ELECTRON.CONCENTRATION DISTRIBUTION IN A POWERFUL VOLUME DISCHARGE WITH IONIZATION OF THE GAS BY AN ELECTRON BEAM

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UDC 533.915

Ionizing a gas with a beam of high-energy electrons turned out to be a useful method for creating conductivity in a gas, ensuring a high density of absorbed energy in powerful lasers at high pressure [1, 2]. At the present time, studies of the spatial homogeneity of the electrical discharge parameters are of fundamental significance. This stems primarily from the necessity of exciting large volumes of optically active media, formed by highquality radiation fluxes, etc. The uniformity with which the gas is ionized also has a large effect on the stability of the volume discharge and its limiting energy characteristics [3].

In order to determine the conditions for the formation of a homogeneous discharge, numerical simulation is most widely used. However, in calculations of the ionization of a gas by fast electrons, the real geometrical dimensions and the energy and angular spectra of the electron beam formed in the diode of an accelerator have not been taken into account [4-8]. Experimental studies of the local electrical parameters of a high-voltage volume discharge at pressures of p $\sim 10^5$ Pa encounter serious difficulty [9].

In this paper, we present for the first time, using the method of optical interferometry, experimental measurements of the electron concentration distribution n(x, t) in a powerful volume electroionization discharge in N₂ and air (here, x is the cathode-anode direction). The rate of ionization of the gas by the electron beam is determined and the effect of the electric field, the elements constituting the discharge gap, and the mechanism for loss of free electrons on the distribution n(x, t) is investigated.

The main attention is focused on the possibility of using, for uniform ionization of a gas, the typical scheme of an accelerator operating on the burst-emission principle with a capacitive energy accumulator, for which a pulsed voltage generator is used. We note that the given scheme for feeding the accelerator, which is usually used for forming powerful electron fluxes with current density $j_b = 1\text{-}100 \text{ A/cm}^2$ and duration $\sim 10^{-8}\text{-}10^{-6}$ sec, has been widely used in electroionizing laser systems [1-3]. However, it has an important characteristic that greatly affects the ionization of the gas. As the capacitive accumulator discharges, the voltage U_b on the diode cathode decreases, which decreases the average energy and, therefore, the mean free path of fast electrons in the gas as well. The pulse duration of the transmission of the foil, through which the electrons are injected into the gas, decreases sharply. Thus, it was interesting to study the possibility of using a beam of electrons, whose average mean-free path R_e in the gas varied during the pulse from R_e \gg d (d is the distance between electrodes) to R_e \ll d, for creating uniform ionization of the gas in the discharge gap.

The investigations were carried out in the discharge chamber D (Fig. 1) of a powerful CO₂ electroionization laser with a radiation energy of 1 kJ [2]. The gas was ionized by the electron beam with an average current density $j_b \approx 0.75 \text{ A/cm}^2$, a pulse duration of 0.75 µsec, and a cross section of 100×10 cm, which was injected into the gas from the vacuum diode (residual pressure p = 10^{-3} Pa) through a titanium foil of 50-µm thickness. A pulsed Marx generator with the following parameters was used as the energy source: starting voltage Ub ≈ 200 kV, working capacitance Cb ≈ 0.046 µF, wave impedance 3 Ω.

Typical current-density oscillations in the beam injected into the gas in the discharge gap and the voltage on the cathode of the vacuum diode are shown in Fig. 2a, b. A pulsed voltage generator served as the source of energy absorbed in the discharge: $U_{dis} = 100-250$

Novosibirsk. Translated from Zhurnal Prikladnoi Mekhaniki i Tekhnicheskoi Fiziki, No. 6, pp. 3-9, November-December, 1981. Original article submitted December 25, 1980.



Fig. 2

kV, capacitance $C_{dis} \approx 0.45 \ \mu\text{F}$, wave impedance $\rho \approx 1.5 \ \Omega$. The distance between the electrodes was d \approx 9.8 cm. The cathode was constructed in the form of a grid, which was placed 1 cm from the output foil of the electron accelerator.

Figure 1 shows a schematic diagram of the scheme for measuring the electron concentration in a powerful volume discharge at atmospheric pressure of the gas, similar to that described in [10, 11]. A doubled Mach-Zender interferometer scheme with a common measuring arm, which permitted carrying out the measurements simultaneously at two wavelengths, 10.6 and 0.63 µm, was used. We note that the use of a two-wavelength interferometer stems from the large contribution of the neutral component of the discharge to the change in the index of refraction of the active medium over the time that energy is absorbed [11]. The electron concentration in this case was determined from the expression [12]

$$n \approx 2.23 \cdot 10^{13} \left(\lambda_1 \delta_1 - \lambda_2 \delta_2 \right) / \left(\lambda_2^2 - \lambda_1^2 \right) L,$$

where λ_1 and λ_2 are the radiation wavelengths; δ_1 and δ_2 are increases in phase in fractions of the band. Here, the quantities $\lambda_{1,2}$ and L are expressed in centimeters. The error in measuring n was $\approx 14\%$.

A 10-mW (at $\lambda_1 = 0.63 \mu$ m) LG-126 He-Ne laser Ll and a 5-W, water-cooled, continuous, CO₂ laser L2 ($\lambda_2 = 10.6 \mu$ m) with a 1-m active medium and change of working gas were used as the sources of radiation. Before the beginning of the experiment, the lasers were brought into a steady state over the course of 1.5 h. No special measures were taken to stabilize the frequency or phase of the radiation and the output power. Phase control and amplitude modulation were carried out by changing the optical length of the reference beam in the interferometer with the help of rotating plane-parallel plates K1 and K2 [11].

The maximum diameter of the light beam, propagating along the direction L = 100 cm parallel to the electrodes, was 0.4 cm, thereby determining the spatial resolution of the scheme.

An FEU-83 photomultiplier Pl was used to detect the $\lambda_1 = 0.63 \ \mu m$ radiation. The radiation was incident on the photocathode through a diaphragm \emptyset 0.3 mm. A set of light filters (F1, F2) was used both to suppress the parasitic lighting and to put the sensors into the linear mode for detecting laser radiation. A GeAu photoresistor P2 at T = 77°K with a working surface of 3 × 3 mm served as a $\lambda_2 = 10.6$ - μm radiation detector. The time resolution of the measuring circuit with the radiation detectors was $\tau \leq 100$ nsec.

Figure 3 shows the typical time dependence of the electron concentration, measured in nitrogen with $U_D \approx 90$ kV for different distances from the foil in the electron accelerator



(curves 1-4 correspond to x = 3, 6.3, 8.6, and 10.1 cm). For comparison, the function n(t) (curve 5) for x = 6.3 cm and $U_p = 0$ is also presented. The change with time of the average mean-free path of fast electrons R_e , formed in the diode, is shown at the bottom of Fig. 3. The vertical lines show the error in determining R_e . The magnitude of R_e was obtained from the data in [13] for electron energy $\mathcal{B}_b(0, t)$, determined by the voltage on the cathode $U_b(t)$ taking into account the energy loss on transmission through a 50-µm Ti foil. It is evident in Figs. 2 and 3 that as the cathode voltage decreases, the mean-free path of electrons drops and, in addition, for t > 0.3 µsec, $R_e < d$. This determined the difference in the time for attaining a maximum in n at different distances from the foil. Thus, for x = 3 cm, the maximum value of n is observed at t \approx 0.45 µsec, while for x \approx 8.6 cm, at t \approx 0.3 µsec. The breakdown of this behavior for n measured near the anode should be noted. As can be seen from Fig. 3, for x = 10.1 cm, i.e., at a distance 0.7 cm from the anode, the value of n increases, and therefore, the gas continues to ionize up to the time t \approx 0.5 µsec.

The corresponding distributions n(x) with respect to the length of the discharge gap at different times are presented in Fig. 4 [points 1-6 correspond to t = 0.2, 0.3, ..., 0.6, 0.8 µsec, respectively for air (a) and nitrogen (b)]. It is evident from Fig. 4 that the value of n decreases with motion from the cathode to the anode, i.e., in the direction of motion of the beam. The degree of nonuniformity in the distribution of the electron concentration $\xi = n_{+}/n_{-}$ (where n_{+} , n_{-} are the maximum and minimum values of n in the discharge gap) attains $\xi \approx 2$ already at t = 0.2 µsec, i.e., for $R_{e} > d$. Apparently, this is related to the beam divergence. Indeed, in accordance with the data in [14], under the conditions of our experiment, in the course of the entire current pulse of low-energy electrons the divergence, stemming from scattering at the outlet foil, is determined by the stationary angular distribution for which <cos $\theta > \approx 0.7$. This decreases the current density



by approximately a factor of two with motion of electrons up to the anode, and therefore, the ionization rate.

The maximum nonuniformity in the distribution of n both in N₂ and in air is attained at the time t \approx 0.5-0.6 µsec, i.e., as can be seen from Fig. 2, practically by the end of the action of the external ionizer. However, the degree of nonuniformity in ξ differs in these gases. The quantity $\xi \approx 4.2$ in air exceeds the corresponding value $\xi \approx 2.5$ in N₂. It is important to note that further evolution of the distribution n(x, t) also differs in these gases. If the degree of nonuniformity for t > 0.5 µsec in air remains practically constant, then in N₂ the electron concentration distribution is observed to level out. Thus, the degree of nonuniformity in N₂ decreases from $\xi \approx 2.5$ at t = 0.5 µsec to $\xi \approx 1.5$ at t = 0.8 µsec.

Apparently, the observed difference in the distribution n in N₂ and in air stems from different mechanisms for loss of free electrons. In air, for which attachment of electrons to the O₂ molecule is characteristic, the stationary electron concentration n₀ depends more strongly on the distribution $\psi(x, t) (n_0 \approx \psi/\eta N_0, \psi$ is the ionization rate, η is the attainment coefficient, and N₀ is the concentration of O₂ molecules) than in nitrogen with its recombination mechanism $[n_0 \approx (\psi/\beta)^{1/2}, \beta$ is the recombination coefficient]. In addition, the characteristic time for n to change in the presence of recombination $\tau \approx 1/(\beta \psi)^{1/2}$ in contrast to the attachment mechanism $(\tau \approx 1/\eta N_0)$ decreases with increasing ψ , which additionally stabilizes n. Therefore, recombination leads to considerable smoothing of the nonuniformity in n(x), stemming from nonuniform distribution $\psi(x)$.

The effect of the electric field on the electron concentration distribution and the ionization rate is shown in Fig. 5. The functions n(x) and $\psi(x)$ in N₂ for E/N = 0 (points 1) and the average value E/N = $36 \cdot 10^{-17}$ V·cm² (points 2) are shown. The data correspond to t \approx 0.3 µsec, i.e., R_e \approx d, j_b \approx 0.75 A/cm² with a diode voltage of \approx 150 kV (here, N is the starting gas concentration).

The value of $\psi(\mathbf{x}, t)$ in N₂ was determined from the expression

(it is easy to show that for the conditions of our experiment, the term ${\rm div}\; j_e,$ where j_e is the discharge current density, $j_e\approx j_p,$ can be neglected in the electron balance equation).

The recombination rate $\beta = f(E/N)$ of electrons in nitrogen was measured beforehand. The decay of the plasma after the external ionizer was switched off was studied in a constant field (E/N = const) and with a uniform distribution n in the discharge gap [15]. The values of β are shown in Fig. 6 as a function of E/N. The continuous line corresponds to the function $\beta = 1.7 \cdot 10^{-7} W^{-0.85}$, where W is the average electron energy in electron volts, determined as a function of E/N from data in [16]. It should be noted that in practice, even in specially purified nitrogen with an impurity content $\leq 0.2\%$ (O_2 , H_2O), there is some effective recombination coefficient, which stems from the complicated molecular composition of ions: N_2^+ , N_3^+ , N_4^+ , $N_2O_2^+$, NO⁺, and so on, which form at a high rate $\sim 10^9 sec^{-1}$ in a discharge at atmospheric pressure [17].

It is evident from Fig. 5 that switching off the field sharply increases the uniformity of the distributions n(x) and $\psi(x)$. The value of ξ increases from 2 to 6.5. For comparison, Fig. 5 shows (curves 3 and 4) the ionization rate ψ , calculated from numerical simulation data on the distribution of energy transferred by the beam electrons to the gas. In order to determine ψ , we used the expression

 $\psi = k_{jb} D(x) / ek_2 \varepsilon,$

where j_b is the beam current density behind the foil in the electron accelerator; $k_1 \approx 0.5$ is the transmittance of the cathode grid in the discharge gap; $k_2 \approx 0.1$ [18] is the transmission of 150 keV electrons through a Ti foil with a thickness of 50 µm; $\epsilon \approx 35$ eV [13] is the energy loss on creating electron—ion pairs in N₂; D(x) is the distribution of energy losses in the gas, calculated per fast electron incident on the foil. The values of D(x) for $U_b \approx 150$ keV, a 50 µm Ti foil with an average magnitude of the electric field in the gas E/N = 0, 15.1 \cdot 10^{-17} V·cm² (curves 3, 4, respectively), are taken from [7].

It is evident from Fig. 5 that the real inhomogeneity of the distribution $\psi(x)$, determined experimentally, greatly exceeds the data obtained by numerical simulation. This apparently stems primarily from the additional decrease in the current density with the motion of fast electrons to the anode due to the divergence of a finite beam, which was not taken into account in the calculations in [7]. The nonuniformity in the ionization of the gas is also affected by scattering of electrons by elements in the structure, for example, by the cathode grid in the discharge gap.

Thus, the experimental data show that the quite uniform $(\Delta\psi/\psi\leqslant 0.2)$ distribution in the intensity of external ionization can be created at a distance $x\leqslant R_e/2$ and only in the presence of an electric field compensating the ionization losses.

The small effect of electrons, scattered by the anode, on the distribution n(x) should be noted. Reflected fast electrons move opposite to the external electric field, which leads to an additional decrease in their mean free path in the gas. For this reason, in air, for which strong inhomogeneity in the distribution n is observed and, therefore, localization of the electric field in the vicinity of the anode, the reflected flux only slows down the decrease in n as the anode is approached (see Fig. 4). The effect of the anode is most clearly manifested in the absence of an electric field, but in this case as well, the characteristic size of the region of action of the scattered electrons is limited at p $\,\approx\,10^{\,\rm s}$ Pa to $\,\approx\,2\,$ cm (see Fig. 5). The presence of ionization processes near the anode for t > 300 nsec (average mean-free path of fast electrons $R_e < d$, see Figs. 3 and 4) could stem from electron impact ionization in a discharge growing with strong field distortion and increase of the parameter $E/N \ge 10.6 \cdot 10^{-16} \text{ V} \cdot \text{cm}^2$ in this region [9], as well as an acceleration of high-energy fast electrons, for which the electric field E compensates the ionization losses. However, the contribution of these processes is significant directly near the anode $\Delta x \lesssim$ 2 cm and does not effect the distribution n in the discharge gap. Thus, scattering of the electron beam by the electrode surfaces in dense gases can apparently greatly affect the preelectrode phenomena in a discharge, as well as the ionization of the gas for small (d \leq 4 cm) interelectrode gaps.

The experimental data and the analysis of the main mechanisms of formation of the electron concentration presented above show that the accelerator with a capacitive supply source, which forms a beam with decreasing electron energy, cannot be used without special measured taken to achieve uniform ionization of the gas in the discharge gap of a powerful laser. As noted in [9], it is necessary to maintain the electron energy above a critical value, for which $R_e > 2d$, over the course of the entire current pulse. For gas mixtures with the N₂ content predominating and, therefore, a recombination electron loss mechanism, a promising method for creating a uniform discharge could be the use of short powerful beams ($\tau \approx 10^{-8}$ sec) and energy absorption in the plasma decay regime.

Thus, in this work, the electron concentration distribution n(x, t) in a powerful volume discharge with electron-beam ionization of the gas was measured for the first time using the method of optical interferometry. The positive effect of recombination compared to attachment on the formation of a uniform distribution n was demonstrated. The weak effect of scattering of fast electrons by the anode on the ionization of the gas in large volumes (d \ge 10 cm) at a pressure of p \approx 10⁵ Pa is noted. The distribution of the rate of electron-beam ionization of the gas in large volumes in the presence and absence of an electric field E in the discharge was determined experimentally. It was shown that a quite uniform distribution of electron concentration n(x, t) can be created at a distance of $x \leq i$ $R_{e}/2$ and only in the presence of an electric field E that compensates the ionization losses in the beam.

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ROTATIONAL RELAXATION IN THE TRANSITION REGIME OF FREE NITROGEN

JETS

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UDC 533.6.011.8

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 $N_k = N_o(2k + 1)\exp[-k(k + 1)\Theta/T_k]$ substituting the measured values of the population densities of the k-th N_k and zeroth N_o rotational levels (Θ is the characteristic temperature). It has been found [3] that the temperature sof 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.

Novosibirsk. Translated from Zhurnal Prikladnoi Mekhaniki i Tekhnicheskoi Fiziki, No. 6, pp. 9-16, November-December, 1981. Original article submitted December 23, 1980.

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