U.S.S.R. Comput. Maths. Math. Phys. Vol. 2, pp. 182–190. 1980
Printed in Great Britain.

0041–5553/80/020182–09$07.50/0

NUMERICAL INVESTIGATION OF THE PROBLEM OF LASER BREAKDOWN OF A DENSE GAS*

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(*Received* 19 *January* 1979)

A MATHEMATICAL model is constructed of breakdown by laser radiation of a dense cold gas close to a metal surface. A method for the numerical solution of the kinetic and energy equations allowing for transfer processes is developed, and computer calculations are carried out using it.

1. In a series of experiments [1–3] the conditions for the breakdown of inert gases and nitrogen by the radiation of a neodymium laser and features of the interaction of laser radiation with solids at low flow densities and high pressures of the surrounding medium have been investigated. The experiments showed that a breakdown close to metal surfaces occurred without noticeable mechanical damage, that is, without extensive vapourization of the target material, and the irradiated surfaces under the influence of the dense plasma changed their physical properties (for example, the microhardness of steel in a nitrogen medium increased by a factor of 2 to 3 in the irradiated zone).

However, the extremely high speed of the processes taking place, the rigid construction of the camera, which must withstand pressures of hundreds of atmospheres, and also the fact that the plasma being formed is opaque, make it difficult to obtain exhaustive information experimentally. The experimental data about the initial stage of formation of the plasma that is most difficult to obtain is the breakdown of a cold gas. At the same time this stage is of great interest, since the mechanism of breakdown in a cold gas at such low radiation fluxes is not completely understood. For this reason the numerical modelling of these processes has become urgent. In turn numerical modelling encounters a number of difficulties the basis of which is the fact that in the problem of laser breakdown in gases it is required that a whole series of physicochemical processes with characteristic times differing by several orders of magnitude be traced, that is, it is required that, against a background of slow processes such as the diffusion of charged particles in a dense gas, high-speed processes be distinguished, for example, the excitation and multistage ionization of particles. The numerical realization of the problem is also complicated by the fact that the solution is of an exponentially increasing nature, and for this reason questions of approximation and stability become of paramount importance.

In this paper we consider a method for the numerical solution of the system of equations describing the phenomenon of breakdown in a dense atomic gas.

It is necessary to note that this method can easily be generalized to the case of molecular gases, the same mathematical difficulties being encountered in the solution of such problems [4]. However, molecular gases involve a greater number of chemical reactions and are accordingly described by a greater number of equations. It is therefore more practical to develop the method for the simpler model for atomic gases.

*Zh. vychisl. Mat. mat. Fiz., **20**, 2, 451–460, 1980.

2. The system of equations describing the kinetics of collisions in atomic gases, the exchange of energies and transfer processes is written as follows:

$$\frac{\partial N}{\partial t} = -(k^+N-\beta^+N^+N_e)N_e-(k^*N-\beta^*N^*)N_e, \tag{1}$$

$$\frac{\partial N^*}{\partial t} = (k^*N-\beta^*N^*)N_e-(k^{*+}N^*-\beta^+N^+N_e)N_e, \tag{2}$$

$$\frac{\partial N_e}{\partial t} = (k^+N-\beta^+N^+N_e)N_e-(k^{*+}N^*-\beta^+N^+N_e)N_e+\frac{\partial}{\partial x}D_a\frac{\partial N_e}{\partial x}, \tag{3}$$

$$\frac{3}{2}\frac{\partial(N_eT_e)}{\partial t} = \left[\frac{4\pi e^2G}{mc\omega^2}-\frac{3}{2}\Lambda(T_e-T_g)\right](\nu_{en}+\nu_{ei})N_e \tag{4}$$
$$-I^*(k^*N-\beta^*N^*)N_e-I^+(k^+N-\beta^+N^+N_e)N_e$$
$$-I^{*+}(k^{*+}N^*-\beta^+N^+N_e)N_e+\frac{\partial}{\partial x}\varkappa_e\frac{\partial T_e}{\partial x},$$

$$\frac{\partial(N_gT_g)}{\partial t} = \Lambda(T_e-T_g)(\nu_{en}+\nu_{ei})N_e. \tag{5}$$

To simplify the problem somewhat we will assume that the discharge takes place in conditions of quasi-neutrality, that is, $N_e \sim N_i$, where N_e, N_i are the electron and ion densities, and we also disregard diffusion and the thermal conduction of the neutral component and losses by radiation.

Equation (1) is the balance equation for the population of the ground level, (2) is the balance equation for the first excited level, (3) is the balance equation for the electron density (4) is the energy balance equation for the electrons, and (5) is the energy balance equation for the heavy particles.

The system of equations (1)–(5) is supplemented by the boundary conditions at the points x=0 and x=L; x = 0 is a point on the surface of the plate. The distance from the right boundary was chosen arbitrarily: $\sim$ 100 μ. At the left boundary x = 0 the surface temperature and the thermoelectric current are determined. The temperature of the target surface is determined by solving the heat conduction problem for a semi-infinite body on which a source of constant intensity G_n acts. Richardson's formula [5] is used to estimate the thermoelectric current from unit surface. It is convenient to specify the boundary conditions on the right boundary x=L as relations connecting the unilateral flows of electrons and heat with the density and temperature functions:

$$\begin{aligned}
&x=0,\\
&T_e(0,t)=T_g(0,t)=2G_n(at/\pi)^{1/2}/\lambda,\\
&D_e(0,t)\frac{\partial N_e(0,t)}{\partial x} = BT_e^2(0,t)\exp\left[-\frac{\varphi}{T_e(0,t)}\right];\\
&x=L,\\
&D_a(L,t)\frac{\partial N(L,t)}{\partial x} = -\frac{1}{2}v_eN_e(L,t),\\
&\varkappa_e(L,t)\frac{\partial T_e(L,t)}{\partial x} = -\frac{1}{2}v_eN_e(L,t)T_e(L,t).
\end{aligned} \tag{6}$$

The following notation is used: T_e and T_g are the temperatures of the electrons and heavy particles, t is the time, x is the spatial coordinate, h is the x-step, τ is the t-step, c is the speed of light, e is the electron charge, ω is the frequency of the laser radiation, κ_ν is the coefficient of the braking absorption, $\upsilon_{ei}, \upsilon_{en}$ are the frequencies of elastic collisions of electrons with ions and atoms, D_e, D_i, D_a are the coefficients of electronic, ionic and ambipolar diffusion, I^*, I^+, I^{*+} are the excitation and ionization energies of atoms from the ground and excited states, k^*, k^+, k^{*+} are the coefficients of the excitation and ionization velocities, $\beta^*, \beta^{*+}, \beta^+$ are the coefficients of the de-excitation and recombination velocities (it being assumed that $\beta^{*+} = \beta^+$), v_g, v_e are the mean thermal velocities of the atoms and electrons, σ is the cross-section of elastic collisions of electrons with atoms, A is the atomic weight, κ_e is the electronic thermal conductivity, m, M are the masses of an electron and an atom, σ_{ei} is the cross-section of elastic collisions of ions with atoms, κ_ν is the coefficient of absorption of laser radiation, G_0 is the flux density of the laser radiation, G_n is the flux of radiation absorbed by the target, G is the total flux of radiation taking into account the reflection from the target, B is the thermionic emission constant, φ is the work function, and λ and a are the thermal conductivity and thermal diffusivity of the target.

The following expressions and numerical values were used:

$$D_e = v_e/\sigma N, \quad D_i = v_a/\sigma N, \quad D_a = D_i(1+T_e/T_g),$$

$$v_e = 6.7\cdot 10^7\, T_e^{1/2}, \quad v_a = 1.56\cdot 10^6 (T_g/A)^{1/2},$$

$$G_n = RG_0 \exp\left(-\int \kappa_\nu dx\right), \quad G = (2-R)\, G_0 \exp\left(-\int \kappa_\nu dx\right),$$

$$\kappa_\nu = \frac{16\pi^2}{3\sqrt{3}} \frac{e^6 Z^2 k T_e}{h^4 c \nu^3} \exp\left(-\frac{I^+ - h\nu}{kT_e}\right), \quad \Lambda = \frac{2m}{M}, \quad R = 0.2,$$

$$\kappa_e = \frac{5}{2}\frac{v_e N_e}{\sigma N}, \quad \hbar\omega = 1.17 \text{ эВ}, \quad I^+ = 14.58 \text{ эВ},$$

$$I = 10.3 \text{ эВ}, \quad I^{*+} = I^+ - I^*, \quad \varphi = 4.1 \text{ эВ}, \quad a = 0.54 \text{ см}^2\cdot\text{с}^{-1},$$

$$B = 60.2 \text{ А}\cdot\text{см}^2\cdot\text{град}^{-2}, \quad \lambda = 1.4 \text{ Вт}\cdot\text{см}^{-1}\cdot\text{град}^{-1}.$$

3. The systems of equations describing the step-by-step ionization, recombination and exchange of energies between the particles are usually referred to stiff systems for which the eigenvalues of the Jacobi matrix have negative real parts and a large spread of absolute values. Gear's method [6–8] is well recommended for solving stiff systems of ordinary differential equations.

With the aim of applying Gear's method to solve system (1)–(5), the partial differential equations are transformed by the straight-line method into a system of ordinary differential equations with difference ratios on the right side (the number of resulting equations is $M\times K$, where M is the number of equations in the original system and K is the number of internal nodes of the mesh):

$$\frac{dN}{dt_1} = [-(k^+N - \beta^+N^+N_e)N_e - (k^*N - \beta^*N^*)N_e]_1, \tag{7}$$

$$\frac{dN^*}{dt_1} = [(k^*N - \beta^*N^*)N_e - (k^{*+}N^* - \beta^+N^+N_e)N_e]_1,$$

$$\frac{dN_e}{dt_1} = [(k^+N - \beta^+N^+N_e)N_e + (k^{*+}N^* - \beta^+N^+N_e)N_e],$$

(Cont'd)

$$+\frac{1}{\hbar_i}\left[\frac{D_{a,i+1}+D_{ai}}{2}\frac{N_{e,i+1}-N_{ei}}{h_{i+1}}-\frac{D_{ai}+D_{a,i-1}}{2}\frac{N_{ei}-N_{e,i-1}}{h_i}\right],$$

$$\frac{3}{2}\frac{d(N_eT_e)}{dt_i}=\left\{\left[\frac{4\pi e^2G}{mc\omega^2}-\frac{3}{2}\Lambda(T_e-T_g)\right](\nu_{en}+\nu_{ei})N_e-I^+(k^+N\right.$$

$$\left.-\beta^+N^+N_e)N_e-I^\cdot(k^\cdot N-\beta^\cdot N^\cdot)N_e-I^{\cdot+}(k^{\cdot+}N^\cdot-\beta^+N^+N_e)N_e^+\right\}$$

$$+\frac{1}{\hbar_i}\left[\frac{\varkappa_{e,i+1}+\varkappa_{ei}}{2}\frac{T_{e,i+1}-T_{ei}}{h_{i+1}}-\frac{\varkappa_{ei}+\varkappa_{e,i-1}}{2}\frac{T_{ei}-T_{e,i-1}}{h_i}\right],$$

$$\frac{d(N_gT_g)}{dt_i}=[\Lambda(T_e-T_g)(\nu_{en}+\nu_{ei})N_e]_i,$$

$$\hbar_i=\frac{h_{i-1}+h_i}{2},\qquad 1\leqslant i\leqslant K.$$

However, the application of Gear's method to (7) does not yield the desired results. The integration was carried out only with a small step and first-order accuracy. On increasing the step τ or the order of the method the solution became unstable. The physical cause of this is as follows. The phenomenon of the breakdown of a dense gas is characterized by the fact that a slight perturbation of the electron density in some section leads to its exponential growth with time, that is, the speed of the ionizing build-up of the perturbation exceeds the speed of its diffusive leakage. The exponential build-up of the solution of system (1)–(5), as is shown by numerical solution of the problem of finding the eigenvalues for the simplified linearized system (1)–(5), is described by eigenvalues Re $\lambda_i > 0$. Therefore, system (1)–(5) is not stiff in the sense indicated above. Moreover, the dependence of the coefficients κ_e, D_e on the temperature and density of the particles leads to the diffusion terms becoming the principal source of error in the solution of system (7). The methods of predictor–corrector type used in Gear's method use the values from the layer f and below to advance t by one step, that is, to pass from the layer j to the layer j+1. It is known [9] that if the values of κ_e and D_e are taken from the layer j, then the solution of the quasilinear equations of diffusion and heat conduction have high accuracy. The existence of a close connection between the equations of kinetics and energy leads to the error committed in the calculation of $\partial N_e/\partial t$, being transferred directly into Eq. (4), where N_e is under the sign of the derivative $\partial/\partial t$. In conditions of exponential growth of the solution this leads to instability.

A more efficient method for solving system (1)–(6) is the method of cumulative approximation [9], in which the process of finding an approximate solution of a complex problem is split into several stages at each of which a simpler problem is solved. For this purpose the boundary value problem (1)–(6) was represented as two problems to be solved successively: for the kinetics and exchange of energy due to collisions we have the problem

$$\frac{\partial N}{\partial t}=-(k^+N-\beta^+N^+N_e)N_e-(k^\cdot N-\beta^\cdot N^\cdot)N_e, \tag{8}$$

$$\frac{\partial N^\cdot}{\partial t}=(k^\cdot N-\beta^\cdot N^\cdot)N_e-(k^{\cdot+}N^\cdot-\beta^+N^+N_e)N_e,$$

$$\frac{\partial N_e}{\partial t}=(k^+N-\beta^+N^+N_e)N_e+(k^{\cdot+}N^\cdot-\beta^+N^+N_e)N_e,$$

$$\frac{3\partial(N_eT_e)}{2\partial t}=\left[\frac{4\pi e^2G}{mc\omega^2}-\frac{3}{2}\Lambda(T_e-T_g)\right](\nu_{ei}+\nu_{en})N_e-I^+(k^+N-\beta^+N^+N_e)N_e \qquad \text{(Cont'd)}$$

$$-I^\cdot(k^\cdot N-\beta^\cdot N^\cdot)N_e-I^{\cdot+}(k^{\cdot+}N^\cdot-\beta^+N^+N_e)N_e,$$

$$\frac{\partial(N_gT_g)}{\partial t}=\Lambda(T_e-T_g)(\nu_{ei}+\nu_{en})N_e$$

and for the transfer processes the problem

$$\frac{\partial N_e}{\partial t}=\frac{\partial}{\partial x}D_a\frac{\partial N_e}{\partial x},\qquad \frac{\partial(N_eT_e)}{\partial t}=\frac{\partial}{\partial x}\varkappa_e\frac{\partial T_e}{\partial x}. \tag{9}$$

Using the straight-line method system (8) is transformed into the difference system:

$$\frac{dN}{dt_i}=[-(k^+N-\beta^+N^+N_e)N_e-(k^\cdot N-\beta^\cdot N^\cdot)N_e]_i,$$

$$\frac{dN^\cdot}{dt_i}=[(k^\cdot N-\beta^\cdot N^\cdot)N_e-(k^{\cdot+}N^\cdot-\beta^+N^+N_e)N_e]_i,$$

$$\frac{dN_e}{dt_i}=[(k^+N-\beta^+N^+N_e)N_e+(k^{\cdot+}N^\cdot-\beta^+N^+N_e)N_e]_i,$$

$$\frac{3}{2}\frac{d(N_eT_e)}{dt_i}=\left\{\left[\frac{4\pi e^2G}{mc\omega^2}-\frac{3}{2}\Lambda(T_e-T_g)\right](\nu_{en}+\nu_{ei})N_e-I^+(k^+N\right. \tag{10}$$

$$\left.-\beta^+N^+N_e)N_e-I^\cdot(k^\cdot N-\beta^\cdot N^\cdot)N_e-I^{\cdot+}(k^{\cdot+}N^\cdot-\beta^+N^+N_e)N_e\right\}_i,$$

$$\frac{d(N_gT_g)}{dt_i}=[\Lambda(T_e-T_g)(\nu_{en}+\nu_{ei})N_e]_i.$$

For Eqs. (9) a conservative difference scheme is written down [10], which is a system of two second-order difference equations:

$$\frac{N_{ei}^{j+1}-N_{ei}^{j}}{\tau}=\frac{1}{\hbar_i}\left[\frac{D_{a,i+1}^{j+1}+D_{ai}^{j+1}}{2}\frac{N_{e,i+1}^{j+1}-N_{ei}^{j+1}}{h_{i+1}}\right.$$

$$\left.-\frac{D_{ai}^{j+1}+D_{a,i-1}^{j+1}}{2}\frac{N_{ei}^{j+1}-N_{e,i-1}^{j+1}}{h_i}\right], \tag{11}$$

$$\frac{(N_eT_e)_i^{j+1}-(N_eT_e)_i^{j}}{\tau}=\frac{2}{3}\frac{1}{\hbar_i}\left[\frac{\varkappa_{e,i+1}^{j+1}+\varkappa_{ei}^{j+1}}{2}\frac{T_{e,i+1}^{j+1}-T_{ei}^{j+1}}{h_{i+1}}\right.$$

$$\left.-\frac{\varkappa_{ei}^{j+1}+\varkappa_{e,i-1}^{j+1}}{2}\frac{T_{ei}^{j+1}-T_{e,i-1}^{j+1}}{h_i}\right].$$

The general difference scheme is a chain of one explicit (10) and one implicit (11) difference scheme. The transition from the layer j to the layer $j+1$ is made via an auxiliary layer $j+½$. The first problem (11) is solved, and then with the results obtained as initial values system (10) is solved.

To solve (10) Nordsieck's method, provided for in Gear's algorithm [11, 12], is used. The three-point difference problem (11) is solved by the method of separate pivital condensations with iterations [13].

The application of the method of cumulative approximation permits both systems of equations (10) and (11) to be computed more accurately than (7). The increased accuracy of the solution of the non-linear system (9) is achieved because of the iterative procedures used in the solution. The segregation of the transfer processes in a separate problem permits the kinetic system of equations to be computed with a higher order of accuracy $p \leqslant 4$.

4. As a model example and to develop the method for the numerical solution of problem (1)–(6), calculations were carried out for atomic nitrogen, for whose coefficients of the rates of excitation and ionization approximate analytic expressions exist in $cm^3 \cdot c^{-1}$ (see [14, 15])

$$k^*(T_e)=1.6\cdot10^{-9}\frac{(I^*(I^*+T_e))^{1/2}}{(I^*+0.13T_e)}\exp\left(-\frac{I^*}{T_e}\right),$$

$$k^+(T_e)=3.1\cdot10^{-7}\left(\frac{1}{I^+ + T_e}\right)^{1/2}\frac{T_e}{I^+ + 0.21T_e}\exp\left(-\frac{I^+}{T_e}\right),$$

$$k^*(T_e)=1.24\cdot10^{-6}T_e^{-1/2}\exp(-I^{*+}/T_e).$$

The recombination rate β^+ was estimated in $cm^6 \cdot c^{-1}$ by Pitaevskii's formula [16]

$$\beta^+(T_e)=\frac{4\sqrt{2}\,\pi^{3/2}e^{10}Z^2\Lambda}{9m^{1/2}(kT_e)^{9/2}}, \quad Z=1, \quad \Lambda=10.$$

The coefficient of the rate of de-excitation of an atom was determined by the principle of detailed balancing (in $cm^3 \cdot c$):

$$\beta^*(T_e)=\frac{k^*(T_e)}{S(T_e)};$$

the equilibrium constant was determined by Saha's formula:

$$S(T_e)=2\frac{g^+}{g}\left(\frac{2\pi mT_e}{h^2}\right)^{3/2}\exp\left(-\frac{I^*}{T_e}\right).$$

As $\sigma(\epsilon)$ the experimental values of [17] were used.

In the calculations a uniform mesh was used with $K = 20$ nodes along the spatial coordinate x with interval length $L = 80\mu$. The value of the first step $h_1 = 0.1\mu$. The succeeding steps were specified as a geometrical progression with denominator 1.3. With the aim of determining the accuracy of the numerical calculations the τ-mesh was halved. The difference in the solution appeared in the fourth decimal place.

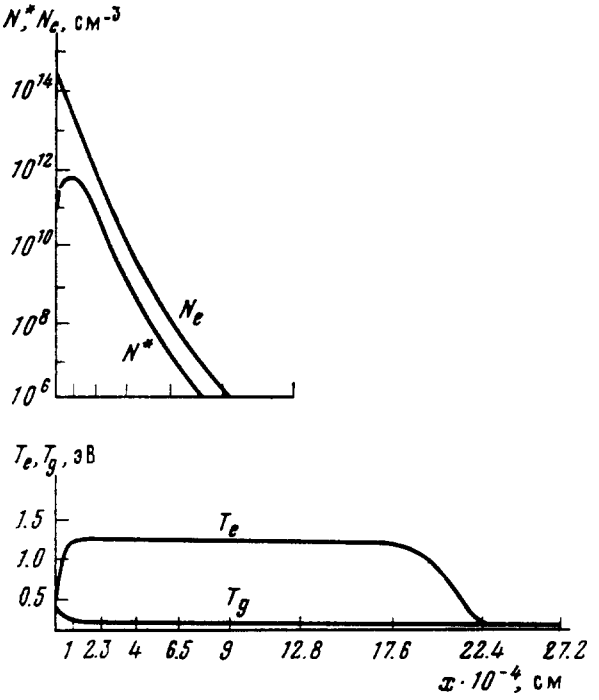
N*, Ne, см-3
10^14
10^12
10^10
10^8
10^6
Ne
N*
Te, Tg, эВ
1.5
1.0
0.5
Te
Tg
1 2.3 4 6.5 9 12.8 17.6 22.4 27.2
x · 10^-4, см

FIG. 1

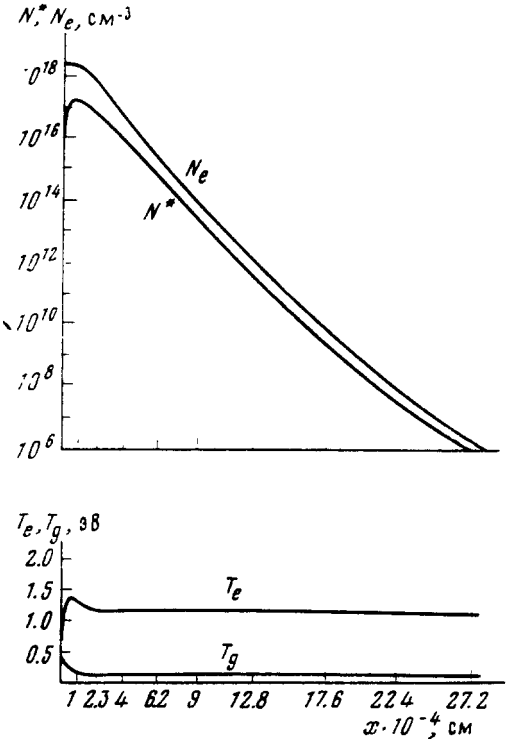
N*, Ne, см-3
10^18
10^16
10^14
10^12
10^10
10^8
10^6
Ne
N*
Te, Tg, эВ
2.0
1.5
1.0
0.5
Te
Tg
1 2.3 4 6.2 9 12.8 17.6 22.4 27.2
x · 10^-4, см

FIG. 2

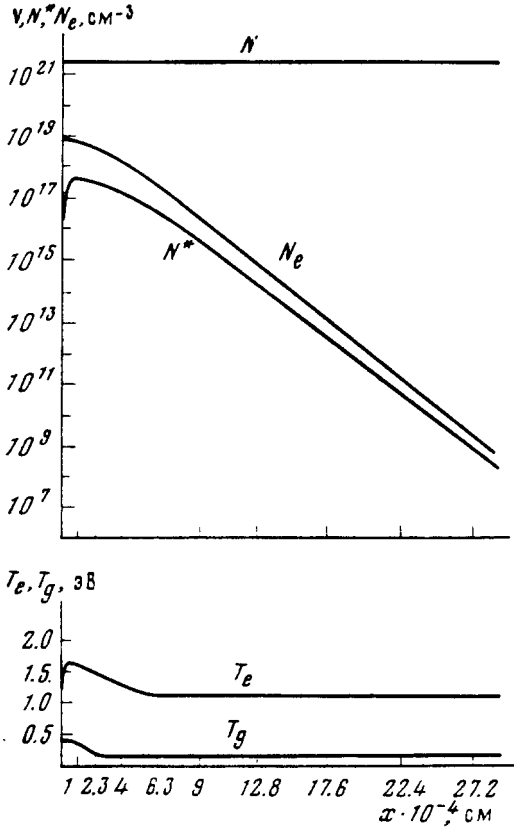

FIG. 3

Figures 1–3 show the spatial profiles of the densities N, N^*, N_e and temperatures T_e, T_g calculated for an initial pressure of 100 atm, which corresponds to $N = 2.7 \cdot 10^{21}$ cm^{-3}, and for the radiation flux density $G_0 = 4.10^6$ W·cm^{-2}. It is obvious from Figs. 1–3 that at a distance above 10μ there are few electrons, since the high pressure of the gas prevents the diffusion of the electrons and the main discharge processes develop at a distance of several microns from the target. The electron temperature for $x \geqslant 0.4\mu$ is very rapidly (within 2.10^{-10}s) established at 1.21 eV. In this time the gas is completely cold (Fig. 1, $t = 6.3 \cdot 10^{-10}$s). Then under the influence of thermionic emission and ionization the electron density at the point x=0.4μ increases to 10^{18} cm^{-3}. At this charged particle density the frequency of the Coulomb collisions becomes predominant, $\nu_{ei} > \nu_{en}$ and T_e increases sharply, which causes cumulative ionization and further strengthing of the inequality $\nu_{ei} > \nu_{en}$ (Fig. 2, $t = 3.65 \cdot 10^{-9}$s). At the instant $5.6 \cdot 10^{-9}$s the temperature T_e at the point x=0.4μ attains its maximum value $T_{e\,\max} = 1.64$ eV, for which $N_e \approx 2 \cdot 10^{19}$cm^{-3} (Fig. 3, $t = 5.6 \cdot 10^{-9}$s). Later the value of T_e at this point falls a little because of the decrease of the flux of laser radiation G_0 due to the growth of absorption in the adjacent region on the right, where cumulative ionization also begins. Because of the high electron density the temperature of the gas increases rapidly due to elastic collisions up to 0.35 eV. The initial stage of discharge of the cold gas may be regarded as ended by this, since a local region of intensive absorption of laser radiation is formed in space. Further calculations were not carried out, since the mathematical model (1)–(6) does not take into account the gas-dynamic motion of the hot gas, which may lead to an inadequate representation of the real picture of the processes taking place.

It must be mentioned that the assumptions made in the formulation of the problem somewhat simplify the real physical picture because of the molecularity of nitrogen, and also because of the infringement of the conditions of quasi-neutrality, due to thermionic emission of electrons. The numerical modelling of the process of laser discharge in a molecular gas will be the subject of further investigations.

The authors thank A. A. Samarskii and N. N. Rykalin, and also V. Ya. Gol'din and N. N. Kalitkin for useful discussions.

Translated by J. Berry.

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