Spherical stratification of a glow discharge

O. A. Nerushev, S. A. Novopashin, V. V. Radchenko, and G. I. Sukhinin Institute of Thermophysics, Novosibirsk 630090, Russia

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Spherically symmetrical strata (striation) in a dc glow discharge were discovered. Experiments were carried out in a low-pressure steel chamber that served as a cathode. An anode was placed in the center of the chamber. The stratification of the glow discharge was observed in some molecular gases: air, N₂, O₂, CO₂, and acetone vapor. A simple relation between strata radii and their number $r_n/r_{n-1} = \alpha$, where $\alpha = 1.4-2$ (depending on gas composition), were found. Gas pressure dependences of strata radii were measured. The values of bulk charge and electric potential radial distributions were estimated. A simplified Boltzmann equation for the electron energy distribution function (EEDF) in spherical geometry was used for theoretical consideration. The so-called "black wall law" assumption was used in the collisional integral. An analytical solution for the isotropic part of EEDF was obtained. The existence of converging and decaying stagnant waves reproducing the variation of the observed strata radii was shown. [S1063-651X(98)03709-X]

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I. INTRODUCTION

The appearance of alternating bright and dark regions in the positive column of a glow discharge in tubes is a wellknown phenomenon (striations). Nonmoving striations were first described in 1843 by Abria [1]. The stratified discharge is the widespread form of the positive column. This phenomenon may be observed in molecular or atomic gases, in the range of pressures from 10^0 to 10^4 Pa. A striation may be static or moving, with the velocities up to 10^5 cm/s. The nature of striations is ionization instability, and this phenomenon is called ionization wave. The main processes that define the characteristics of the striations are ionization by an electron impact and ambipolar diffusion to the walls of the discharge tube. Some data on striations in tubes are presented in reviews [2-7] and recent papers [8-10]. The authors are not familiar with experimental investigations or theoretical considerations of spherical strata in bulk discharges. This paper presents, to our knowledge, the first observation of spherical striations in a three-dimensional (3D) glow discharge and the first attempt at theoretical treatment of this phenomenon. In this paper, to distinguish between discharges in tubes and in 3D glow discharges the term "strata" will be used instead of the term "striation."

II. EXPERIMENTAL

Experimental setup is shown in Fig. 1. The experiments were carried out in a cylindrical vacuum chamber (60 cm in height and 50 cm diam) made of steel. It could be pumped up to 10^{-2} Pa and then filled with different gases. The pressure was controlled by a pressure gauge. The experiments were carried out in the range of pressures 5–50 Pa. A high-voltage insulated copper wire was placed radially in the middle of the chamber, so that its noninsulated end (1 cm in length) was in the center of the chamber. The diameter of the electrode was 0.3 cm. The steel chamber walls served as the second electrode. The dc power supply with a maximum voltage of 1 kV was used. The voltage and current of the discharge were measured. The maximum value (\approx 40 mA) of

the current was restricted by active resistance. As a rule the voltage drop on the discharge was about 400-500 V.

The vacuum chamber was supplied with four glass windows placed in the middle of the cylinder height. They were used for visual observation and for taking pictures of the glow discharge from different directions.

III. RESULTS

Several spherical concentric strata nested one into another were observed while the positive voltage was applied to the central electrode. The photograph of the strata is shown in Fig. 2 (the anode can also be recognized in the picture). The lighting of the strata has a surprisingly spherical symmetry. The typical size of the central lighting sphere is about 2 cm. Four strata can be seen in the picture. The number of strata depends on many parameters. They were observed from one to more than ten strata (and also a nonstratified discharge).

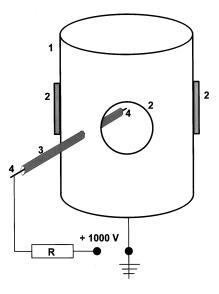


FIG. 1. Experimental setup. (1) vacuum chamber, (2) glass windows, (3) insulator, and (4) copper wire.

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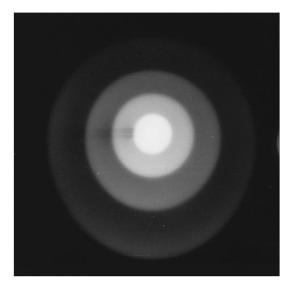


FIG. 2. Photograph of stratified discharge in mixture of air+acetone, P = 15 Pa.

The dimensions of the strata depend on the pressure and gas composition. Figure 3 shows the radius r_n of the stratum as a function of its number *n*. The radii are normalized on the radius of the central lighting ball r_1 . Starting with the second stratum, the dependence may be approximated as

$$r_n/r_1 = \alpha^n = \exp(\beta n), \tag{1}$$

The coefficient α is gas composition dependent and it was observed in the range from 1.4 to 2 ($\beta = 0.35 \div 0.7$). The absolute values of the strata radii increased with the decrease of pressure. These dependencies are close to inverse proportionality as it is shown in Fig. 4.

The spherical strata were observed in molecular gases (air, pure N₂, O₂, CO₂). The addition of some polyatomic gas (acetone, benzene) led to an increase of strata number and to a decrease of their radii [decreasing of the coefficient α in Eq. (1)]. The strata were not observed in Ar. The strata lifetimes depended on gas conditions and changed in the range from a few seconds to tens of minutes.

It should be stressed that the strata were not observed if the central electrode served as a cathode and the walls served as an anode. Under these conditions, the discharge voltage was much higher (approximately 1 kV) and the electric current was very low ~ 2 mA.

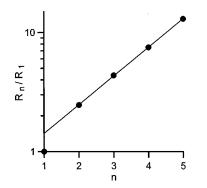


FIG. 3. Normalized strata radii vs its number.

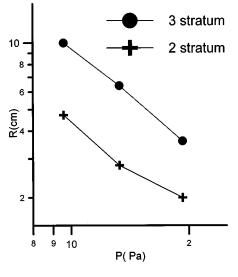


FIG. 4. Strata radii vs pressure.

IV. THEORY

The problem of a rigorous kinetic description of electrons and ions in spatially nonhomogeneous plasma in the presence of dc electric fields is extremely complicated. The solution of this problem should be self-consistent. For the calculation of the electron and ion distribution functions in nonhomogeneous plasma, it is necessary to know the electric field in plasma. However, the electric field has to be determined from the Poisson equation with the known distributions of charged particles. Even in a quasi-one-dimensional case of the positive column of discharges in tubes, this problem has not been solved until now. Usually, the assumption of a constant or slightly modulated longitudinal electric field along the discharged tube is used [11-13]. In a kinetic consideration of ionization waves (striations) in discharge tubes [11], a simplified spatially modulated electric field consistent with the experimental data has been used.

To treat the problem of three-dimensional spherical strata in this paper, we will use some assumptions on spatial distribution of electric field, which is consistent with our experimental data.

It is known [8–13] that voltage drops between the neighboring strata are the same in the positive column in discharge tubes. These drops are connected with electron energy losses in nonelastic collisions. Let us assume that a similar situation occurs in the 3D case. Taking into account the distance between the strata and voltage on discharge Φ measured experimentally, it is possible to estimate the bulk charge $\rho(r)$, and electric potential $\varphi(r_n)$. The potential for the *n*th stratum radius r_n can be presented in the form

$$\varphi(r_n) = \Phi - \varphi_0 n = \Phi - \varphi_0 \ln(r_n/r_1)/\beta, \qquad (2)$$

where r_1 is the radius of the stratum nearest the anode. The value of φ_0 depends on the kind of gas, its molecular energy levels, and cross sections of electron energy losses. Usually, φ_0 is equal to several electron volts and correlates with the first electron excitation level ε_1 (or the ionization potential of gas) [8–13].

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We suppose that the smoothed potential for every point between the anode and cathode sheaths can be presented by the same expression:

$$\varphi(r) = \Phi - \varphi_0 \ln(r) / \beta. \tag{3}$$

The electric field then will be

$$E(r) = -\partial \varphi(r) / \partial r = \varphi_0 / \beta r.$$
(4)

From the Poisson equation,

$$-\frac{1}{r^2}\frac{\partial}{\partial r}\left(r^2\frac{\partial\varphi}{\partial r}\right) = 4\pi\rho(r),\tag{5}$$

we can obtain a positive bulk (space) charge distribution,

$$\rho(r) = \varphi_0 / 4\pi \beta r^2. \tag{6}$$

Outside the electrode sheaths, the electron current density $j_e(r)$ is much higher than the ion current density $j_i(r)$. Hence, we can obtain that

$$j_e(r) = J/4\pi r^2, \tag{7}$$

where J is a discharge current. On the other hand, the electron current density can be presented as

$$j_e(r) = -en_e(r)\mu_e E(r), \qquad (8)$$

where μ_e is the mobility of electrons and -e is the charge of electron. It turns out that electron density is inversely related to r. This dependence seems to contradict the radial dependence of the bulk charge (6). However, using the values of electron mobilities μ_e (Ref. [7]) and the experimental values of electron current density, it can be shown that number densities of the bulk charge n(r), ions $n_i(r)$, and electrons $n_e(r)$ satisfy the following relation:

$$n(r) = n_i(r) - n_e(r) \ll n_e(r).$$

Let us consider the problem of spherical strata on the basis of Boltzmann equation, with some simplifications that are common, however, for treating the strata problem in onedimensional discharge tubes [8,10]. The traditional two-term approximation for electron distribution function will be used:

$$f(r,v) = \mathbf{f}_0(r,\varepsilon) + (\mathbf{v}/v)\mathbf{f}_1(r,\varepsilon)$$

where v is the vector of the mean electron velocity, $v = |\mathbf{v}|$, $\varepsilon = mv^2/2$, m is the mass of electron, $f_0(r,\varepsilon)$ is the isotropic part, and $(v/v)\mathbf{f}_1(r,\varepsilon)$ is the first anisotropic term in the expansion of the electron energy distribution function (EEDF) f(r,v) in Legendre polynomials. The functions $f_0(r,\varepsilon)$ and $f_1(r,\varepsilon)$ are related with the electron density n_e and electron current density j_e :

$$\int_{0}^{\infty} f_{0}(r,\varepsilon) \sqrt{\varepsilon} d\varepsilon = n_{e}(r), \qquad (9)$$

$$\frac{1}{3} \int_0^\infty f_1(r,\varepsilon)\varepsilon r^2 d\varepsilon = -\sqrt{m_e/2} \,\frac{j_e(r)}{e} \,r^2 \propto J. \tag{10}$$

For stationary conditions for the spherical case with the electric field (4), the Boltzmann equations for $f_0(r,\varepsilon)$ and $f_1(r,\varepsilon)$ can be presented in the form

$$\frac{1}{r^2} \frac{\partial}{\partial r} (r^2 f_1(r,\varepsilon)\varepsilon) - \frac{1}{3} \frac{e\varphi}{\beta r} \frac{\partial}{\partial \varepsilon} (f_1(r,\varepsilon)\varepsilon)$$
$$= N_g \Biggl(\sum_j \left[\sigma_j(\varepsilon + \varepsilon_j)(\varepsilon + \varepsilon_j) f_0(r,\varepsilon + \varepsilon_j) - \sigma_j(\varepsilon)\varepsilon f_0(r,\varepsilon) \right] + \frac{2m}{M} \frac{\partial}{\partial \varepsilon} \left[\sigma_m(\varepsilon)\varepsilon^2 f_0(r,\varepsilon) \right] \Biggr)$$
$$+ S_{\rm ion}(f_0(r,\varepsilon)) + S_{\rm rec}(n_i, f_0(r,\varepsilon)), \qquad (11)$$

$$\frac{\partial}{\partial r} f_0(r,\varepsilon) - \frac{e\varphi_0}{\beta r} \frac{\partial}{\partial \varepsilon} f_0(r,\varepsilon) = -N_g \left[f_1(r,\varepsilon)\varepsilon r^2 \left(\frac{\sigma_m(\varepsilon)}{\varepsilon} \right) \right] \frac{1}{r^2}, \qquad (12)$$

where $\sigma_m(\varepsilon)$ and $\sigma_i(\varepsilon)$ are momentum and excitation cross sections, N_{σ} is the gas number density, and M is the mass of gas molecule. The terms in the figure brackets in the right side of Eq. (11) corresponds to conservative inelastic and elastic collisions of electrons with gas molecules. The last two terms in Eq. (11), $S_{ion}(f_0(r,\varepsilon))$, $S_{rec}(n_i, f_0(r,\varepsilon))$, represent ionization and recombination collisional terms that are responsible for electron and ion production and losses. In stationary discharge conditions, these two terms must be equal and will be omitted from further consideration. This mutual ionization-recombination compensation might be nonlocal. A spatial variation of EEDF results in the separation of these processes in the radial direction within the limits of each stratum. Equations (11) and (12) could be and should be solved numerically for given electron-gas collisional cross sections. Energy dependencies of cross sections are rather complicated. Fortunately, it is well known that the cross sections of energy losses in molecular gases dramatically increase for energies higher than the threshold of the first electronic excitation level, ε_1 . The electrons energy distribution function, $f_0(r,\varepsilon)$, is exponentially decreasing in this energy region. Let us assume that $f_0(r,\varepsilon)$ is zero for ε $> \varepsilon_1$. This assumption is the so-called "black wall law" [8,9].

The purpose of this paper is to show a specific and periodical structure of EEDF (in energy and coordinate space) that explains the presence of spherical strata in 3D glow discharge. A peculiar energy dependence of EEFD is not important in this context. Thus, we can simplify the collisional terms in Eq. (11) to receive an exact analytical solution, which provides us with a clear physical understanding of spherical strata.

Let us also assume that the right side of Eq. (11) can be presented in the form of continuous energy losses for elastic collisions, rotational and vibrational excitations, and that $f_0(r,\varepsilon)$ is zero for energies higher than ε_1 —the first electronic excitation level of the molecule. where $L(\varepsilon)$ is the stopping cross section of electrons in a molecular gas:

$$L(\varepsilon) = \left[\sum_{j} \sigma_{j}(\varepsilon)\varepsilon_{j} + \frac{2m}{M} \sigma_{m}(\varepsilon)\varepsilon\right] = \sigma_{ex}(\varepsilon)\varepsilon_{ex}.$$

For example, below the excitation threshold of low-lying electronic states of N_2 at several eV, the total stopping cross section is extremely small except in the resonance region where the vibrational stopping cross section is remarkably large ($\sim 10^{-16}$ cm² eV for 2 eV < ε < 3 eV). For energies higher than the effective excitation threshold ($\varepsilon_1 \approx 7.5$ eV), the total stopping cross section is increasing abruptly from the value of $L(\varepsilon) \sim 2 \times 10^{-18}$ cm² eV up to 2 $\times 10^{-15}$ cm² eV at $\varepsilon \approx 11$ eV [14]. This fact can be the reason for the use of the "black wall" assumption often used in papers devoted to striation problems in discharge tubes [8,9].

The set of equations (12) and (13) can be solved analytically if we substitute the quantities

$$[\sigma_{\rm ex}(\varepsilon)\varepsilon_{\rm ex}\varepsilon], \ \ [\sigma_m(\varepsilon)/\varepsilon]$$

for their averaged values in the energy region $\varepsilon = (0 \div \varepsilon_1)$ and introduce an averaged dimensionless quantity:

$$\Lambda^2 = 3N_g^2 \sigma_m \sigma_{\rm ex} r_c^2 \frac{\varepsilon_{\rm ex}}{\varepsilon_1}, \qquad (14)$$

where r_c is the radius of the largest strata. Usually, r_c has the order of half distance to the cathode. According to the set of electron- N_2 cross sections [14], $\varepsilon_1 \approx 10 \text{ eV}$, $\langle \sigma_{\text{ex}}(\varepsilon) \varepsilon_{\text{ex}} \rangle \approx 1 \times 10^{-16} \text{ cm}^2 \text{ eV}$ for 2 eV $\langle \varepsilon \langle 3 \text{ eV} \rangle$. Thus, the averaged parameter $\Lambda \approx 1.5$ for $N_g \approx 3.5 \times 10^{15} \text{ cm}^{-3}$ (pressure p = 13 Pa, T = 300 K) and $r_c = 10 \text{ cm}$. It is obvious that Λ is proportional to the gas density. From two equations (12) and (13), we can obtain one equation for the function $h(r,\varepsilon) = rf_0(r,\varepsilon)$:

$$r^{2} \frac{\partial^{2} h}{\partial r^{2}} - 2 \phi r \frac{\partial^{2} h}{\partial r \partial \varepsilon} + \phi^{2} \frac{\partial^{2} h}{\partial \varepsilon^{2}} + (\phi + \Lambda^{2} r^{2}) \frac{\partial h}{\partial \varepsilon} = 0.$$
(15)

Here, the energy ε and radius *r* are normalized by ε_1 and r_c and the parameter $\phi = e \varphi_0 / \beta \varepsilon_1$ has to be defined from a comparison with the experiments.

We must solve this equation backward from $r = r_c$ using boundary conditions for distribution functions $f_0(r,\varepsilon)$ and $f_1(r,\varepsilon)$ at the edge of the cathode sheath. It can be seen from Eq. (15) that it is possible to introduce new variables: $x = \Lambda r/\phi^{1/2}$ and $t = \varepsilon/\phi$. Then, Eq. (15) changes to the universal form

$$x^{2} \frac{\partial^{2} h}{\partial x^{2}} - 2x \frac{\partial^{2} h}{\partial x \partial t} + \frac{\partial^{2} h}{\partial t^{2}} + (1 + x^{2}) \frac{\partial h}{\partial t} = 0, \quad (15')$$

and must be solved with appropriate initial conditions.

Equation (15) can be solved using Fourier analyses with the help of substitution,

$$h(r,\varepsilon) = \sum_{m=-\infty}^{\infty} \exp\{2\pi i m(\varepsilon + \phi \ln r)\} H_m(r).$$
(16)

The function $H_m(r)$ is satisfied by the equation

$$\frac{\partial^2 H_m}{\partial r^2} + 2\pi i m \Lambda^2 H_m(r) = 0, \qquad (17)$$

which has the solutions

$$H_m(r) = C^{\pm}_m \exp\{\pm \sqrt{\pi m}\Lambda r(1-i)\}.$$
 (18)

A general solution of Eq. (15) has the form

$$h(r,\varepsilon) = \sum_{m=-\infty}^{\infty} C^{\pm}_{m} \exp\{2\pi i m (\varepsilon + \phi \ln(r)) + \sqrt{\pi m} \Lambda r (1-i)\}.$$
(19)

The complex constants C^{\pm}_{m} have to be determined from boundary conditions at $r=r_c$. The solution $h(r,\varepsilon)$ must be real. It turns out that $C^{\pm}_{m}=(C^{\pm}_{m})^*$. A solution with the superscript "+" must be used for boundary conditions at the external radius r_c (on the cathode sheath). This solution leads to energy dissipation. On the other hand, the second solution (labeled as "-") leads to energy gain even in the absence of electric field ($\phi=0$).

Thus, after simple mathematics, the solution can be presented in the form

$$f_{0}(r,\varepsilon) = \frac{h(r,\varepsilon)}{r} = \frac{r_{c}}{r} \sum_{k} \left\{ A_{k} \cos \left[2 \pi k \left(\varepsilon + \phi \ln \left(\frac{r}{r_{c}} \right) \right) + \sqrt{\pi k} \Lambda \frac{(r_{c} - r)}{r_{c}} \right] + B_{k} \sin \left[2 \pi k \left(\varepsilon + \phi \ln \left(\frac{r}{r_{c}} \right) \right) + \sqrt{\pi k} \Lambda \frac{(r_{c} - r)}{r_{c}} \right] \right\} \exp \left(- \sqrt{\pi k} \Lambda \frac{(r_{c} - r)}{r_{c}} \right).$$

$$(20)$$

The coefficients A_k , B_k can be obtained from Fourier expansion of the distribution function $h(r=r_c,\varepsilon)$:

$$h(r=r_c,\varepsilon) = \sum_k \{A_k \cos[2\pi k\varepsilon] + B_k \sin[2\pi k\varepsilon]\}.$$
(21)

It can be seen that solution (20) presents a converging and decaying stagnant wave. For the parameter $\Lambda \ll \pi$, the decay is small and the electron distribution function $h(r,\varepsilon) = rf_0(r,\varepsilon)$ is reproduced according to the variation of observed strata radii (1). This gives us some number of strata. For $\Lambda > \pi$, there are no spatial oscillations in distribution function due to damping connected with the energy losses in vibrational excitation of molecular gas.

Figure 5 shows an example of the spatial-energy distribution function $h(r,\varepsilon)$ for $\Lambda = 0.5$. The initial distribution function at $r = r_c$ is chosen in form (21) with the coefficients $A_k = (-1)^k a^{-1} \{1 + (k/a)^2\}^{-1}$, a = 1, and $B_k = 0$ (*k*

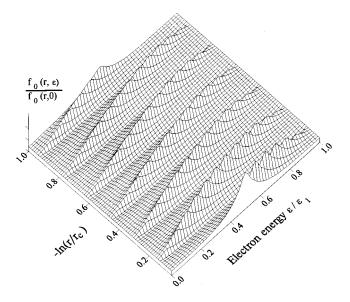


FIG. 5. Electron energy distribution function $h(r,\varepsilon) = rf_0(r,\varepsilon)$ obtained according to Eq. (20) with boundary conditions $A_k = (-1/a)^k \{1 + (k/a)^2\}^{-1}$, $B_k = 0$, $\Lambda = 0.5$, a = 1.

=0-50). It can be seen that while *r* is decreasing, electrons gain energy in the electric field, reach the "black wall" at $\varepsilon = \varepsilon_1$, lose energy, and appear with small energy. Then, the cycle is repeated.

Figure 6 shows the spatial variation of electron density n_e (r) for $\Lambda = 0.5$ (this value corresponds to pressure $P \sim 10$ Pa). Due to linearity of solved equations (12) and (13) n_e is proportional to the current density. It can be seen that the electron density has spatial oscillations in accordance with the experimental data.

From Eq. (15) and its dimensionless form (15'), it follows that the distance between strata will be inversely proportional to Λ (or to gas density N_g) for the region where $\Lambda r/r_c \leq 1$ in full accordance with the experimental data.

V. DISCUSSION AND CONCLUSION

In this paper, spherical bright and dark alternating regions in a 3D glow discharge in molecular gases have been reported to our knowledge, for the first time. The radii dependencies on their number and gas pressure have been measured. For the explanation of the observed phenomena, a

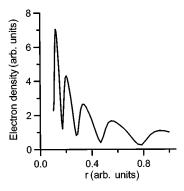


FIG. 6. Radial distribution of electron density n_e , corresponding to boundary conditions with $A_k = (-1/a)^k \{1 + (k/a)^2\}^{-1}$, $B_k = 0$, $\Lambda = 0.5$, a = 10.

simplified model has been proposed. It was assumed that the spherical strata are of the same nature as the striations in discharge tubes. It has been possible for us to deduce the electric potential from the experimentally observed radii of strata. An analytical solution of a simplified Boltzmann equation for the deduced electric potential has been received. The solutions show the existence of spherical strata with radii inversely proportional to gas pressure that is in agreement with the experimental data.

The analysis of strata radii has resulted in the conclusion that a bulk charge plays an important role in spatial potential distribution. This may be the reason why the stratification has been observed only in molecular gases. The main processes that define the bulk charge are the ionization, recombination, and diffusion of charged particles. The recombination processes of noble and molecular gases are known to differ significantly. The recombination in a noble gas discharge is suppressed because it needs triple collisions. The principal process that leads to electron and ion losses in discharge tubes for atomic gases is connected with the diffusion of charged particles to the tube wall and their recombination there. In the case of molecular gas discharges, dissociative recombination plays an important role in two-body electronmolecular ions collisions. With respect to strata formation, this process should be studied both theoretically and experimentally.

The finite "life" duration of strata is an interesting property of the spherical discharge. We assume that the processes of dissociation (because of molecular gases), plasma chemical reactions, and excitation of metastable molecular states may result in a change of gas composition, spatial distribution of volume charge, and electric potential and lead to the disappearance of stratification.

In the model, the assumption of a "black wall" for the electron distribution function $[f_0(r,\varepsilon)$ is zero for $\varepsilon > \varepsilon_1]$ has been used. The smoothed logarithmic electric potential (3) consistent with the observed strata radii (1) in the region between the anode and cathode sheaths has been adopted. The obtained solution shows the existence of strata for the averaged energy losses parameter $\Lambda \leq 1$ [see Eq. (14)] for low-energy electrons $\varepsilon < \varepsilon_1$. For atomic gases, the corresponding averaged parameter Λ must be much lower than for molecular gases. It seems that spherical strata could exist at the same gas pressures and chamber radius. However, there have been no strata observed in argon and helium discharges (for a given range of pressures). The only explanation for this fact is that the electric potential between the anode and cathode in atomic gas has a different spatial dependence for the reason discussed above. It should be noted that the model may be improved using a real potential distribution and a real set of cross sections of elastic and nonelastic collisions. However, a rigorous approach requires a self-consistent consideration including the solution of nonlocal Boltzmann equation for electron energy distribution function, equation for ions motion, and Poison equation for an electric potential.

It should be noted that some analogy can be drawn between the observed spherical strata and the double layers that were observed in the 3D glow discharge in argon at low pressures of order $P \sim 0.2-1$ Pa [15]. However, in the latter case the glow discharge was observed in a gas flow with substantial gradients of gas concentration due to a fast expansion of gas from the anode region.

The spherical gas discharge considered in this paper, i.e., the discharge with a small central anode and remote cathode, has some characteristics that make it somewhat similar to a positive corona discharge, at least, in the region around the central electrode. In the anode sheath, the value of the electric field must be very large. This small region serves as a source of ions. For infinitely remote walls, the spherical discharge tubes into a low-pressure corona [16,17]. In such a corona, the ion discharge current density would be several orders smaller than the total current density in our case. For a large but finite radius of the chamber, there is a cathode sheath adjusted to the chamber wall. A strong electric-field region serves as a source of electrons that drive to the central electrode. As a result, in the region between the anode and cathode sheaths, the discharge has the features of a glow discharge. Thus, we believe that the spherical discharge described in the paper is some mixture of corona (in the nearanode region) and glow discharges.

ACKNOWLEDGMENT

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