Determination of Anisotropic Ion Velocity Distribution Function in Intrinsic Gas Plasma. Theory.

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Abstract. The first seven coefficients of the expansion of the energy and angular distribution functions in Legendre polynomials for Hg⁺ ions in Hg vapor plasma with the parameter E/P ≈ 400 V/(cm Torr) are measured for the first time using a planar one-sided probe. The analytic solution to the Boltzmann kinetic equation for ions in the plasma of their parent gas is obtained in the conditions when the resonant charge exchange is the predominant process, and ions acquire on their mean free path a velocity much higher than the characteristic velocity of thermal motion of atoms. The presence of an ambipolar field of an arbitrary strength is taken into account. It is shown that the ion velocity distribution function is determined by two parameters and differs substantially from the Maxwellian distribution. Comparison of the results of calculation of the drift velocity of He⁺ ions in He, Ar⁺ in Ar, and Hg⁺ in Hg with the available experimental data shows their conformity. The results of the calculation of the ion distribution function correctly describe the experimental data obtained from its measurement. Analysis of the result shows that in spite of the presence of the strong field, the ion velocity distribution functions are isotropic for ion velocities lower than the average thermal velocity of atoms. With increasing ion velocity, the distribution becomes more and more extended in the direction of the electric field.

1. Introduction

The ion velocity distribution function (VDF) is interesting for many reasons among which there are problems associated with the analysis of plasma chemical reactions involving ions, determination of the ion mobility in a plasma object, heating of the neutral component of the plasma, etc. Among the technical applications one can mention modern plasma nanotechnologies, fine cleaning of surfaces of objects with ions and the technology of producing reliefs on surfaces by selective etching by means of bombardment with ion beams [1]. Peculiarities of ion drift were studied theoretically in [2–13], but the results are described most comprehensively in [5–13]. Perel [6] reported on the results of calculation of the drift velocity of ions of inert gases in their parent gas; however, the ion VDF was not calculated explicitly. The drift velocity of ions of inert gases in their parent gas plasma was also calculated in [5] in the strong field approximation; the author of [5] believed that the distribution of atoms has the form of a delta function. The author of [8], who determined the distribution function of ions in their parent gas, disregarded the generation of ions with a Maxwellian velocity distribution as a result of charge exchange. It will be shown below that this may lead to considerable errors in the ion VDF.

The time during which an ion moving in its parent gas (if we disregard all processes except charge exchange) has a velocity component in the direction of the electric field in the interval from \( v_{\parallel} \) to \( v_{\parallel} + dv_{\parallel} \) [7]. Clearly, this time is proportional to the ion distribution function of the velocity

\( dv_{\parallel} \) [7]. Clearly, this time is proportional to the ion distribution function of the velocity
projection onto the electric field direction; however, it is difficult to obtain from these results the total velocity distribution function.

In [9, 10], a new method for calculating matrix elements of the collision integral was developed, which was used for solving the time-dependent Boltzmann equation by the method of moments for ions in the conditions when the resonant charge exchange is the predominant process. However, the application of this method for calculating the time-independent VDF for ions is hampered in strong field, when the ratio of the thermal energy of atoms and the energy of an ion acquired over the mean free path is less than 0.05.

In [11], the analytic solution to the Boltzmann equation for ions in the Bhatnagar–Gross–Krook (BGK) model [14] is compared with the numerical solution for a constant resonant charge-exchange cross section. It is shown that in strong fields, the analytic solution gives an incorrect asymptotic form for the drift velocity of an ion.

The author of [12] developed the moments method for solving the Boltzmann equation for charged particles for different potentials of their interaction with atoms.

In [13], a review of the methods for solving the Boltzmann equation for charged particles is given both for the regimes close to hydrodynamic conditions and in the opposite case. In the latter, the principles of the moment method (and its modifications) are given. The cases of both constant and varying fields (in particular, RF discharge) are considered.

As regards the measurement of the VDF for ions in the plasma of a self-consistent dc discharge, we are not aware of publications except [15], in which the Doppler shift of the ion lines is measured spectroscopically for a discharge in argon and observing along the discharge axis. From this shift, the average velocity of ions was estimated, which was found to be \(10^4\) cm/s in the given experimental conditions.

Various aspects of ion drift in the plasma of low-pressure capacitive discharge are studied theoretically in [16–18].

This study is devoted to experimental and theoretical determination of the ion VDF in their parent gas of the glow discharge taking into account the generation of slow ions with the atomic temperature as a result of charge exchange, which was treated as the dominating process. We assumed that the velocity of ions prior to collisions considerably exceeds the velocity of atoms.

2. Theoretical determination of the ion distribution function

2.1. Derivation of Basic Relations for a Constant Cross Section in Zero Ambipolar Field

The mechanisms of the interaction of ions with remaining components of a low-temperature plasma have been studied comprehensively [2, 19, 20]. It was found that the dominating process in collisions of an ion in its parent gas is resonant charge exchange as a result of which an ion colliding with an atom transfers to it its charge due to the tunnel effect, but preserves the energy and momentum. Thus, the motion of the ion in its parent gas is a relay-race process (Sena effect [2]).

Considering the main mechanism of interaction of ions with neutral atoms, we analyze the steady-state velocity distribution under the following conditions: (i) gas atoms move in accordance with the Maxwell law; (ii) ions move in their parent gas with a low degree of ionization; (iii) the dominating process that forms the ion velocity distribution function is resonant charge exchange; (iv) longitudinal gradients of the plasma parameters are equal to zero. The last condition is necessary because otherwise the ion VDF will also depend on the longitudinal coordinate. The solution to the problem in this case can also be obtained easily, but will require the imposition of physically substantiated boundary conditions along the \(z\) coordinate.

In a stationary plasma under the above assumptions, the Boltzmann equation has the form:

\[
\bar{v}_i \frac{d}{d\varphi} \bar{f}_i + eE \frac{1}{m} \bar{v}_i \frac{d}{d\varphi} \bar{f}_i = S_i,
\]  

(1)
where $e$ and $m$ are the ion charge and mass, $E$ is the electric field strength, $v_i$ is the velocity of ions, $S_i$ is the collision integral. Considering that an ion appearing as a result of charge exchange as well as a result of an electron impact has the velocity of the atom, we define the collision integral as

$$S_i (v_i) = n_a \int \sigma v_a f_a (v_a^*) f_i (v_i^*) \left[ \delta (v_i^* - v_i) - \delta (v_i^* - v_i^* - v_i) \right] dv_a d v_i^*, \quad (2)$$

where $f_a (v_a^*)$ is the atomic distribution function, $n_a$ is the concentration of atoms, $\sigma$ is the charge-exchange cross section, $v_r$ is the magnitude of the relative velocity of ion–atom, and $v_a$ is the velocity of atoms. In the collision integral, we disregard the ionization and electron–ion recombination processes since under the given assumptions, their characteristic time is large as compared to the resonant charge-exchange time. The ion distribution function is normalized to the concentration and the distribution function for atoms is normalized to unity.

First of all, let us specify the functions appearing in the collision integral. It is well known that the charge exchange cross section is a logarithmic function of the ion–atom relative velocity [5, 20, 21]:

$$\sigma (v) = \sigma_0 \left[1 + a \cdot \ln \left( \frac{v_0^2}{v^2} \right) \right] \equiv \sigma_0 \kappa (v), \quad (2a)$$

where $\sigma_0, a, v_0$ are the constants depending on the type of the gas. For example, $\sigma_0 = 2.79 \cdot 10^{-15} \text{ cm}^2$ and $a = 0.0557$ for helium with $\frac{mv_0^2}{2} = 1 \text{ eV}$ [21], while $\sigma_0 = 12 \cdot 10^{-15} \text{ cm}^2$ and $a = 0.073$ for resonant charge exchange of Hg$^+$ ions [22]. At the same time, the author of [23] mentioned the following values: $\sigma_0 = 20 \cdot 10^{-15} \text{ cm}^2$ and $a = 0.048$. In view of such a weak energy dependence in a comparatively narrow velocity range typical of the motion of ions, the charge-exchange cross section can be treated as constant with a high degree of accuracy. Indeed, in the energy range 0.1–0.3 eV, the charge-exchange cross section (e.g., for helium) changes by approximately 5% according to the results from [9, 21].

For this reason, we will first solve the problem assuming that the resonant charge-exchange cross section is constant and then write the formulas taking into account its dependence on velocity. Let us consider the conditions of motion of ions in which the velocity acquired by them over the mean free path is considerably higher than the average velocity of atoms; such conditions hold if

$$\left( \frac{P}{T_a} \right) \ll 1,$$

where $P$ [Torr] is the gas pressure, $T$ is measured in $[\text{V/cm}]$, and $\sigma [10^{-15} \text{ cm}^2]$. (This inequality is equivalent to

$$(E \cdot P^{-1} \cdot \sigma^{-1})^{1/2} \gg 1, \quad (2b)$$

where $P$ [Torr] is the gas pressure, $E$ is measured in [V/cm], and $\sigma [10^{-15} \text{ cm}^2]$.)

It should be noted that in the case of inert gases, inequality (2b) holds only for small values of the product $PR \ll 1$ ($R$ is the characteristic size of the volume in which the plasma is produced), when the electric field in the plasma increases substantially due to the diffusion induced by the perish of electrons [24]. In this case, we assume that the ion–atom relative velocity prior to the collision has only one component along the electric field strength $\vec{E}$ and is determined only by the velocity of the ion. We also assume that the atomic collision frequency is considerably higher than the frequency of ion–atom collisions. This allows us to use the Maxwell equilibrium distribution for atoms, but imposes a limitation on the degree of ionization. Under the above assumptions, the collision integral can be reduced to the expression:
\[ S_i(\vec{v}_i) = n_\alpha \sigma [f_\alpha(\vec{v}_i)] \int [v_{iz}] f_i(\vec{v}_i) d\vec{v}_i - [v_{iz}] f_i(\vec{v}_i)]. \] (3)

Let us first consider the case when the plasma is not confined by the walls and the electric field has one component directed along the \( z \) axis. We assume that this field component, as well as the concentrations of atoms and ions, is independent of the coordinate. Then, distribution function \( f_i \) depends only on velocity \( \vec{v}_i \) and we can, using expression (3), reduce the kinetic equation to the following form up to terms of the order \( \frac{\alpha_0}{\beta} \):

\[ \frac{\delta f_i}{\delta v_{iz}} + 2\alpha_0 v_{iz} f_i = \gamma f_\alpha(\vec{v}_i), \] (4)

where \( O(x) \) is a quantity of the same order of magnitude as \( x \) for \( x \to 0 \); \( 2\alpha_0 = \frac{m}{eE} n_\alpha \sigma \); \( \gamma = 2\alpha_0 n_i \vec{v}_{iz} \); \( f_\alpha(\vec{v}_i) = \left( \frac{\beta}{\pi} \right)^{1.5} \exp(-\beta v_i^2) \); \( \beta = \frac{m}{2kT_\alpha} \); \( T_\alpha \) - is the temperature of atoms, \( \vec{v}_{iz} = \frac{\int v_{iz} f_i(\vec{v}_i) d\vec{v}_i}{\int f_i(\vec{v}_i) d\vec{v}_i} \) is the average velocity of ions in the direction of the electric field, and \( n_i \) is the ion concentration.

Let us now assume that the plasma is produced in a cylindrical discharge tube of radius \( R \) with dielectric walls and the possibility of the plasma being heated along the radius non-uniformly. In this case, the left hand side of Eq. (4) acquires three additional terms \( \frac{m}{eF} v_\rho \frac{\delta f_i}{\delta \rho} \), \( E_\rho(\rho) \frac{\delta f_i}{\delta \rho} \), \( \frac{m}{eF} n_\rho \frac{\delta n_i}{\delta \rho} f_i \), where \( E_\rho \) is the ambipolar field, \( \rho \) is the radial coordinate, and \( v_\rho \) is the ion velocity component in the radial direction. The last term is of the order of \( \frac{m}{eF} n_\rho \frac{\delta n_i}{\delta \rho} f_i \) for \( \lambda_i \ll R \), where \( \lambda_i \) is the path length of an ion in respect with the resonant charge exchange, and therefore can be disregarded. The second term is much smaller than \( \frac{\delta f_i}{\delta v_{iz}} \) if the inequality \( E_\rho(\rho) \ll E \) holds in the self-sustained plasma discharge. This inequality is observed near the discharge axis (where the total ambipolar field is zero) or in the entire volume if the following relation holds [24]:

\[ PR \gg 10^{-3} \cdot T_a \cdot \sigma_e^{-1} \cdot \delta^{-1/2}, \] (4a)

where \( \sigma_e \) is the elastic scattering cross section of an electron from a neutral particle and \( \delta \) is the average relative fraction of the electron energy lost by in the elastic scattering from neutral particles of the plasma; \( P \) [Torr], \( R \) [cm], \( T_a \) [K], \( \sigma_e \) [10^{-16} cm^{2}].

The first additional term is obviously of the order of the last term; therefore, all three terms can be disregarded. If inequalities (4a) and \( \lambda_i \ll R \) hold, in the case of a nonuniform heating of the gas, the \( f_i(\rho) \) dependence can be taken into account just assuming that quantities \( \alpha_0, \beta, \gamma \) are functions of the radius.

Let us first assume that these inequalities hold and parameters \( \alpha_0, \beta, \gamma \) that determine the properties of the plasma are specified a priori (including their possible dependence on radial coordinate \( \rho \)). It should be noted that parameter \( \alpha_0 \) determines the velocity acquired by an ion in the electric field, parameter \( \beta \) characterizes the temperature of neutral atoms and \( \gamma \) is the rate of formation of ions due to charge exchange.

The solution to kinetic equation (4) when inequalities (4a) and \( \lambda_i \ll R \) take place (inequality (4a) for small \( \frac{\rho}{R} \) holds because \( E_\rho(0) = 0 \) ), taking into account the behavior of the ion velocity distribution function for \( v_{iz} \to -\infty \), has the form

\[ f_i(\vec{v}_i) = A \exp \left[ -\beta v_i^2 + (\beta - \alpha_0)v_{iz}^2 \right] \text{erfc} \left( -\sqrt{\beta - \alpha_0} v_{iz} \right) \left\{ 1 + O \left( \frac{\alpha_0}{\beta} \right) \right\} \text{при } v_{iz} \geq 0; \]

\[ f_i(\vec{v}_i) = A \exp \left[ -\beta v_i^2 + (\beta + \alpha_0)v_{iz}^2 \right] \text{erfc} \left( -\sqrt{\beta + \alpha_0} v_{iz} \right) \left\{ 1 + O \left( \frac{\alpha_0}{\beta} \right) \right\} \text{при } v_{iz} \leq 0, \] (5)
where \( A = \frac{\gamma \beta}{2 \pi} \) is a constant factor, \( v_i \) is the magnitude of the ion velocity, and \( \text{erf}(x) = \frac{2}{\sqrt{\pi}} \int_{0}^{x} \exp(-t^2) \, dt \). Under the above assumptions, the inequality \( \frac{a_0}{\beta} \ll 1 \) holds. Note that expression (5) for \( v_{iz} = v_i \mu; \mu = \cos \theta \) (\( \theta \) is the angle between the \( z \) axis and the ion velocity) gives the angular velocity distribution for ions. Integrating the relation (5) with respect to angular coordinates, we obtain the ion distribution function over velocity modulus, normalized to unity:

\[
F_i(v_i) \equiv \frac{f_i(v_i)}{n_i} v_i^2 = \frac{2}{\sqrt{\pi}} a_0 \beta v_i \exp(-\beta v_i^2) \int_{-\sqrt{\beta a_0 v_i}}^{\sqrt{\beta a_0 v_i}} \exp(t^2) \text{erf}(t) dt \cdot \left\{ 1 + O\left( \frac{a_0}{\beta} \right) \right\}. \tag{6}
\]

The ion concentration can be expressed in terms of the constants defined above as follows:

\[
n_i = \frac{\gamma \beta}{2 \sqrt{a_0}}. \]  

It should be noted that to take into account terms \( \frac{\gamma}{\beta} \), it is sufficient to specify the normalization in this formula i.e., divide the function by \( F_i(v_i) v_i^2 dv_i \).

Integration of relation (5) with respect to the azimuthal angle and the velocity component orthogonal to the electric field direction gives the distribution of ions over the \( z \) component of velocity \( v_{iz} \), normalized to the concentration:

\[
f_{iz}(v_{iz}) = \frac{n_i A}{\beta} \exp(-\alpha_0 v_{iz}^2) \text{erf}(\sqrt{\beta - \alpha_0 v_{iz}}) \left\{ 1 + O\left( \frac{\alpha_0}{\beta} \right) \right\} \;) \] \quad \text{при } v_{iz} \geq 0; \tag{6a}
\[
f_{iz}(v_{iz}) = \frac{n_i A}{\beta} \exp(\alpha_0 v_{iz}^2) \text{erf}(\sqrt{\beta + \alpha_0 v_{iz}}) \left\{ 1 + O\left( \frac{\alpha_0}{\beta} \right) \right\} \; \) \quad \text{при } v_{iz} \leq 0.
\]

The ion energy distribution function (IDF) \( F_i(\varepsilon_i) \), normalized to unity can be written (with the same accuracy as relation (6)) in the form:

\[
F_i(\varepsilon_i) = \frac{1.16 \times 10^4 \sqrt{a_0}}{\sqrt{\pi} r_a} \exp(-E(\varepsilon_i)) \int \sqrt{\frac{(1-\frac{\alpha_0}{\beta}) E(\varepsilon_i)}{1+\frac{\alpha_0}{\beta} E(\varepsilon_i)}} \exp(t^2) \text{erf}(t) dt \left\{ 1 + O\left( \frac{\alpha_0}{\beta} \right) \right\}. \tag{6b}
\]

where \( \varepsilon_i \) is the ion energy in electron volts, \( E(\varepsilon_i) = \frac{1.16 \times 10^4 \varepsilon_i}{r_a} \), terms \( O\left( \frac{\alpha_0}{\beta} \right) \) are taken into account analogously to formula (6) (i.e., by specifying the renormalization), and the azimuthal angle and energy IDF have the form:

\[
F_i(\varepsilon_i) = \frac{1.16 \times 10^4 \sqrt{a_0}}{\sqrt{\pi} r_a} \sqrt{\frac{E(\varepsilon_i)}{\frac{1}{2} + \frac{\alpha_0}{\beta} E(\varepsilon_i) \mu}} \exp \left[ -E(\varepsilon_i)(1 - \mu^2) - \frac{\alpha_0}{\beta} E(\varepsilon_i) \mu^2 \right] \text{erf} \left[ -\sqrt{1 - \frac{\alpha_0}{\beta} E(\varepsilon_i) \mu} \right] \tag{6c}
\]

for \( \mu > 0 \)

\[
F_i(\varepsilon_i) = \frac{1.16 \times 10^4 \sqrt{a_0}}{\sqrt{\pi} r_a} \sqrt{\frac{E(\varepsilon_i)}{\frac{1}{2} + \frac{\alpha_0}{\beta} E(\varepsilon_i) \mu}} \exp \left[ -E(\varepsilon_i)(1 - \mu^2) + \frac{\alpha_0}{\beta} E(\varepsilon_i) \mu^2 \right] \text{erf} \left[ -\sqrt{1 + \frac{\alpha_0}{\beta} E(\varepsilon_i) \mu} \right] \]

for \( \mu < 0 \).

It follows from the expression for \( n_i \) and from definition of parameters \( \alpha_0 \) and \( \gamma \) that the average velocity of an ion is defined to within terms of the order of \( \frac{\alpha_0}{\beta} \) as:

\[
\bar{v}_i = \sqrt{\frac{1}{n a_0} \left[ 1 + O\left( \frac{\alpha_0}{\beta} \right) \right]} \approx \frac{2eE}{\pi \alpha a \sigma}. \tag{7}
\]
The same result can be obtained by evaluating the average velocity directly using the distribution function obtained above. The calculation of the square mean velocity with the same degree of accuracy gives

\[ \sqrt{\langle v_i^2 \rangle} = \left( \frac{1}{n_i} \int_0^\infty F_i(v_i) \, dv_i \right)^{0.5} = \frac{1}{\sqrt{2\alpha_0}} \left[ 1 + O\left( \frac{a_0}{\beta} \right) \right], \]

(7a)

while for the most probable velocity \( v_{ip} \) in the approximation \( \alpha_0 \to 0 \) we find from the solution of the transcendental equation \( \frac{dF_i(v_i)}{dv_i} \bigg|_{v_i=v_{ip}} = 0 \), that \( v_{ip} = \frac{1.5}{\sqrt{\beta}} \). The allowance for the finite value of parameter \( \alpha_0 \ll \beta \) shifts the position of the peak of the ion VDF towards lower velocities.

2.2. Inclusion of the Ambipolar Field and Dependence of the Charge-Exchange Cross Section on Velocity

In the case when the ambipolar field must be taken into account (i.e., if we seek the solution not in the axial region of the discharge or if inequality (4a) does not hold) but the inequality \( \lambda_i \ll R \) still takes place, we proceed as follows. Taking into account the fact that we can disregard in our case the spatial gradients in the Boltzmann equation and that the resonant charge exchange is the main process determining the form of the distribution function, we will solve the problem for a certain real coordinate \( \rho \) by choosing a local system of coordinates in which the \( z \) axis coincides with the direction of the total electric field in the plasma for this value of \( \rho \):

\[ E_\rho = E \sqrt{1 + \epsilon(\rho)^2}, \]

(8)

where \( \epsilon(\rho) = \frac{E_\rho(\rho)}{E} \).

Then, the Boltzmann equation has the form (4), but the parameter \( \alpha_0 \) depends on the ratio of the ambipolar and axial (directed along the discharge tube axis) fields:

\[ \alpha(\rho) = \alpha_0 \sqrt{1 + \epsilon(\rho)^2}. \]

(8a)

Respectively, expressions (5) and (6) hold after the substitution of \( \alpha_0 \) for the parameter \( \alpha(\rho) \). In addition, a simple geometrical analysis shows that we must replace \( v_\rho = v\mu \) in formula (5) by the expression \( v\sqrt{\frac{\mu + \epsilon(\rho)\sqrt{1-\mu^2}}{1+\epsilon(\rho)^2}} \), and \( v_\rho = v\sqrt{1-\mu^2} \) by \( \sqrt{\frac{\epsilon(\rho)\mu - \sqrt{1-\mu^2}}{1+\epsilon(\rho)^2}} \). It should be noted that we have not imposed any constraints on the relation between the axial and ambipolar fields. As regards the expression for the drift velocity in the axial direction, taking into account the fact that the relation \( \tilde{v}_i = \tilde{v}_{ix}\sqrt{1 + \epsilon(\rho)^2} \),

(9)

holds in the chosen system of coordinate, we can write

\[ \tilde{v}_{ix} = \frac{2eE}{m_{e\sigma}^2} \frac{1}{[1+\epsilon(\rho)]^{0.25}}. \]

(10)

Concluding this section, let us consider how the resultant formulas will change if we take into account the dependence of the resonant charge-exchange cross section on the velocity. Calculations
show that the values of quantities \( \frac{x^2 \kappa(x)}{2} \) and \( \int_0^x \kappa(y) dy \), (e.g., for inert gases) change with the variation of the quantity \( x = \sqrt{\beta} v_i \) in the range \( x \in [0, 10] \) by not more than 5\%, where \( \kappa(x) \) is defined by formula (2a). We can easily show that all expressions derived above describe to the same degree of accuracy the corresponding quantities in the case of dependence of the charge-exchange cross section on the velocity in accordance with expression (2a) after the substitution of \( \alpha(\rho) \) for \( \alpha(\rho)\kappa(v_i) \).

The problem was solved under the assumption that inequality \( \lambda_i \ll R \) holds. This is necessary for disregarding the term with \( \bar{V}_r(f_i) \) in kinetic equation (1). However, even if this inequality does not hold, the above results are still valid in the vicinity of the tube axis because \( \bar{V}_r(f_i)_{r=0} = 0 \). We can easily prove that for an arbitrary ratio , the exact solution to the Boltzmann equation tends to the solution to Eq. (4) for \( \rho \to 0 \).

3. Experimental determination of the ion distribution function in plasma glow discharge

We measured the energy IDF by the probe method in the positive column of the low-pressure glow discharge in mercury. The discharge was initiated in a quartz glass tube 30 mm in diameter and 300 mm in length between the planar impregnated indirectly heated cathode 11 mm in diameter and a molybdenum anode with a diameter of 20 mm. Preliminary thermal preparation of the device and oil-free evacuation ensured a vacuum of \( 10^{-8} \) Torr in the working regime.

The cathode temperature was measured by a W–Re microthermocouple and was maintained by the electron stabilization system at a level of 1000–1500 K to within \( \pm 5 \) K. The stability of the emission cathode current and the discharge current during measurements at various mercury pressures were controlled by the electronic stabilization system.

Mercury was fed to the tube using the vacuum distillation method. The saturated vapor pressure was controlled by the temperature of the thermostat containing the liquid phase of mercury. The mercury pressure varied in the range \( 5 \cdot 10^{-4} \)–1 Torr and was monitored by magnetodischarge sensors. The upper boundary of the pressure was determined by the sizes of probes and the applicability conditions for the collisionless theory of the probe current. The discharge current varied from 0.05 to 0.5 A. The estimates of the near-probe layer thickness show that the collisionless layer approximation [25, 26] holds in experimental conditions, and the thickness of this layer is much smaller than the probe diameter.

The main experimental difficulty in the IDF measurements with a planar probe is that it must rotate and simultaneously move along the discharge axis. To overcome this difficulty, a rotating planar disk-shaped one-sided probe made of 30-\( \mu \)m-thick tantalum foil 0.5 or 0.8 mm in diameter is introduced into the plasma through the lateral boundary. The probe is located on the discharge tube axis. A tantalum wire lead of diameter 0.1 mm is welded to the probe. The lead and one side of the probe are protected with a special alundum coating annealed in vacuum. The probe was mounted in accordance with a three-coordinate micrometric remote displacement system, which ensured its spatial fixation to within \( \pm 0.01 \) mm and orientation relative to the symmetry axis of the discharge in the angular range 0–180° with a step of 5° with accuracy not worse than \( \pm 10'' \). The installation of the probe was monitored with the help of an ocular micrometer.

Legendre coefficients \( F_{\frac{n}{2}}^m \) of the IDF were measured by recording the second derivatives of the probe current \( I''_y \), obtained by double modulation of the probe potential. The experimental setup and the method for measuring the second derivative of the probe current with respect to the probe potential were described in detail in [27, 28]. We used a modulating signal of the form \( u(t) = \Delta \varepsilon(1 + \cos \omega_1 t) \cos \omega_2 t \).

The consideration of hardware distortions, the choice of the optimal amplitudes of the differentiating signal \( \sqrt{2} \omega_1 \) and the control over the recording system linearity were carried out in accordance with the technique described in [29, 30]. We used the following values: \( \Delta \varepsilon = 0.05, 0.1, \) and \( 0.2 \) V, \( \omega_1 = 6 \cdot 10^3 \) Hz and \( \omega_2 = 6 \cdot 10^5 \) Hz.
In the range of negative probe potentials, the value of $I_U$ is determined by the electron component of the probe current and is proportional to the electron VDF. It is well known [31] that for a positive probe potential, the electron current to the planar probe is almost independent of this potential. Ions move in this case in the retarding field, and the second derivative of the ion current carries information on their distribution function. For this reason, $I_U$ in the potential range 0–1 V is mainly describes the ion distribution. Special attention in recording $I_U$ was paid to the stability of the discharge current.

In our experiment, we tried to eliminate distortions emerging during IDF measurement due to the finite conductivity of plasma [32, 33]. For this purpose, a planar one-sided probe and a spherical probe of diameter 0.3 mm were soldered into the tube at distances of 20 and 25 cm from the cathode, respectively, for monitoring the accuracy in the reconstruction of the isotropic component $F_{0z}$ of the IDF. The absence of distorting effect of vibrations in measurements was monitored by recording $I_U$ in the absence of a differentiating signal.

The special measures taken in our experiment for stabilizing the electric discharge parameters ensured the reproducibility of the results not worse than 0.5%. The oscillation spectrum of the discharge current and the voltage were monitored in a frequency range up to 300 MHz.

The special calibration of the measuring system ensured not only relative, but also absolute measurements of $I'_U$, which in turn made it possible to determine angular harmonics of the distribution function $F_{0z}$ in an absolute way.

The spatial potential was determined as the zero of the second derivative of the probe current [26]. The electron and ion concentrations were calculated by integrating the distribution functions for electrons and ions. Comparison of these concentrations shows their coincidence to within 10%, which is a satisfactory result considering the error of probe measurements.

The experiment was controlled with help of a PC based multichannel measuring–computational system. Special software and radio-engineering elemental base made it possible to perform digital recording of the data obtained in pulse and stationary regimes and complex processing in real time scale.

Digital recording made it possible to substantially improve the accuracy and sensitivity of the diagnostic method and elevated the authenticity of experimental results.

4. Discussion of results and comparison of experimental and computation data

It should be noted that expression (7) for the ion drift velocity coincides with the results obtained in [6, 19] in the limit case of a strong field. Analogously, formula (10) coincides with the result reported in [15]. As mentioned above the time during which an ion has a velocity component along the electric field in the interval from $v_{iz}$ to $v_{iz} + dv_{iz}$ was calculated in [7]. This time determines the IDF for the $z$ component of velocity. Distribution (6a) obtained in this study coincides with expressions (47) (for $v_{iz} \geq 0$) and (46) (for $v_{iz} \leq 0$) from [7] for $\frac{\beta}{a_0} \gg 1$. When the relation $\frac{\beta}{a_0} \to \infty$ takes place, distribution (6a) is transformed to the Maxwellian distribution identical to those obtained in [8]. As noted above, this corresponds to the disregard of ions generated as a result of charge exchange with thermal velocities. Thus, we can state that the result obtained here for the IDF are in conformity with the results of other authors.

Let us now consider some peculiarities of distribution function (6). It can be seen that the ion velocity modulus distribution function depends on two parameters, $\alpha_0$ and $\beta$, which distinguishes this distribution from the well-known equilibrium Maxwellian distribution.
Figure 1. Distribution function $x^2 f_i(x)$ for ions and $x^2 f_g(x)$ for ions (see formula (11) from [8]): $x = \sqrt{\beta} v$, (1) $x^2 f_i(x)$ for $\alpha_0/\beta = 0.1$, (2) $x^2 f_i(x)$ for $\alpha_0/\beta = 0.01$, (3) $x^2 f_m(x)$; and (4) $x^2 f_g(x)$ for $\alpha_0/\beta = 0.01$.

Figure 2. Distribution functions $x^2 f_i(x)$ and $x^2 f_g(x)$ for $\alpha_0/\beta = 0.02$ for different ratios $\epsilon(\rho)$ of the ambipolar field to the axial field: (1) $x^2 f_i(x)$ for $\epsilon(\rho) = 0$; (2) $x^2 f_i(x)$ for $\epsilon(\rho) = 0.5$; (3) $x^2 f_i(x)$ for $\epsilon(\rho) = 1$; (4) $x^2 f_i(x)$ for $\epsilon(\rho) = 2$; and (5) $x^2 f_g(x)$.

Figure 1 shows the curves describing functions $x^2 f_i(x)$, where $x = \sqrt{\beta} v$, for various values of the parameter $\frac{\alpha_0}{\beta}$ according to (6), of the Maxwellian distribution $x^2 f_m(x) = \frac{4}{\sqrt{\pi}} x^2 \exp(-x^2)$ and the distribution obtained in [8] for the constant resonant charge-exchange cross section:

$$x^2 f_g(x) = \frac{4}{\sqrt{\pi}} \left(\frac{\alpha_0}{\beta}\right)^{1.5} x^2 \exp\left(-\frac{2\alpha_0 x^2}{\beta}\right).$$

(11)

It can be seen that the number of particles for function $x^2 f_i(x)$ in the range of high velocities is much larger than for the equilibrium distribution with atomic temperature. The opposite relation is observed in the range of low velocities. This can easily be explained by the fact that the excess of ions in the range of high velocities is associated with their acceleration by the electric field, while the deficit of ions in the range of low velocities as compared to the Maxwellian distribution is due to persistent departure of ions from this region as a result of the same acceleration in the electric field. At the same time, the departure of particles in the range of low energies in the case of the equilibrium distribution is a discrete process that occurs only during their collisions with other particles (some of the collisions lead to energy loss).

It should also be noted that the group of slow ions forming the ion VDF in the region of its peak has the characteristic velocity on the order of the atomic velocity. However, this does not mean that the assumption that the ion velocity prior to the collision must be much higher than the thermal velocity of atoms is violated for this group of ions. Indeed, the group of ions with thermal velocities appears precisely as a result of charge exchange; i.e., these are the ions that have just been generated and that must traverse before the next collision a distance of the order of the path length over which they are accelerated in the field to high (as compared to thermal) velocities.

Comparison of distributions (6) and (11) (Figs. 1, 2) shows that these distributions differ radically. This is due to the difference in the physical models used for them. As mentioned above, the author of
[8] disregarded the generation of ions as a result of charge exchange assuming that this can be done since the velocity of an ion generated in this way is much lower than the average ion velocity (acquired due to the field over the path length) in accordance with the conditions of the problem. This is true indeed, but the concentration produced by the ion accelerated in the field in the case of equal fluxes is disregarded. To clarify this situation, it is sufficient to consider the 1D problem of motion of a charged particle beam in the field. In the stationary case, since the flux is constant, the concentration decreases upon an increase in the velocity. It is for this reason that the most probable velocity (obtained in this study) is determined by the temperature of atoms (i.e., ions that have just been generated).

It can be seen from results (7) and (7a) that upon an increase in the ambipolar field, the average and square mean velocities of ions become higher, which is quite clear because ions are additionally accelerated in the ambipolar field. At the same time, the average velocity along the axial field decreases in accordance with the relation (10). This is completely confirmed by the data depicted in Fig. 2, which shows IDFs for various parameters $\alpha(\rho)$. The same figure shows distribution function $x^2f_g(x)$ [8]. It is shown above that the most probable velocity of ions has the same order of magnitude as the most probable velocity of neutral atoms and is determined by parameter $\beta$. On the other hand, average and square mean velocities are determined by parameter $\alpha(\rho)$ in accordance with expressions (7) and (7a).

![Figure 3](image-url)

**Figure 3.** Distribution functions for He$^+$ ions for $\rho = 0$, $\frac{\alpha_0}{\beta} = 0.01$, $T_a = 300$ K; (1) calculation disregarding the velocity dependence of the charge-exchange cross section; (2) calculation taking this dependence into account.

![Figure 4](image-url)

**Figure 4.** Comparison of the dependence of the drift velocity of He$^+$ ions in He, Hg$^+$ ions in Hg vapor, and Ar$^+$ in Ar in strong fields, calculated from distribution function (5), with experimental data, $P_0 = P \times 273.16 / T_a (K)$; (1) calculations based on the developed theory for He$^+$ in He; (2) experimental data [34]; (3) experimental data [35]; (4) calculations based on the developed theory for Hg$^+$ in Hg vapor; (5) experimental data [34, 36]; and (6) calculations based on the developed theory for Ar$^+$ in Ar.

Figure 3 shows the results of comparison of the IDFs for $\rho = 0$ and $\frac{\beta}{\alpha_0} = 100$ for He$^+$ ions at room atomic temperature taking into account the dependence of the charge-exchange cross section on the velocity and disregarding this dependence. It can be seen that these functions are slightly different in
the region of the low temperature extremum in view of the difference in the cross sections at such moderate velocities.

To verify our results, we calculated the drift velocity of the ions in the parent inert gas plasma and in mercury vapor using the IDF obtained theoretically. The experimental data and the results of calculation of distribution function (5) for the drift velocity of He⁺ ions in He, Hg⁺ in Hg vapor, and Ar⁺ in Ar [34–36] are compared in Fig. 4. In the calculation of this velocity, we assumed that expression (6) obtained for the concentration is correct, strictly speaking, only for \( \frac{a_0}{\beta} \to 0 \). Therefore, we have used the normalization factor \( \int_0^\infty F_{ie}(\varepsilon_1) d\varepsilon_1 \) in these calculations and everywhere below. The calculations were performed taking into account the dependence of the charge-exchange cross section on the velocity. The data on the cross sections for He⁺ were borrowed from [21, 22], for Hg⁺ from [22], and for Ar⁺ from [36]. The references to experimental data are given in the figure. It can be seen that the correlation between the measured and calculated drift velocities is satisfactory, which confirms the correctness of the defined formulas once again. It should be noted that in spite of the fact that expressions (6) and (6b) were derived under the assumption that inequality (2b) holds, the results of comparison lead to the conclusion that the given theory can be used if the parameter appearing on the left-hand side of inequality (2b) exceeds 3. Figure 5 shows the dependence of the resultant ion distribution function (5) on the azimuthal angle \( \theta \) for \( \frac{a_0}{\beta} = 0.01 \) (for example, in He this corresponds to \( E \approx 300 \frac{V}{\text{cm Torr}} \)) for different relative velocities \( x = v_i \sqrt{\beta} = 0.1 \) and 5 in the absence (\( \varepsilon = 0 \)) and presence (\( \varepsilon = 1 \)) of the ambipolar field. It is interesting to note that for \( x = 0.1 \), the distribution is close to isotropic in spite of the strong field and the presence of the ambipolar field. This is due to the fact that the group of ions with velocities \( x \ll 1 \) is formed by ions that have been generated as a result of charge exchange, that have the Maxwellian (and isotropic) distribution, and have not been accelerated by the electric field. With increasing relative velocity, the angular velocity distribution for ions becomes more and more extended in the direction of the field (see Fig. 5). The results represented in this figure also show that in the presence of the ambipolar field, the peak of the angular distribution function deviates from the discharge axis (\( \theta = 0 \)); for example, for parameter \( \varepsilon = 1 \) (ambipolar field is equal to the axial field), the peak corresponds to angle \( \theta = \frac{\pi}{4} \).

As noted above, apart from theoretical calculation of the IDF in the parent gas in strong fields, we also measured it in mercury vapor at a low pressure (of the order of \( 10^{-3} \) Torr) for the parameter \( E \approx 400 \frac{V}{\text{cm Torr}} \) using a planar one-sided probe [27, 28]. The second derivative of the probe current was recorded using the demodulation method [28]. It is well known that the instrument function in this method has the form [30]:

\[
A(z) = \frac{2}{\pi} \int_{|u|}^{1} \left[ \frac{u^2 - z^2}{u} (1 - u) \right]^{0.5} du \text{ for } |z| < 2\sqrt{z}
\]

\[
A(z) = 0 \text{ for } |z| > 2\sqrt{z},
\]

(12)

where \( z = \frac{\sqrt{\varepsilon(\varepsilon - \varepsilon)}}{\Delta \varepsilon} \); and the amplitude of the differentiating signal is \( \Delta \varepsilon \).
Figure 5. Angular velocity distribution function for ions at $\frac{\alpha}{\beta} = 0.01$; (1) $x = 0.1$, $\varepsilon = 0$; (2) $x = 5$, $\varepsilon = 0$; (3) $x = 0.1$, $\varepsilon = 1$; and (4) $x = 5$, $\varepsilon = 1$.

Figure 6. Comparison of the IDF (normalized to unity) for Hg$^+$ ions in Hg vapor, calculated by formula (6) and measured using the method developed in [29] with a planar one-sided probe for different values of the differentiating signal of the probe method: (1) $F_{\text{calc}}(\varepsilon)$; (2) calculation of $F_{\text{calc}}(\varepsilon), \Delta \varepsilon = 0.1$; (3) experimental $F_{\text{exp}}(\varepsilon), \Delta \varepsilon = 0.1$; (4) calculated $F_{\text{exp}}(\varepsilon), \Delta \varepsilon = 0.2$; (5) experimental $F_{\text{exp}}(\varepsilon), \Delta \varepsilon = 0.2$; (6) calculated $F_{\text{exp}}(\varepsilon), \Delta \varepsilon = 0.05$; (7) experimental $F_{\text{exp}}(\varepsilon), \Delta \varepsilon = 0.05$. Current density $j = 100$ mA/cm$^2$, pressure $P = 10^{-3}$ Torr, parameter $E/P = 400$ V/(cm Torr), and atomic temperature $T_a = 410$ K.

Figure 6 shows the experimental data and the results of corresponding calculations of the convolution of the IDF normalized to unity (6b) and the above instrument function $A(z)$ for various differentiating signals. It can be seen that the measured and calculated dependences are in good agreement.

Figures 7 and 8 show the calculated and measured energy dependences of the coefficients of the IDF expansion in Legendre polynomials of degrees 0– 3 and 4–6, respectively, for the conditions of Fig. 6. It should be noted that the calculations were performed without fitting parameters using the measured value of parameter $\frac{E}{p} = 400$ V/cm Torr and the atomic temperature (equal to 410 K) determined earlier from the coincidence of the calculated and experimental energy distribution functions. It can be seen that good agreements with the results of calculations take place.

Figures 9–11 show the angular dependences for various energies of the calculated IDF normalized to unity, of the calculated sum of the first seven terms of the IDF expansion in the Legendre polynomials, and the same sum, but using the experimentally determined expansion coefficients for the Hg$^+$ ions under the same conditions as before. It can be seen that for the smallest anisotropy of the distribution function, which corresponds to the minimal energies at 0.05 eV, the coincidence of all three function is very good. Bearing in mind that anisotropy decreases with energy, we can state that for energies lower than 0.05 eV, the coincidence will also be good. With increasing energy (i.e., with increasing anisotropy), the discrepancy is observed between the distribution functions calculated from
the first seven Legendre polynomials and the exact function, which is quite natural for such a high
g value of $\frac{E}{p} = 400 \frac{V}{cm \text{Torr}}$.

**Figure 7.** Dependence of the first four Legendre coefficients in the IDF expansion over the directions of their motion under the same discharge conditions as in Fig. 6. The instrument function width is $\Delta \varepsilon = 0.05$ eV; experiment: (1) $n = 0$, (2) $n = 1$, (3) $n = 2$, (4) $n = 3$; theory: (5) $n = 0$, (6) $n = 1$, (7) $n = 2$, and (8) $n = 3$.

**Figure 8.** The same as in Fig. 7 for Legendre coefficients 4–6: experiment: (1) $n = 4$, (2) $n = 5$, (3) $n = 6$; theory: (4) $n = 4$, (5) $n = 5$, and (6) $n = 6$.

**Figure 9.** Comparison of angular dependence of the calculated IDF (formula (6)), calculated sum of the first seven terms of the DF expansion in Legendre polynomials, and the same sum determined from measurements for ion energy $\varepsilon = 0.05$ eV; the instrument function width in the probe method is $\Delta \varepsilon = 0.05$ eV. The discharge conditions are the same as in Fig. 6: (1) experimental reconstruction using the coefficients of the DF expansion in Legendre polynomials, $n = 0, \ldots, 6$; (2) calculated sum of the first seven terms of the DF expansion in Legendre polynomials; and (3) calculated DF.
Figure 10. The same as in Fig. 9 for $\varepsilon = 0.2$ eV.

Figure 11. The same as in Fig. 9 for $\varepsilon = 0.5$ eV.

It should be noted that for the plasma of inert gases, the simultaneous fulfillment of inequalities (2b) and (4a) required for the validity of the theory formulated above is ruled out in view of a low ratio $E/p$ for a small role of ambipolar diffusion, a high resonant charge exchange cross section, and a relatively small cross section of elastic scattering from a neutral atom [24]. Thus, to solve the problem of determination of the ion VDF in a glow discharge of inert gases in strong fields outside the discharge symmetry axis, the ambipolar field should be taken into account.

5. Conclusions

The velocity distribution of ions during their motion in the parent gas under the condition that the velocity acquired by an ion over the mean free path is higher than the average velocity of thermal motion of atoms is described by a function differing substantially from the equilibrium Maxwellian distribution. The average and the square mean velocities of ions in strong fields are determined by the energy acquired by an ion over its mean free path, while the most probable velocity is close to the most probable velocity of neutral atoms. For this reason, the representation of the velocity IDF as an equilibrium function with a temperature differing from the temperature of neutral atoms is a quite rough approximation.

The presence of the ambipolar field comparable with the axial field considerably distorts the ion velocity distribution function.

It was measured the energy dependence of the Legendre components of IDF $F^{n}_{la}$ and it was analyzed the accuracy of the measurements. The theoretical and experimental values of $F^{n}_{la}$ have been compared. It was obtained a good coincidence of experimental and theoretical results. In the positive column of the electric discharge, the concentrations of electrons and ions are almost identical; therefore, comparison of concentrations obtained from the processing of the experimental curve $I_U$ is also one of criteria of the validity of the above interpretation. The difference in the concentrations in our study did not exceed 10%, which is a satisfactory results on account of the error in probe measurements.

Good coincidence of the calculated and measured IDFs leads to the conclusion that the DF of the Hg+ ion under the given conditions is formed as a result of resonant charge exchange, and elastic collisions do not affect its form. It should be noted that, for example, elastic collisions in He plasma in strong fields should be taken into account in the calculation of the DF for the He+ ions [37].

Thus, the probe method developed here makes it possible to reliably determine angular harmonics of IDF $F^{n}_{ia}$, which considerably expands its potentialities. It was measured the energy dependence of
the harmonics of the ion distribution function $F_{0}^{a} - F_{1a}^{b}$. New possibilities of application of the probe method in determining IDF and the convective velocity of ions in a plasma have been illustrated.

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