RADIAL DISTRIBUTION OF PLASMA PARAMETERS IN THE POSITIVE COLUMN OF A GLOW DISCHARGE IN LOW-PRESSURE IODINE VAPOR

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The radial distribution of the plasma potential and charged particles was measured in the positive column of a discharge in the low-pressure iodine vapor. The experimental data are qualitatively explained. The results obtained in this work are consistent with the plots of plasma potential and charged particle density against the radial coordinates obtained by J. B. Thompson.

1. Electronegative substances in the ionized state and their mixtures with other gases are widely used to solve a number of applied problems of plasma chemistry and to construct gas-discharge lasers and sources of negative-ion beams [1–4; 5, Chap. 15]. Information on the properties of such a plasma is also of great academic interest since elementary and collective processes that occur in them are known to possess a number of specific features as compared to processes occurring in a plasma that contains no negative ions.

If the ratio of the negative ion concentration $n_-$ to the electron density $n_e$ becomes much greater than unity, we then have what is called an ion-ion plasma. Its properties differ strongly both qualitatively and quantitatively from the characteristics of an electron-ion plasma in which $n_-/n_e \ll 1$. Under conditions of $n_-/n_e \gg 1$, there are many unsolved problems of the balance of charged particles and of establishing steady-state parameters of such a plasma [5, Chap. 15; 6, Chap. 8].

In the presence of an appreciable amount of negative ions in a plasma, there usually arise intense electromagnetic oscillations, standing and running strata, and a phenomenon of pinching.

All these phenomena and the chemical activity of most electronegative elements complicate experimental investigations of the ionized gas in which $n_+ / n_e \approx n_- / n_e \gg 1$. Because of scarcity of experimental data and of the complexity of their interpretation, no quantitative theory of plasma in electronegative gases has been worked out so far. The development of such a theory is only in its initial stage (see, e.g., [7, 8] and the references therein).

2. Thompson [9] (see also [5, p. 697]) determined the radial-coordinate dependence of the densities of electrons ($n_e$) and of positive ($n_+$) and negative ($n_-$) ions and of the space potential $V_0$ in the positive column (PC) of a discharge in an electronegative gas (oxygen) at low pressures, when $n_- / n_e \approx 20$. The results of these measurements were quite different from the corresponding plots obtained for the PC of discharges in electropositive gases: over a large part of the tube radius ($0 \leq r/R \leq 0.78$) the values of $n_e$ and $V_0$ were independent of the distance from the PC axis.

We decided to measure the above-indicated dependences in the PC of the iodine vapor, a substance which differs appreciably from oxygen studied in [9] with respect to mass ($M(O_2) = 32, M(I_2) = 254$) and electron affinity ($E_a(O_2) = 0.44, E_a(O) = 1.46, E_a(I_2) = 2.51, E_a(I) = 3.06$ eV [5, pp. 211, 59, 240]). We also made an attempt to explain the unusual behavior of the dependences indicated above.

The iodine vapor pressure $p(I_2) = 0.05$ and 0.075 mm Hg and the discharge current density ($j_d = 1.8 \times 10^{-4}$ and $4.4 \times 10^{-4}$ A/cm$^2$) were chosen such as to be close to the experimental conditions used in...
Moreover, at the chosen pressure and current density values the PC was diffuse, devoid of standing strata and pinching, and the plasma noises were weaker, which facilitated the conduction of experiments. The discharge tube was described in [10].

Measurements under conditions close to those used by us in a discharge in the iodine vapor have shown that most of charged particles were $I_2^+$ and $I^-$ ions and electrons [4, 11].

According to estimates [12, p. 412], the discharge PC in the iodine vapor studied by us was in the diffusion conditions for positive and negative ions: the gas-kinetic mean free paths of the ions were $\lambda_+(I_2^+) \approx \lambda_-(I^-) = 0.25 - 0.37$ cm for the iodine vapor pressure $p(I_2) = 7.5 \times 10^{-2}$ and $5 \times 10^{-2}$ mm Hg, i.e., the condition $\lambda_+ \approx \lambda_- < R = 1.9$ cm was fulfilled.

3. The electron concentration was determined by a standard procedure from measured values of electron current to the probe using the Bohm formula. The concentration of negative ions was determined from the quasi-neutrality condition: $n_+(I_2^+) \approx n_-(I^-)$.

Apart from the determination of $n_-(I^-)$, the results of measurement of $n_+(I_2^+)$ were used to estimate $n_-(I^-)$ through the solution of the balance equation, which takes account of the principal processes of formation and decay of negative ions $I^-$. The dominant reactions of formation of charged particles under our experimental conditions are the dissociative sticking and ionization of iodine molecules upon collisions with electrons. The removal of positive and negative ions $I_2^+$ and $I^-$ from the plasma bulk occurs due to ion-ion recombination.

A certain, though lesser, role is played by other processes [4, 11]. At low current densities, as is the case in our experiments, the degree of dissociation is small: $n(I_2) \gg n(I)$ [4].

The balance equation for the formation and decay of negative ions under our experimental conditions may be written in the form

$$K_d n_e(I_2) - K_r n(I^-) n(I_2^+) = 0, \quad (1)$$

where $K_d$ and $K_r$ are the rate constants for dissociative sticking and ion-ion recombination, respectively, with $K_r = 1.47 \times 10^{-8}$ cm$^3$ s$^{-1}$ [11].

The quantity $K_d$ was estimated according to the formula

$$K_d = \int q_d(v_e) f(v_e) v_e \, dv_e. \quad (2)$$

The effective cross section of dissociative sticking $q_d(v_e)$ was taken from [4]. We measured the electron distribution function over the rates $f(v_e)$; it was found to be close to a Maxwell distribution with $kT_e \approx 30$ eV. In our conditions $K_d$ was of the order of $10^{-11}$ cm$^3$ s$^{-1}$.

The estimated values of the negative ion density exceeded the measured ones by no more than two-fold. The measured values of the charged particle concentrations along the PC axis varied with varying iodine vapor pressure and discharge current density within the limits indicated above: $n_e(0)$ from $4 \times 10^7$ to $7 \times 10^7$ cm$^{-3}$, $n_+(0) \approx n_-(0)$ from $3 \times 10^9$ to $9 \times 10^9$ cm$^{-3}$, i.e., we observed an ion-ion plasma with $n_-/n_e \approx 100$, which exceeds the corresponding value found in [9] by about 5 times.

In the case of $n_+ \approx n_+ \gg n_e$, the Debye radius is determined by the concentration of ions and their temperature ($T_+ \approx T_- \approx 400$ K) and is about $10^{-2}$ cm. The ion mean free path $\lambda_+ \approx \lambda_- < R = 1.9$ cm. This means that the object of our investigations is an ion-ion plasma in the diffusion regime for ions.

The dependences $n_+(r/R)$, $n_+(r/R) \approx n_-(r/R)$ measured by us in the discharge PC in the iodine vapor are shown in Fig. 1. The measurements could be performed up to $r/R \approx 0.78$ (up to $r = 1.5$ cm). At distances along the PC axis $r/R \approx 0.6$ the values of $n_+(r/R)$ remained nearly constant, which in our conditions is due to three factors. First, at the iodine vapor pressures used by us the electron mean free path $\lambda_e \gtrsim R$ [12, p. 107]. In these conditions $n_e(r/R) \approx \text{const}$ in the discharge PC in all gases. Second, when the loss of charged particles in the plasma due to recombination prevails over their removal due to diffusion, the concentration distribution over the radius is constant in the central part and experiences a sharp drop near the walls [13, pp. 204–205]. Estimates have shown that this condition is satisfied in our case: the diffusion lifetime of charged particles exceeds by more than 100 times their decay time because of the recombination. This is a natural result of high values of electron affinity of iodine atoms and molecules and of low velocities
Concentrations of electrons \( n_e \), positive ions \( n_+ \), and negative ions \( n_- \) as a function of distance from the PC axis in iodine vapor at \( p(\text{I}_2) = 5 \times 10^{-2} \text{ mm Hg} \), \( i_d = 2 \text{ mA} \), \( n_e(0) = 4 \times 10^7 \text{ cm}^{-3} \); \( n_+(0) \approx n_-(0) \approx 3 \times 10^9 \text{ cm}^{-3} \); \( R = 1.9 \text{ cm} \). The dash line represents the zero-order Bessel function of the first kind.

The third factor responsible for the fact that the electron concentration is almost independent of the radial coordinates at a distance exceeding half of the PC radius is the small flow of negative charges to the tube walls. The electron density is low, which is why the density of the electron flow to the walls is also low. The concentration of negative ions is high, but their temperature is low, and therefore even a weak repelling field does not allow them to get to the walls. Because of this, the density of negative charges on the walls is insignificant.

With increasing distance from the PC axis, the concentration of negative and positive ions monotonically decreases less sharply than the Bessel function (see Fig. 1). This seems to be accounted for by the presence of electrons, whose distribution along the radius is nearly constant.

The space potentials \( V_0(r/R) \) at different distances from the PC axis were measured using mobile probes in two ways: (1) at the point of inflection of the current-voltage curve of the electron current to the probe, \( i_p(V_p) \), and (2) at the point on the current-voltage curve of the second derivative of the current to the probe, where \( i'' = 0 \). Both procedures gave results which are in satisfactory agreement within the experimental error (\( \pm 15 - 20\% \)).

The radial distribution of the space potential \( V_0(r/R) \) is almost independent of the radial coordinates (Fig. 2) approximately up to the same distances from the axis \( (0 \leq r/R \leq 0.6) \) as \( n_e(r/R) \), following which \( n_e \) somewhat decreases as \( r/R \) increases to about 0.78, and the plasma potential \( V_0 \) decreases (see Figs. 1 and 2).

Figure 2 demonstrates that the negative potential drops by about 3 V in the discharge PC in the iodine vapor at a distance \( r \approx 0.78R \approx 1.5 \) from the axis. This change of the potential occurs predominantly at the end of the studied distance \( r \) from the axis: \( \Delta r \approx 0.3 \text{ cm} \) (from \( r \approx 0.6R \) to \( r \approx 0.78R \)).

The behavior of the PC parameters at distances of \( r/R \geq 0.78 \) could not be determined for technical reasons. We measured the potential difference between the PC axis and the tube walls using a wall probe.
It was equal to \( V_0(0) - V_0(R) \approx -45 \) V. The estimate of this quantity according to the Poisson formula for the average value \( n_e(r) \approx 10^7 \text{ cm}^{-3} \) gave a value which is close in order of magnitude to that measured: 
\( V_0(0) - V_0(R) \approx -60 \) V. Hence, in a layer of thickness \( \Delta r = R - 0.78R \approx 0.5 \) cm there occurs a change of the potential by more than \(-40\) V, i.e., in this layer the radial field is much stronger than in the region with the radial coordinates between \( r/R = 0 \) and \( r/R \approx 0.78 \).

Figure 2 also shows the dependence \( V_0(r) \) in the discharge PC in helium. It has a shape characteristic of electronegative gases: the potential \( V_0(r) \) becomes monotonically less and less negative as the distance from the axis increases.

The radial distribution of electrons \( n_e(r) \) and of the potential \( V_0(r) \) is mutually determined by the Boltzmann law [14, p. 240]. Therefore, at distances from the axis where \( n_e(r) \approx \text{const} \) (see Fig. 1) \( V_0(r) \) must also be nearly constant, which is what can be seen from Fig. 2.

4. The radial distribution of negative ions observed in [9] in the discharge in oxygen and in our measurements in the iodine vapor as shown in Fig. 1, can only take place when there are an ion concentration gradient and a retarding field from the walls. This means that the potential \( V_0(r/R) \) in the discharge PC in electronegative gases is such that the space potential slightly decreases (within the measurement accuracy, so it cannot be seen in Fig. 2) with increasing distance from the axis. This generates a radial field \( E_r \neq 0 \) which retards the motion of negative particles to the walls. This field is not strong enough to hinder the motion of high-energy electrons but sufficient for slowing down negative ions with their low temperature. In contrast to the electron-ion plasma, in the ion-ion plasma a specific regime of ambipolar diffusion sets in, which is controlled by negative ions [5, pp. 694–698]. Here we see that the quantitative changes of the quantity \( n_-/n_e \) are responsible for a qualitatively different mechanism of displacement of charged particles from the plasma bulk to the tube walls. For reasons indicated in Sec. 2, the contribution of ambipolar diffusion controlled by negative ions to the removal of ions from the plasma bulk in our experimental conditions is negligibly small compared to the role of bulk recombination.

5. Figure 2 shows that at a distance of \( r \approx 0.6R \approx 1.1 \) cm from the axis the PC in the radial direction may be tentatively divided into two layers: an inner \((0 \leq r \leq 0.6R)\) cylindrical layer, where \( n_e(r) \approx \text{const} \) and \( V_0(r) \approx \text{const} \), and an outer layer \((0.6R \leq r \leq R)\) in the form of a hollow cylinder, where \( n_e(r) \) and \( V_0 \) begin to fall off.

The distance equal to the outer layer wall thickness \( \approx 0.4R \approx 0.76 \) cm) accommodates two or three mean free paths of the ions. The drop of the space potential \( V_0(0) - V_0(R) = -45 \) V occurs mainly in this layer.

In the inner layer the quasi-neutrality condition has the form \( n_+(r) \approx n_-(r) \) and the formation of negative ions I\(^-\), positive ions I\(^+\), and electrons and their disappearance occur as a result of the reactions listed in Sec. 3.

The strong field near the walls, which retards the motion of negatively charged particles, must completely repel the negative ions and attract the positive ions. A high proportion of electrons overcome the retarding potential of the walls because of their high energy. In the stationary regime the flow densities of positive (I\(^+\) ions) and negative (electrons) particles must be the same \((n_+v_+ = n_ev_e)\), so one should assume that an electron-ion bipolar motion of positive ions and electrons in the direction to the walls will set up in the outer layer.

6. It should be pointed out that in discharges of electronegative gases and vapors, when \( n_+ \approx n_- \gg n_e \), the notion of “positive column” (PC) of the discharge assumes a meaning that differs somewhat from that implied in discharges in substances with a negative electron affinity of their particles. Many of the properties and processes that occur in the first type of PC are noticeably different from those in the second. Almost all the characteristics of the PC in electropositive gases are quite satisfactorily described by the Schottky theory and the Langmuir-Tonx theory [12, Sec. 33–37], which cannot be used to estimate many of the PC parameters in electronegative gases since in the latter case certain important assumptions of the above theories are not fulfilled. These include primarily the assumption of the predominance of the mechanism of ambipolar diffusion toward the tube walls in the course of removal of charged particles over their bulk recombination. When \( n_+ \approx n_- \gg n_e \), this assumption holds true since \( m_e/m_+ \ll 1 \) and \( T_e/T_+ \gg 1 \), which gives rise to a large difference in mobility between electrons and ions: \( \mu_e \gg \mu_+ \approx \mu_- \). In the so-called PC, where \( n_+ \approx n_- \gg n_e \), we have an opposite situation: the masses, temperatures and mobilities of the major part of charge carriers (positive and negative ions) are almost equal. In such an ionized gas the principal cause of decay of charged particles is their recombination in the bulk even with such low ionization
degrees and low gas pressures at which in electropositive gases the contribution of bulk recombination may be neglected as compared to particle diffusion to the walls. This is also favored by the nearly complete absence of a radial electric field in the greater part of the PC cross section, which in turn is caused by the low diffusion rate of charged particles controlled by negative ions and directed to the walls, and by the low electron density. These factors are responsible for the fact that in an ion-ion plasma there is no permanent excess of the density of positive charges over negative ones.

The neutral plasma zone located between the anodic and cathodic parts of the discharge in electronegative gases may be tentatively called a positive column, which we do in the present paper, keeping in mind the above remarks concerning the difference between the PC properties in electronegative and electropositive substances.

What has been said about the differences in the PC properties between iodine and electropositive gases should be qualitatively valid not only for O\textsubscript{2} and I\textsubscript{2} but also for other substances whose atoms and molecules have positive values of electron affinity, and also for other values of pressure and current density. Naturally, the distances \( r/R \) at which \( n_\text{e}(r) \) and \( V_0(r) \) remain constant will be different.

In conclusion, the following inferences may be made.

1. The dependences \( n_\text{e}(r/R) \) and \( V_0(r/R) \) obtained in [9] and in our work are apparently characteristic of the discharge PC in all electronegative gases when \( n_\text{e}/n_\text{e} \approx n_-/n_\text{e} \gg 1 \).

2. In spite of the fact that \( n_\text{e} \ll n_\text{e} \approx n_- \) in such discharges, the dependence \( V_0(r) \) is determined by the radial distribution \( n_\text{e}(r) \). The latter also affects the form of \( n_\text{e}(r) \approx n_- (r) \).

3. The discharge PC in electronegative gases may be divided in the radial direction into two layers, which are sharply different with respect to the behavior of \( V_0(r) \) and \( n_\text{e}(r) \) and the mechanisms that control the balance of charged particles.

In order to solve many of the physical problems of an ionized gas in electronegative gases, it is necessary to conduct systematic studies of its parameters by widely varying the values of electron affinity of the gas particles, pressure, current density, and size of the discharge tube.

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**REFERENCES**


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