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THE ELECTRONEGATIVE GLOW DISCHARGE IN THE CYLINDRICAL AND COAXIAL GEOMETRY: THE COMPARISON OF OPTICAL RADIATION EMISSION ABILITY

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The transition from the traditional cylindrical discharge geometry to the coaxial one (where the discharge plasma is located in a gap between two coaxially placed cylindrical tubes) has been theoretically investigated for the positive column of an electronegative middle pressure glow discharge in the mixture of chlorine and inert gases. Here a new electron's loss mechanism appears, i.e. the electron's diffusional outgoing onto the inner wall. The discharge existence was proved to be only made possible by sufficient increasing of the ionization frequency and hence the electron temperature as well. The elevation of the electron temperature would cause a growth of the specific power of discharge ultraviolet radiation. The analytical expressions for estimating the electron temperature of the discharge plasma in the coaxial geometry were derived.

Keywords: plasma radiation, electronegative discharge, electron temperature, ionization frequency, coaxial geometry

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СРАВНЕНИЕ СПОСОБНОСТИ ИСПУСКАНИЯ ОПТИЧЕСКОГО ИЗЛУЧЕНИЯ ЭЛЕКТРООТРИЦАТЕЛЬНОГО ТЛЕЮЩЕГО РАЗРЯДА В ЦИЛИНДРИЧЕСКОЙ И КОАКСИАЛЬНОЙ ГЕОМЕТРИИ

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Для положительного столба электроотрицательного тлеющего разряда среднего давления в смеси хлора с инертным газом теоретически исследован переход от традиционной цилиндрической геометрии разряда к коаксиальной, когда плазма разряда локализована в зазоре между двумя коаксиально расположенными цилиндрическими трубками; при этом возникает новый механизм потерь электронов – их диффузионный уход на внутреннюю стенку. Показано, что существование разряда становится возможным лишь при существенном увеличении частоты ионизации и, соответственно, электронной температуры; последнее вызовет возрастание удельной мощности ультрафиолетового излучения разряда. Получены аналитические выражения для оценки электронной температуры плазмы указанного разряда в коаксиальной геометрии.

Ключевые слова: излучение плазмы, электроотрицательный разряд, электронная температура, частота ионизации, коаксиальная геометрия

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Introduction

Continuing the terminology adopted in [1], we understand the coaxial geometry of the discharge as the configuration where the discharge is initiated in the gap between two coaxial cylindrical tubes, plasma has a tubular shape, and the longitudinal field E_z and the discharge current are directed along the axis of the tubes. Such electronegative discharges in mixtures of inert gases with chlorine are used as working media in powerful and effective mercury-free sources of ultraviolet radiation [2, 3]. The emitting particles were inert gas chlorides that are excimers.

We have found previously [1] that the ionization rate is increased for a positive column of electronegative glow discharge of moderate pressure (up to 40 Torr) in coaxial geometry, compared to cylindrical geometry, even with a small (0.05–0.10) ratio of the radii of the inner to the outer wall of the discharge tube (R_1/R_2). Therefore, it seems logical to assume that the population rates of excited states of gas atoms and the specific radiation power of the discharge increase as well. However, while this issue is important for practical applications of gas-discharge light sources, it is currently poorly understood.

The goal of this study has consisted in proving theoretically that transition from cylindrical to coaxial geometry of this type of discharge increases the electron temperature T_e, and, accordingly, the concentration of excimers in the positive column. An additional task has been to obtain quantitative estimates for the increase in T_e and the specific power of UV excimer radiation.

Description of the model

It was established in [4, 5] that positive ions in plasma of halogen discharges are mostly molecular (Cl^+_2 for chlorine), and the dominant mechanism of decay of negative ions Cl^- is dissociative ion-ion recombination:

$$Cl_2^+ + Cl^- \rightarrow 3Cl.$$

Notations. Let us introduce the following notations: ν_i, ν_a are ionization and attachment rates; ρ_i is the ion-ion recombination coefficient; T_j, μ_j and n_j are the temperature, mobility and absolute density of the jth type of charged particles; indices $j = e$, p, n correspond to electrons, positive and negative ions; D^*_{ap}, D^*_{an} are reduced ion diffusion rates; n_{e0} is the maximum electron density;

$$D^*_{ap} = \mu_p T_e / \left(eR_2^2\right);$$
$$D^*_{ap} = D^*_{ap} / \mu_{pn};$$
$$\mu_{pn} = \mu_p / \mu_n; \mu_{np} = \mu_n / \mu_p; \quad (1)$$
$$\nu = \nu_i / D^*_{ap}; \alpha = \nu_a / D^*_{an};$$
$$\eta = n_{e0}\rho_i / D^*_{ap};$$

ν, α, η here are the dimensionless normalized rates of ionization, attachment, and ion-ion recombination, respectively.

Next, X is the transverse normalized (dimensionless) radial coordinate, X_1 is the normalized coordinate of the inner wall;

$$X = r/R_2;\ X_1 = R_1/R_2;\ X \le X \le 1;$$

$n(X)$, $N(X)$, $P(X)$ are relative spatial distributions of electrons, negative and positive ions along the X coordinate (transverse to the current direction); τ_p τ_n are the temperature ratios of ions and electrons;

$$n(X) = n_e(X) / n_{e0};$$
$$N(X) = n_n(X) / n_{e0};$$
$$P(X) = n_p(X) / n_{e0}; \quad (2)$$
$$\tau_j = T_j / T_e; \tau_p \approx \tau_n \approx \tau.$$

The same as [1, 6], the model accepted assumes quasineutrality for

$$P(X) = n(X) + N(X), \quad (3)$$

as well as constant ν_i, ν_a, ρ_i, μ_j along the plasma cross-section, and non-isothermal plasma, i.e., $T_e >> T_p$, T_n. However, the τ_p, τ_n values are not taken to be negligibly small.

Mathematical description. The following equations for the variables N, P and n were obtained in [1, 6]:

$$-\Delta N \cdot \tau_n + \nabla\left(\frac{N}{n}\nabla n\right) \approx \alpha n - \mu_{pn}\eta NP, \quad (4)$$

$$-\Delta P \cdot \tau_p - \nabla\left(\frac{P}{n}\nabla n\right) \approx \nu n - \eta NP \quad (5)$$

with zero boundary conditions on the walls for any type of charged particles. Derivatives in Eqs. (4) and (5) are taken for the normalized coordinate X.

It is known [6–8] that plasma in electronegative discharges stratifies in the direction transverse to the current flow into the central region of ion-ion (i-i) plasma (referred to as the “core” from now on) with relatively low electron density and the peripheral region of electron-ion (*e-i*) plasma (the "shell"), which is nearly free of negative ions, with plasma diffusion close to ambipolar. We shall confine our attention to the case of strong electronegativity, when $\alpha >> 1$, and

$$N \approx P >> n \sim 1, \quad (6)$$

in the core, while

$$P \approx n >> N, \quad (7)$$

in the shell, which is thin and can be treated as flat even for cylindrical discharge geometry [7, 8].

Summing Eqs. (4) and (5), we obtain, in view of conditions (6), the following relation for the core:

$$\Delta N \cdot \tau_S \approx \left(\mu_{pn}+1\right)\eta N^2 - \left(\nu+\alpha\right), \quad (8)$$

where $\tau_S = \tau_p + \tau_n$.

Taking into account inequality (7), valid for the shell, where there are few negative ions and a strong transverse field E_x [7], allowing to neglect ion diffusion, we obtain:

$$\Delta n = -(\nu+\alpha)n \quad (9)$$

We considered Eq. (8) in [1], where it was proved that if the inequality

$$0.01 \le \tau < 1.0 \quad (10)$$

holds true, then, provided that

$$0.3 \le R_1/R_2 < 1.0, \quad (11)$$

solution (8) for coaxial geometry is close to symmetric with respect to a point with the coordinate $(X_1+1)/2$. Such symmetry makes it possible to move the origin of the X coordinate to this point and write the solution as

$$N\left(X\right) \approx \frac{\nu+\alpha}{2\tau_S} X_0^2 \left[1-\left(\frac{X}{X_0}\right)^2\right] \quad (12)$$

and carry out further calculations for coaxial geometry as for plane geometry (let us call this the PG approximation). Here X_0 is the coordinate measured from the new origin, with N(X) taken equal to zero, i.e., X_0 is the normalized coordinate of the boundary between the *i-i* and *e-i* plasma. The actual value of X_0 is still unknown and remains to be calculated.

The normalized coordinate of the outer wall (equal to unity before the coordinate shift) equals, in PG approximation,

$$\sigma = \frac{R_2 - R_1}{2R_2} < \frac{1}{2},$$

and the X coordinate lies between 0 and σ.

Due to lack of data obtained in any real experiments, result (12) was verified to be valid by comparing it with the result of a computational experiment (CE) [1, 6] that is a model numerically solving a system of steady-state equations for flows of charged plasma particles and their densities in a positive column of electronegative glow discharge as an eigenvalue problem with a minimum of a priori assumptions. Using this approach, we were able to calculate the spatial profiles of electron and ion densities, the rates of plasma-chemical processes and to verify the obtained analytical expressions and find their range of applicability. As a result, we found that using expression (12) for coaxial geometry yields errors of $N(X)$ and $n(X)$ no more than 12%, provided that inequalities (10) and (11) are satisfied, and no more than 6% for $0.5 \le R_1/R_2 < 1.0$.

For non-isothermal plasma, $\tau < 1$, so negative ions do not fall on the walls and decay only in the plasma volume. Therefore, the average rates of production and decay of negative ions over its cross section should be equal, i.e.,

$$\alpha \int_0^{\sigma} n\left(X\right)dX = \mu_{pn}\eta \int_0^{X_0} N^2\left(X\right)dX. \quad (13)$$

We are going to use relation (13) to calculate the value of X_0; however, the function $n(X)$ should be known to take the integral on the left-hand side. It is taken for the core (see inequality (6)) that $n(X) \approx 1$; it was argued in [6] based on the results of CE that the dependence of $n(X)$ distribution is close to linear with $\tau \ge 0.01$.

Linear dependence $n(X)$

Let us theoretically confirm that the dependence $n(X)$ is linear.

According to expression (9), the following equality holds true in the shell for the PG approximation:

$$n\left(X\right) = n_m \sin\left[\sqrt{\nu+\alpha}\left(\sigma - X\right)\right]; \quad (14)$$

from this we obtain

$$n'\left(\sigma\right) = -n_m\sqrt{\nu+\alpha}.$$

On the other hand, according to Eq. (5) and condition (7),

$$-\Delta n\left(1+\tau\right) \approx \nu n$$

in the shell, or (for the PG approximation)

$$n'\left(\sigma\right) = -\frac{\nu}{1+\tau}\int_0^{\sigma} n\left(X\right)dX.$$

The shell is thin in case of strong electronegativity, the value of the function is $n(X) \approx 1$ in the core; therefore, the core makes the main contribution to the integral. Then n′(σ) can be estimated as

$$n'(\sigma) \approx -\frac{\nu\sigma}{1+\tau}. \quad (15)$$

Equating both expressions for n′(σ), we obtain the following relation:

$$n_m \approx \frac{\nu\sigma}{(1+\tau)\sqrt{\nu+\alpha}}. \quad (16)$$

Results of calculations by Eqs. (15) and (16) compared to the CE data are given in Table 1. An example is given for $\alpha = 8$ and for plane geometry, i.e., for $\sigma = 1$.

The data in Table 1 confirm that expression (15) ensures that the value of n′(1) agrees with the computational experiment with an error of no more than 22%. However, the main result here is that the values of n_m are considerably greater than unity and increase with increasing t. This does not contradict Eq. (9), but $n(X) \le 1$ by definition. Consequently, the $n(X)$ dependence in the shell should correspond only to a small initial segment of sinusoidal dependence (14), i.e., $n(X)$ near the wall should be close to linear, namely (in PG approximation):

$$n(X) \approx \frac{\nu\sigma}{1+\tau}(\sigma - X).$$

From a physical standpoint, $n(X)$ is linear in the shell at $\tau > 0$ due to diffusive penetration of negative ions from (*i-i*) plasma into the shell against the transverse field of the shell

$$E_x \infty \frac{\nabla n_e}{n_e},$$

which is weak near the peak of sinusoidal dependence (14). Firstly, such penetration increases the size of the core while reducing the shell size, and secondly, negative ions neutralize the field E_x inhibiting the escape of electrons from the region that they manage to penetrate. Electrons from this region quickly escape to the remaining part of the shell, with their diffusion close to free rather than ambipolar.

As a result, it can be assumed for coaxial geometry (in the PG approximation) that the approximate equality

$$n(X) \approx (\sigma - X)/(\sigma - X_0). \quad (17)$$

holds true in the shell. If we substitute expressions (10) and (17) into equality (13), then, after a series of transformations, we can obtain the following dependence:

Table 1

Calculated n′(1) and n_m as functions of normalized ionization rate and relative ion temperature

ν	τ	$n'(1)$		n_m
CE		CE	By formula (15)	By formula (16)
24.9	0.01	–24	–24.7	4.3
55.2	0.05	–47	–52.6	6.6
92.7	0.10	–107	–84.0	8.4
167	0.20	-177	-140	10.5

Notations: ν is the normalized ionization rate, obtained in the computational experiment (CE) for different relative ion temperatures τ, n′(1) is the derivative of electron density profile on the wall, n_m is the amplitude of sinusoidal dependence (14).

Note. The calculated data are given for the case $\alpha = 8$ and for plane geometry, i.e., for $\sigma = 1$.

$$X_0 \approx \left[\frac{15\alpha a}{2\mu_{pn}\eta} \left(\frac{\tau_S}{\nu+\alpha} \right)^2 \right]^{1/5}, \qquad (18)$$

where $a = (\sigma + X_0)/2$.

We can estimate X_0 from expression (18), including it in an implicit form. If $a \geq 0.7\sigma$, i.e., $0.4\sigma \leq X_0$, then X_0 can be estimated with an error no more than 5–10% using a simpler and more explicit formula:

$$X_0 \approx \left[\frac{15\alpha\sigma}{2\mu_{pn}\eta} \left(\frac{\tau_S}{\nu+\alpha} \right)^2 \right]^{1/5}. \qquad (19)$$

Positive ions actually flow from the shell to the wall in ambipolar mode [7, 8] (since $n_n << n_e \approx n_p$ in the shell):

$$\Gamma_p = -D_{ap} \frac{dn_p}{dx}.$$

Since in case of strong electronegativity,

$$n_p \approx n_n >> n_e \text{ and } n_e \approx n_{e0}$$

in the core, (see condition (6)), while $n_p \approx n_e$ in the shell, then, in accordance with expression (17),

$$\frac{dn_p}{dx} \approx -\frac{n_{e0}}{(\sigma R_2 - x_0)},$$

where x_0 is the absolute coordinate of the boundary between *i-i* and *e-i* plasma.

A flux Γ_p is formed in the plasma core; with x_0 it corresponds to the expression

$$\Gamma_p = \nu_i \int_0^{x_0} n_e(x)dx - \rho_i \int_0^{x_0} n_p(x) n_n(x) dx.$$

If we equate the above expressions for Γ_p with x_0 taking into account equality (13), we obtain the following expression (for the PG approximation):

$$\nu - \frac{\alpha}{2\mu_{pn}} \cdot \frac{\sigma + X_0}{X_0} \approx \frac{1}{X_0(\sigma - X_0)}. \qquad (20)$$

The value of ν can be calculated from this expression by substituting X_0 from Eq. (19). Notably, the discrepancy between the CE data and the values of ν calculated by Eqs. (20) and (19) does not exceed 18% (Table 2). If ν and α are expressed as functions of T_e, the latter can also be calculated by regarding expression (20) as a transcendental equation for T_e.

Thus, for an electronegative glow discharge in coaxial geometry, expression (20) is an equivalent of the Schottky formula used to estimate T_e in plasma of an electropositive glow discharge in cylindrical geometry [9].

Example calculation

Let us consider an electronegative glow gas discharge in a mixture of 6 Torr Xe and 0.25 Torr Cl_2 (inlet) at a current of 10 mA. The radius of the outer cylindrical tube is taken equal to R2= 6 mm, the gas temperature T_g = 500 K; Xe concentration with account for thermal displacement is equal to

$$N_{\text{Xe}} = 1.3 \cdot 10^{17} \text{ cm}^{-3},$$

and Cl concentration with account for dissociation of chlorine molecules by electron impact [8] is

$$N_{\text{Cl2}} = 1.75 \cdot 10^{14} \text{ cm}^{-3}.$$

Approximations of the ionization rate

$$\nu_i \cong N_{\text{Xe}} \cdot 9.2 \cdot 10^{-8} \exp(-12.9/T_e) 1/\text{s}$$

and attachment rate

$$\nu_a \cong N_{\text{Cl}_2} 3.69 \cdot 10^{-10} \times$$
$$\times \exp\left(-1.68/T_e + 1.457/T_e^2 - 0.44/T_e^3\right) 1/\text{s}$$

(T_e is measured in eV, concentrations in cm^{-3}) were taken from [10]. We take

$$\tau_p = \tau_n = \tau = 0.05.$$

We should note that using the concept of electron temperature T_e, which is equivalent to assuming that the electron energy distribution function (EEDF) is Maxwellian, is a simplified idealization for plasma discharges in mixtures of inert gases with chlorine. In fact, if T_e is defined as a parameter of Maxwellian EEDF adequately describing the excitation of higher levels of an inert gas atom and its ionization by electron impact (the so-called “fast electron temperature” [11]), then the model we described in [8] yields the value $T_e \approx 1.2$ eV for the above discharge in cylindrical geometry. However, if the values of D_e and μ_e (determined by slow electrons) are calculated within the same model for the same discharge conditions using the EEDF derived from the kinetic equation, and the electron temperature is then calculated using the Einstein relation as $T_e = eD_e/\mu_e$, the value of this temperature T_e is more than 6 eV.

To account for difference in the

“temperatures” of fast and slow electrons in further calculations, the quantities D^*_{ap} and D^*_{an}, included in the expressions for ν and α, were calculated as

$$D^*_{ap} = \mu_p \beta T_e / \left(eR_2^2\right),$$

$$D^*_{an} = \mu_n \beta T_e / \left(eR_2^2\right),$$

and β = 6 was selected to ensure the best agreement between the calculation results and the data of the computational experiment. The ion-ion recombination coefficient is taken equal to

$$\rho_i \approx 2.9 \cdot 10^{-9} \sqrt{T_g},$$

in accordance with the data given in [12, 13].

The calculation results are given in Table 2 for cylindrical and coaxial geometry with different ratios of the radii of the inner to the outer walls R_1/R_2, with a constant radius of the outer wall R_2. The “temperature of fast electrons” is given as T_e. S_{rel} refers to the relative (with respect to cylindrical geometry where $R_1/R_2 = 0$) cross-sectional area of the plasma in coaxial geometry.

The concentration of excimer molecules N_{XeCl^*} (the specific power of UV radiation of discharge plasma is proportional to this concentration) was calculated by the relation [14]:

$$N_{\text{XeCl}^*} \infty \left\langle \sigma_{0M} \nu_e \right\rangle,$$

where

$$\left\langle \sigma_{0M} \nu_e \right\rangle = 4.26 \cdot 10^{-9} \exp\left(-7.986 / T_e\right) + $$
$$+ 1.36 \cdot 10^{-8} \exp\left(-9.753 / T_e\right),$$

and σ_{0M} is the total cross-section for excitation of metastable and resonant Xe levels by electron impact (taken from [15]).

Discussion

The main physical cause for the increase in T_e and the specific power of UV radiation of the discharge upon transition from cylindrical to coaxial geometry (with a constant R_2) is the qualitative change in discharge conditions: an additional channel of electron loss appears, that is, their diffusion to the inner wall. Increased ionization rate ν_i is required to compensate for this loss, i.e., to maintain the discharge in coaxial geometry, compared with the case of cylindrical geometry. The increase in T_e with this said transition (see Table2) is attributed to increasing ν_i. As the gap between the walls narrows further in coaxial geometry, i.e.,

Table 2

Calculated T_e and N_{XeCl^*} for coaxial and cylindrical glow discharges in gas mixture of Xe and Cl_2 depending on ratio of inner to outer radii of discharge tube

R_1/R_2	S_{rel}	T_e, eV (calculated from (20))	ν		α	N_{XeCl^*}, c.u.
			CE	from calculated T_e		
0.00	1.00	1.20	120	99.2	8.00	1.0
0.15	0.98	1.31	231	217	7.48	1.8
0.28	0.92	1.36	330	304	7.24	2.3
0.50	0.75	1.48	–	615	6.75	4.0
0.64	0.59	1.62	1142	1174	6.31	6.6
0.80	0.35	1.94	–	3672	5.51	16.7

Notations: ν is the normalized ionization rate, T_e is the electron temperature, α is the normalized attachment rate, S_{rel} is the relative cross-sectional area of the plasma in coaxial geometry (compared to cylindrical geometry), N_{XeCl^*} is the concentration of excimer molecules proportional to the specific power of UV radiation of the plasma.

Note. The first row of the table refers to the case of cylindrical geometry of the glow discharge.

with $R_1/R_2 \rightarrow 1$, electron losses increase even more, necessitating a further increase in both ν_i and, accordingly, T_e (as follows from the data in Table 2). The increase in T_e, in turn, leads to increased excitation rates of Xe energy levels, which, with chlorine present in the gas mixture, provides increased concentration of excimer molecules and specific capacity of the UV radiation emitted by the plasma.

Conclusion

We have theoretically studied the transition from traditional cylindrical geometry of electronegative glow discharge in coaxial geometry at moderate pressure, with the discharge plasma located in the gap between two coaxial cylindrical tubes. We have quantitatively analyzed an additional mechanism of electron loss that is their diffusive escape on the inner wall and its effect on electron temperature and the specific power of UV radiation of discharge plasma, using the example of a discharge in a mixture of xenon and chlorine. We have confirmed that these parameters of discharge plasma increase considerably upon transition from cylindrical to coaxial geometry. The result obtained can be effectively used for designing and operating gas-discharge sources of light and UV radiation.

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