Fundamental Nonambipolarity of Electron Fluxes in 2D Plasmas

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It is demonstrated that in two-dimensional plasmas there is in general a vortex component of the electron motion, which means that electron and ion fluxes do not satisfy the ambipolarity condition.

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The general balance equation for density of a single-component plasma was formulated by Schottky in 1924 by way of example of a longitudinally homogeneous positive column of a glow discharge in a tube [1].

The fundamental balance equations are

$$\frac{\partial n_{e,i}}{\partial t} + \nabla \cdot \mathbf{\Gamma}_{e,i} = S, \tag{1}$$

where fluxes $\mathbf{\Gamma}_{e,i}$ (subscripts e, i indicate electrons and ions, respectively) are assumed to be of the drift-diffusion form

$$\mathbf{\Gamma}_{e,i} = -\nabla(D_{e,i}n_{e,i}) \pm \mu_{e,i}n_{e,i}\nabla\varphi, \tag{2}$$

where n is density, D is a diffusion coefficient, μ is mobility, φ is an electric potential, and S is the volume generation rate. Note that expressions (2) account for thermodiffusion. The drift-diffusion treatment of transport is valid when the mean free path of electrons is less than the discharge size.

The difference between two equations (1) for ions and electrons, given plasma quasineutrality $n_e = n_i = n$, produces the continuity equation for the current density

$$\nabla \cdot \mathbf{j} = \nabla \cdot e(\mathbf{\Gamma}_i - \mathbf{\Gamma}_e) = 0. \tag{3}$$

The substitution of both expressions (2) for fluxes in $\mathbf{j} = e(\mathbf{\Gamma}_i - \mathbf{\Gamma}_e)$ gives the expression for the electric field [2]:

$$-\nabla\varphi = -\frac{\nabla((D_e - D_i)n)}{(\mu_e + \mu_i)n} + \frac{\mathbf{j}}{e(\mu_e + \mu_i)n} = \mathbf{E}_d + \mathbf{E}_c. \tag{4}$$

The first term $\mathbf{E}_d$ in (4) is the ambipolar field determined by the gradient of plasma density. It could be obtained from a simpler than (3) ambipolarity condition

$$\mathbf{\Gamma}_e = \mathbf{\Gamma}_i, \tag{5}$$

as it is usually done in textbooks on plasma physics [3]. The second term $\mathbf{E}_c$ is due to electric current. Thus, in a current-free plasma $\mathbf{E}_c$ vanishes. In this case the field $-\nabla\varphi$ is ambipolar. In a 1D case the only solution to (3) is a constant $\mathbf{j}$ ensured by an external source. A multidimensional case is more complicated, because (3) no longer means that $\mathbf{j}$ is a constant.

In the special investigation of a 2D inductively coupled plasma (ICP) discharge in argon [4,5], it was shown that in case of conductive walls, vortex electron currents exist in plasma, closing on conductive boundaries. For nonconductive walls, on which the fluxes of electrons and ions are locally equal, it was concluded in [4,5] that condition (5) is valid throughout the 2D discharge volume.

However, a more careful analysis shows that while in a 1D case equality (5) for fluxes in the whole volume follows from the same equality on a nonconductive boundary, the extrapolation of this fact to a 2D ICP discharge with dielectric walls, allowed in [4,5], requires a proof. Thus, in the analysis of fluid strata in [6] it was noted that in 2D plasma with nonuniform profiles of n and T_e significant radial electronic currents may exist that are not subject to (5).

In this work it is shown that it is already in a two-dimensional case that the spatial nonuniformity of n and T_e profiles necessarily leads to the presence of a vortex component in electron current, and hence the fluxes of charged particles do not satisfy ambipolarity condition (5). Furthermore, in this case an ambipolar field $\mathbf{E}_d$ is not a potential field.

To prove such important propositions it is enough to consider the simplest case of a single-component plasma with constant mobilities μ_e, μ_i in a discharge volume bounded by dielectric walls, because any complication (conducting walls, nonuniformity of mobility profiles, multicomponent plasma, presence of magnetic field, etc.) will only aggravate the violation of ambipolarity condition (5).

Further, several propositions of a more mathematical nature are used with the following conventions: Ω is the plasma domain (assumed to be a closed set), $\partial\Omega$ is its boundary, and $\mathbf{k}$ is the outer normal vector to $\partial\Omega$. We assume that particle fluxes are described by drift-diffusion equations (2), D_e, μ_e are functions of T_e, D_i, and μ_i are constants. All quantities are assumed to be smoothly varying.

First let us prove that if ∇n and ∇T_e are parallel throughout Ω, i.e., $\nabla T_e \times \nabla n = 0$, and the normal component of the current density vanishes at the boundary $\partial\Omega$: $\mathbf{j} \cdot \mathbf{k}|_{\partial\Omega} = 0$, then $\mathbf{j} = 0$ throughout Ω.

Since ∇n and ∇T_e are parallel, the values of T_e on the contours of n, which are always normal to its gradient, are constant. Thus T_e is a single-valued function of n: $T_e = g(n)$. In fact this statement follows from a general theorem on functional dependence (see [7]). The current density $\mathbf{j}$ can be written in the form

$$\mathbf{j} = -e[-\nabla((D_e - D_i)n) + (\mu_e + \mu_i)n\nabla\varphi] = -e(\mu_e + \mu_i)n\left(-\frac{D_e - D_i}{\mu_e + \mu_i}\nabla \ln((D_e - D_i)n) + \nabla\varphi\right). \quad (6)$$

Further, using the fact that D_e is a function of T_e, and hence a function of n, the expression for $\mathbf{j}$ can be written as

$$\mathbf{j} = -e(\mu_e + \mu_i)n\left(u(n)\frac{dv(n)}{dn}\nabla n + \nabla\varphi\right), \quad (7)$$

where u and v are functions of the single argument n. The exact form of these equations is not important, but assuming $w(n) = \int_0^n u(\xi)v'(\xi)d\xi$, we obtain

$$\mathbf{j} = -e(\mu_e + \mu_i)n\nabla(w(n) + \varphi) = -p\nabla\psi, \quad (8)$$

where $p = e(\mu_e + \mu_i)n$, $\psi = (w(n) + \varphi)$. Equation (3) and the boundary condition of $\mathbf{j}$ vanishing there then take the form

$$\nabla\cdot(p\nabla\psi) = 0, \qquad \mathbf{k}\cdot(p\nabla\psi)|_{\partial\Omega} = 0. \quad (9)$$

Now in the theory of differential equations [8], if $p > 0$ anywhere in Ω, then a solution to the boundary value problem (9) is a constant $\psi = \psi_0$. In other words, for sufficiently smooth p and ψ it follows from (8) that $\psi = \psi_0 = \text{const}$, and hence $\mathbf{j} = 0$.

Thus, the ambipolarity of fluxes (5) is fulfilled if $\nabla T_e \times \nabla n = 0$. Furthermore, in special cases, where D_e and μ_e are constants, or $(D_e - D_i)/(\mu_e + \mu_i)$ is a constant, $\mathbf{j} = 0$, as follows from (8).

Conversely, if $\nabla T_e \times \nabla n \neq 0$ at some point x_0 of Ω, and $d[(D_e - D_i)/(\mu_e + \mu_i)]/dT_e \neq 0$ throughout Ω, then there exists a point at which $\mathbf{j} \neq 0$.

If we suppose that $\mathbf{j} = 0$ everywhere, then

$$u(T_e)\nabla T_e + v(T_e)\nabla(\ln n) = \nabla\varphi, \quad (10)$$

where

$$D_e^T = \frac{dD_e(T_e)}{dT_e}, \qquad u = \frac{D_e^T}{\mu_e + \mu_i}, \qquad v = \frac{D_e - D_i}{\mu_e + \mu_i}. \quad (11)$$

Taking the curl of both sides of (10) and remembering that the curl of a gradient $= 0$, (10) becomes

$$(\nabla u) \times \nabla T_e + (\nabla v) \times \nabla(\ln n) = 0. \quad (12)$$

Since both u and v depend only on T_e, (12) can be rewritten as

$$\nabla T_e \times \nabla n = 0, \quad (13)$$

and (13) must be satisfied everywhere in Ω, which contradicts the original proposition that $\nabla T_e \times \nabla n \neq 0$ at a point x_0. Hence there is a point at which $\mathbf{j} \neq 0$.

In turn, if in Ω there are points where $\mathbf{j} \neq 0$, then there are also points where $\nabla \times \mathbf{j} \neq 0$. Indeed, the normal component of the current has to vanish on $\partial\Omega$, as the current cannot flow through the dielectric. Then from the assumption that $\nabla \times \mathbf{j} = 0$ throughout Ω, it follows that $\mathbf{j} = \nabla\psi$ for some potential ψ. Then from Eq. (3) $\Delta\psi = 0$ throughout Ω, and, as stated above, $\mathbf{j}\cdot\mathbf{k} = \nabla\psi\cdot\mathbf{k} = 0$ at the boundary $\partial\Omega$. Then any constant A is a solution to this boundary value problem. Since the solution of a Neumann boundary problem for a Laplace equation is unique up to constant [8], all possible solutions have the form of $\psi = A$. Hence $\mathbf{j} = \nabla A = 0$ throughout Ω. But this contradicts the assumption made. Consequently, there is a point in Ω where $\nabla \times \mathbf{j} \neq 0$. It means that $\mathbf{j}$ is a vortex current.

Thus, our analysis demonstrates that even in an unmagnetized 2D plasma with spatially nonuniform profiles of density and electron temperature the current density is not zero. It means that (5) is violated. Moreover, the current has a vortex nature even in case of a potential electric field $-\nabla\varphi$.

The $\mathbf{E}_c$ term in Eq. (4) is determined by electric current $\mathbf{j}$. Therefore, in the case of a vortex current, $\mathbf{E}_c$ also has a vortex nature (because the directions of $\mathbf{j}$ and $\mathbf{E}_c$ are the same and $\mathbf{j}$ has closed streamlines). Hence, to ensure the potential character of the total electric field $-\nabla\varphi$, the ambipolar component $\mathbf{E}_d$ of the field has to be vortex too. Therefore, the ambipolar electric field in this case is not a potential field.

It should be noted that in the case of constant mobilities μ_e, μ_i, the sum of Eq. (1) for electrons and ions, multiplied by $\mu_i/(\mu_e + \mu_i)$ and $\mu_e/(\mu_e + \mu_i)$, respectively, gives the equation of ambipolar diffusion

$$\frac{\partial n}{\partial t} - \Delta(D_a n) = S, \quad (14)$$

where $D_a = (D_e\mu_i + D_i\mu_e)/(\mu_e + \mu_i)$ is the ambipolar diffusion coefficient. In this case the equation of ambipolar diffusion is valid in spite of the violation of ambipolarity condition (5). The amazing fact that at constant mobilities Eq. (14) does not depend on a current flowing through plasma was mentioned in [2,6].

Likewise, while the fluxes of electrons and ions are not equal, the ion flux in a fluid model is approximately equal to the ambipolar flux. Indeed, within the drift-diffusion approximation the electron flux (2) is much smaller than each of its components given by drift and by diffusion. This means that $\nabla\varphi \approx \nabla(D_e n)/(\mu_e n)$, and substituting this into (2) we obtain for the ion flux

$$\mathbf{\Gamma}_i \approx -D_a\nabla n - \frac{\mu_i}{\mu_e}D_e^T n\nabla T_e, \quad (15)$$

where D_e^T is defined by (11) and the fact that $\mu_e \gg \mu_i$ has been taken into account.

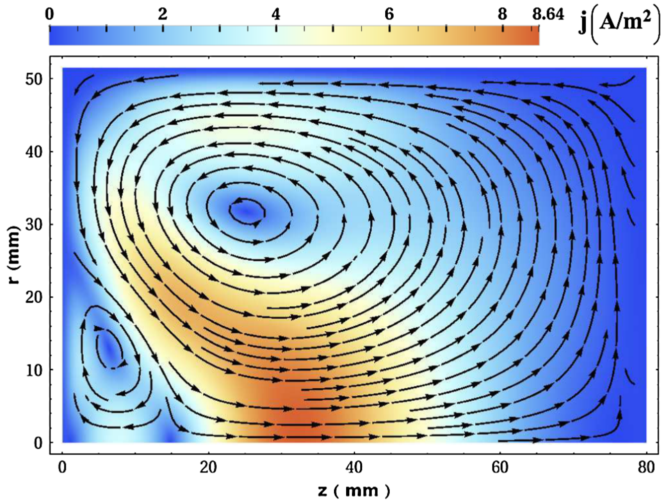

FIG. 1 (color online). A composite plot of the total electric current density. The colors show the absolute value of the flux. The current directions are shown by streamlines with arrows. Streamlines are closed, indicating that the current is a vortex one.

In the case of constant mobilities, and when the Einstein relation for electrons $D_e = T_e \mu_e$ holds, $D_e^T = \mu_e$ and (15) takes a simpler form

$$\mathbf{\Gamma}_i \approx -D_a \nabla n - \mu_i n \nabla T_e \approx -\mu_i \nabla (n T_e). \tag{16}$$

Since the gradient of the electron temperature in plasmas is generally small in comparison with the density gradient [6], the direction of the ion flux for the case of $\mu_e = \text{const}$ is almost perpendicular to the contour of plasma density. This remains true in a more general case when μ_e is a function of T_e, as the second term in (16) is small in comparison with the first one. This is confirmed by results of numerical simulations.

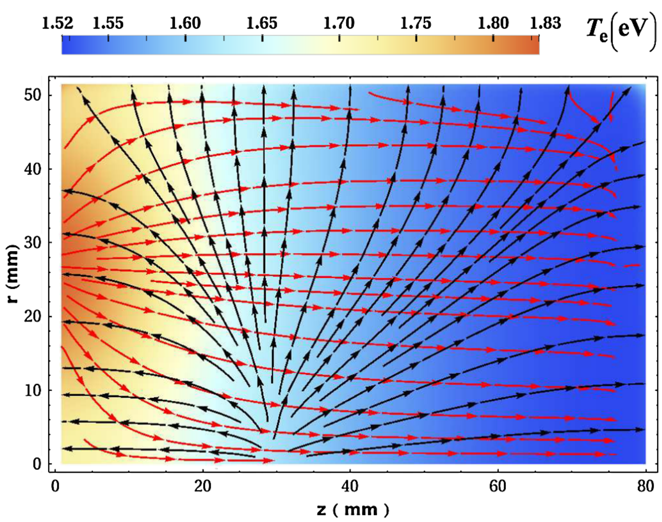

FIG. 2 (color online). A composite plot. The color indicates electron temperature values. Black and gray (red) streamlines show the directions of ∇n and ∇T_e, respectively.

The simulations were carried out for an ICP discharge in argon, in a cylindrical chamber of length $L = 8$ cm and radius $R = 5.15$ cm, with dielectric walls. Inductive coils are placed at one end of the cylinder (on the left in the figures that follow). The simulations employed a fluid model. A self-consistent system of equations for electron and ion densities and electron energy together with Poisson's equation were used.

A detailed description of the system of equations and the boundary conditions has been given in [9]. For simplicity, the volume processes took into account only direct ionization for argon by convolving a Maxwellian distribution for given T_e with the electron impact cross section. μ_e was calculated using the momentum transfer cross section, and D_e was determined by the Einstein relation.

The simulations were carried out for conditions where the gas pressure p was 200 mTorr and the absorbed in plasma power was 1 W. Quasineutrality $n_e = n_i$ was found to hold to a high degree of accuracy. Figure 1 shows that the streamlines are closed and the electric current has a vortex nature. Figure 2 shows the spatial distribution of electron temperature T_e and the directions of the gradients of density and temperature by black and gray (red) streamlines, respectively. It can be seen that in general these vectors are not parallel.

Additionally, simulations were performed for the same conditions as the previous ones but with the constant electron diffusion coefficient. The value chosen was the one obtained by averaging over the volume from the first simulation. In this case we could confirm that $\mathbf{j} = 0$; therefore, a streamline plot, such as the one given in Fig. 1, cannot be given here; in this case $\mathbf{\Gamma}_e = \mathbf{\Gamma}_i = -D_a \nabla n$. The profiles of n, T_e, and φ in both simulations are almost the same.

Thus, to show the differences, the profiles of the electric current density and its electronic and ionic components are represented in Fig. 3. It can be seen that the electronic component significantly exceeds the ionic one. Our results show that the ratio of total current to the ambipolar one increases with pressure, when T_e becomes more

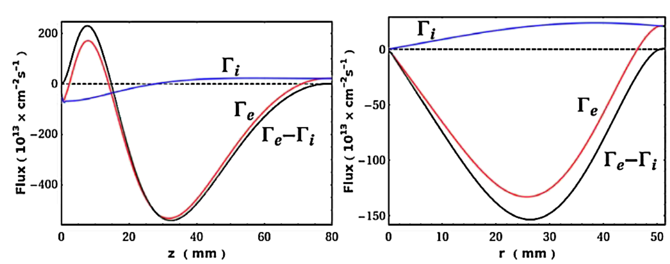

FIG. 3 (color online). Profiles of the electron flux [light gray (red)] and the ion flux [dark gray (blue)] axially (left) and radially at $L/2$ (right), showing their difference which is proportional to the current density. The axial components of fluxes are shown on the first plot and the radial components on the second one.

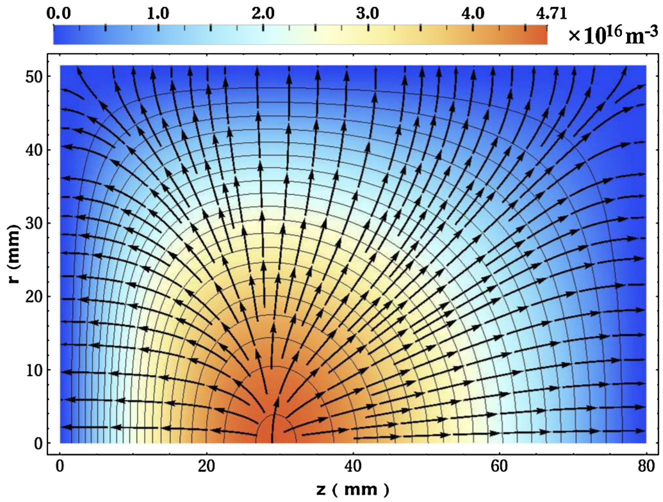

FIG. 4 (color online). A composite plot. The plasma density n is shown in color. Thick lines with arrows are the ion streamlines.

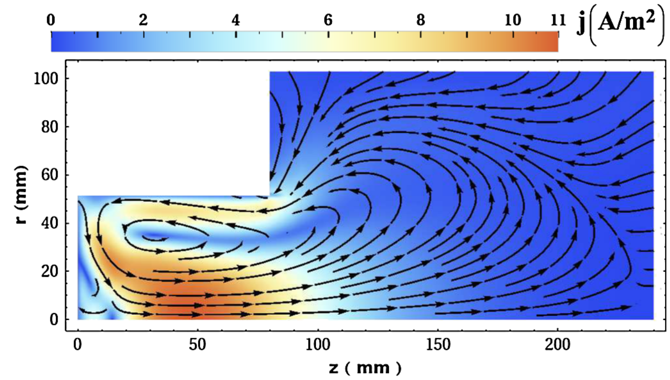

FIG. 5 (color online). A composite plot of the total electric current density for a two-chamber ICP discharge in oxygen with metal walls. The colors show the absolute value of the flux. The direction of the current is shown by streamlines with arrows.

nonuniform. Figure 4 gives the contours of plasma density and the directions of ion flux, indicated by streamlines with arrows. It shows that the ion flux is almost normal to plasma density contours, in agreement with (16). Thus, even for an unmagnetized single ion species plasma bounded by dielectric walls in 2D cylindrical geometry, (5) is violated and a vortex electric current occurs. Since other complications, such as conducting walls and the presence of a magnetic field, will only further constrain validity, it is possible to state that the classical concept of ambipolar diffusion is not generally valid in two dimensions. That is, it is not a paradigm for the transport of charged particles in plasmas, and we conclude that the case of a longitudinally uniform positive column of a dc discharge is a special case [10].

We also made simulations for a more complicated case, which is also more interesting for practical applications (and especially for plasma processing). Namely, we made the calculation of an ICP discharge in oxygen (with pressure $p = 20$ mTorr, the power absorbed in plasma $=$ 25 W) in a discharge chamber with metal walls. The discharge volume has more complex geometry and consists of two chambers with the small chamber dimensions $L =$ 8 cm, $R = 5.15$ cm and $L_1 = 16$ cm, $R_1 = 10.3$ cm for the large ballast volume (see Fig. 5). rf coils are at the left side of the chamber. The system of equations is similar to that used for argon, but it accounts for two kinds of ions (positive O_2^+ and negative O^-) and a more complex plasmachemistry model that includes processes of attachment, detachment, ionization, dissociation, and excitation of a singlet level of an oxygen molecule [11]. Figure 5 shows streamlines of total electric current density for this discharge. It can be seen that in this case there is also a vortex current with closed streamlines, but some streamlines start and finish at walls.

In conclusion we should note that the presence of a vortex component of current necessarily leads to additional Joule heating of the electron gas, which manifests itself in the formation of fluid strata [6]. Additionally, the inequality of electron and ion fluxes can lead to such a nontrivial phenomenon as arising of friction forces which may affect a neutral gas [12]. In particular, these forces may cause rotation of the gas and thus possibly cause magnetomechanical effects [13].

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