "The calculation and measurement of cross sections for rotational and vibrational excitation," Ann. Rev. Phys. Chem., 27, 225 (1976).
3. G. Luiks, S. Stolte, and J. Reuss, "Molecular beam diagnostics by Raman scattering," Chem. Phys., 62, 217 (1981).
4. D. Coe, F. Robben, et al., "Rotational temperatures in nonequilibrium free jet expansion of nitrogen," Phys. Fluids, 23, 706 (1980).
5. J. Verberne, I. Ozier, et al., "Molecular beam magnetic resonance study of intra- and intermolecular effects in H_2 in high rotational states," Mol. Phys., 35, 1649 (1976).
6. M. Faubel and E. R. Weinar, "Electron beam fluorescence spectrometry of internal state populations in nozzle beams of nitrogen/rare gas mixtures," J. Chem. Phys., 75, 641 (1981).
7. E. P. Muntz, "Static temperature measurements in a flowing gas," Phys. Fluids, 5, 80 (1962).
8. P. V. Marrone, "Temperature and density measurements in free jets and shock waves," Phys. Fluids, 10, 521 (1967).
9. R. B. Smith, "N_2 first negative band broadening due to electron excitation," in: Rarefied Gas Dynamics: Proceedings of the Sixth International Symposium, Vol. 2, Academic Press, New York (1969).
10. J. C. Lengrand and M. Cloupeau, "Application of Smith model to rotational temperature measurement in nitrogen," AIAA J., 12, 812 (1974).
11. R. G. Sharafutdinov, "Interaction of background molecules with a low density free jet," in: Rarefied Gas Dynamics: Proceedings of the Seventh International Symposium, Vol. 1, Pisa, Italy (1971).
12. N. V. Karelov, A. K. Rebrov, and R. G. Sharafutdinov, "Populations of rotational levels of nitrogen molecules during nonequilibrium condensation in a freely expanding gas," Zh. Prikl. Mekh. Tekh. Fiz., No. 3 (1978).
13. A. A. Bochkarev, E. G. Velikanov, A. K. Rebrov, R. G. Sharafutdinov, and V. N. Yarygin, "Low-density gasdynamic installations," in: Experimental Methods in Rarefied-Gas Dynamics [in Russian], Inst. Teplofiz., Sib. Otd. Akad. Nauk SSSR, Novosibirsk (1974).

14. A. E. Belikov, N. V. Karelov, A. K. Rebrov, and R. G. Sharafutdinov, "Measurements using an electron beam. The role of secondary processes in the excitation of the $B^2\Sigma_u^+$ state of the nitrogen ion," in: Diagnostics of Rarefied Gas Streams [in Russian], Inst. Teplofiz., Sib. Otd. Akad. Nauk SSSR, Novosibirsk (1979); A. K. Rebrov, G. I. Sukhinin, R. G. Sharafutdinov, and Zh.-K. Lengran, "Electron-beam diagnostics in nitrogen. Secondary processes," Zh. Tekh. Fiz., 51, No. 5 (1981).
15. R. G. Sharafutdinov, G. I. Sukhinin, A. E. Belikov, N. V. Karelov, and A. E. Zarvin, "Electron beam diagnostics in nitrogen: multiquantum rotational transitions," in: Thirteenth International Symposium on Rarefied Gas Dynamics: Book of Abstracts, Vol. 1, Nauka, Novosibirsk (1982).
16. R. G. Sharafutdinov and P. A. Skovorodko, "Rotational levels population kinetics in nitrogen free jets," Prog. Astronaut. Aeronaut., 74 (1981).

INFLUENCE OF HIGH-FIELD EFFECTS ON THE CHARACTERISTICS OF THE NEAR-CATHODE LAYER IN A MOLECULAR GAS PLASMA

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UDC 533.932

It is well known that for quite high values of the electric field intensity E the kinetic and transport coefficients of a weakly ionized plasma depend on E. This paper is concerned with estimating the influence of this effect on the characteristics of the charged layer near the cold cathode in the plasma formed by combustion products with a potassium additive under conditions realized in the channels of open-cycle MHD generators.

The distribution of ion and electron densities n_i and n_e and of the electric field intensity E in the near-wall layer of a volume discharge (Debye layer) at the cathode is described by the following nonlinear boundary-value problems [1]:

$$\begin{gathered} J_i' = f, \quad j' = 0, \quad J_i = \mu_i n_i E, \quad J_e = -\mu_e n_e E, \\ j = e(J_i - J_e), \quad E' = 4\pi e(n_i - n_e); \\ y = 0, \; J_e = 0; \; y = y_D, \; E = 0, \; \frac{J_i}{D_{iD}} + \frac{J_e}{D_{eD}} = -2n_D x_D'. \end{gathered} \tag{1}$$

Here J_i, J_e, μ_i, μ_e, D_i, D_e are the diffusion fluxes, mobilities, and coefficients of diffusion of ions and electrons; j is the electric current density (fixed quantity); the terms on the right sides of the first equation and the last boundary condition take into account, respectively, the increase in the number of charged particles due to volume ionization and transport out of the quasineutral region due to concentration diffusion; the y axis is oriented along the normal away from the electrode surface; y_D is the coordinate of the external boundary of the Debye layer; the prime indicates differentiation with respect to y; and the index D indicates the value of the corresponding quantity at $y = y_D$.

In writing down the boundary condition on the cathode surface, it was assumed that there was no emission current.

To solve the problem formulated it is necessary to know the ionization function f and the transport coefficients μ_i, μ_e, D_{iD}, D_{eD}.

The ionization function f can be represented as the sum

$$f = f_1 + f_2, \tag{2}$$

where f_1 corresponds to stepped ionization of the atoms of the additive with the participation of heavy particles (primarily water molecules) and electrons; f_2 describes direct ionization of heavy particles by electron bombardment.

The quantity f_1 in the limit of a weak field (at an electron temperature T_e equal to the gas temperature T) can be represented, with acceptable accuracy, as a sum [2]

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