Ion–ion plasmas and double layer formation in weakly collisional electronegative discharges

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Plasmas of electronegative gases often separate into two distinct regions: an ion–ion core and an electron–ion periphery. Under certain conditions, a double layer may form at the boundary between the two regions. In weakly collisional three-component electronegative plasmas, formation of a double layer depends on the ratio of the electron to negative ion temperatures, and the ratio of the electron to positive ion densities. © 1998 American Institute of Physics. [S0003-6951(98)00606-8]

FIG. 1. Ion–ion and electron–ion plasmas separated by a double layer. The thickness of the double layer and the sheath is exaggerated for clarity.
Fig. 2. Function $Q(\Psi)$ for $\delta=20$ and different $\alpha$. A single maximum of $Q(\Psi)$ (curves 1 and 3) corresponds to no double layer formation, and the potential at the maximum, $\Psi_f$, corresponds to the sheath edge. Two maxima of $Q(\Psi)$ (curve 2) correspond to double layer formation. The potential at the boundary between the ion–ion plasma and the double layer is then $\Psi_f$, and that between the plasma and the sheath is $\Psi_s$ (see also Fig. 1). Stability of positive ions in the free-flight regime (no collisions) is determined by the spatial distributions of ionization rate $I(x)$ and potential $\Psi(x)$.\(^1\)

\[
p(x) = \frac{1}{v_{th}} \int_0^x \frac{d x'}{\sqrt{\Psi(x) - \Psi(x')}} = \frac{1}{\sqrt{\Psi(x) - \Psi(x')}}. \tag{2}
\]

where $v_{th} = (2kT_e/M)^{1/2}$ is the ion thermal velocity. The potential distribution in the plasma is found from the quasineutrality condition

\[
\int_0^x \frac{I(x') dx'}{\sqrt{\Psi(x) - \Psi(x')}} = \Gamma \left( 1 - \alpha \exp(-\Psi) + \alpha \exp(-\Psi/\delta) \right) \tag{3}
\]

Here $\alpha = n_+ / n_e$, $n_0 = n_{i0} + n_{e0}$ is the plasma density at the center, and $\Gamma = n_0 v_{th}$ is an ion flux. Assuming that $I = n_e \nu$, where $\nu$ is the ionization frequency (assumed spatially constant), the solution of (3) can be found in the form

\[
\frac{\pi \alpha \nu x}{v_{th}} = Q(\Psi). \tag{4}
\]

The function $Q(\Psi)$, shown in Fig. 2, is given by

\[
Q(\Psi) = \int_0^\Psi F(\Psi') \exp(\Psi'/\delta) d\Psi'. \tag{5}
\]

where

\[
F(\Psi) = \Psi^{-1/2} - 2(1 - \alpha)D(\sqrt{\Psi}) - 2\alpha \delta^{-1/2}D(\sqrt{\Psi}/\delta), \tag{6}
\]

and $D(z)$ is the Dawson integral.\(^7\) The spatial distribution of $\Psi(x)$ found from (4) has a singularity $d\Psi/dx = \infty$ (an infinitely high field) where $Q(\Psi)$ has a maximum. The quasineutrality condition is violated at these points. The maxima of $Q(\Psi)$ correspond to roots of $F(\Psi)$. These roots are independent of the shape of the ionization rate and are determined solely by $\alpha$ and $\delta$.\(^1\)

Setting $F(\Psi)=0$ gives

\[
\alpha = \frac{1/2x-D(z)}{D\left(z/\sqrt{\delta}\right)/\sqrt{\delta}-D(z)}, \tag{7}
\]

where $z = \sqrt{\Psi}$. For a given $\delta$, Eq. (7) defines the potential $\Psi(\alpha)$ at the points where quasineutrality is violated. For $\delta < 10.8$, $\Psi(\alpha)$ has a single value for each $\alpha$ which corresponds to the plasma-sheath boundary, $\Psi_s$. In this case, the potential drop in the plasma changes continuously from 0.855 to 0.855 $\delta$ with an increase of $\alpha$ (curve 1 in Fig. 3). For $\delta > 10.8$, there is an inflection point of $\Psi(\alpha)$ at $\alpha = 0.33$ (curve 2). For $\delta > 10.8$, there are three different values of $\Psi(\alpha)$ in a range $\alpha_0 < \alpha < \alpha_1$ and a single value for $\alpha = \alpha_0$ or $\alpha > \alpha_1$ (curves 3 and 4). In the range $\alpha_0 < \alpha < \alpha_1$, the potential in the plasma changes discontinuously with changing $\alpha$.

The case of three different roots of $F(\Psi)$ corresponds to the formation of a double layer in the plasma.\(^1\) In the quasineutral model, the double layer is formed where $d\Psi/dx = \infty$. The spatial location of the double layer ($x_0$ in Fig. 1) depends on the specific ionization mechanism whereas the values of $\Psi_f$ or $\Psi_s$ do not. Equation (3) is not valid in the double layer and the potential profile is to be found from Poisson’s equation. The potential drop in the layer, $\Psi_f - \Psi_s$, can be derived from

\[
\int_{\Psi_f}^{\Psi_s} d\Psi' (p - n) = 0. \tag{8}
\]

At $\Psi > \Psi_f$, in the electron–ion plasma, the quasineutrality condition is valid again, and the potential profile is given by Eq. (3). To estimate the maximum potential drop in the double layer, we equalize the density of positive ions created at $x < x_0$ and accelerated in the layer to the electron density at the boundary of the electron–ion plasma where $\Psi = \Psi_f$. For $\alpha = 1$, when $\Psi_f = 0.855$, this gives for $\Psi_f$

\[
0.344/\sqrt{\Psi_f} = \alpha \exp(-\Psi_f/\delta). \tag{9}
\]

According to (9), the maximum value of $\Psi_f = \delta/2$ is reached at $\alpha = 0.94(\delta/2)^{-1/2}$. The potential drop in the double layer can therefore be of the order of the electron temperature.

Figure 4 shows the normalized electron and negative ion densities, and the potential profiles in the plasma for the three different cases shown in Fig. 2. At low $\alpha$ the potential...
drop in the plasma is fairly small and the ion–ion plasma extends up to the sheath edge. This is the case when \( Q(\Psi) \) has only one maximum and there is no double layer formed (Curve 1 in Fig. 2). With an increase of \( \alpha \), a double layer forms as an abrupt potential jump separating the ion-ion plasma from the electron–ion plasma. The potential at the plasma-sheath boundary, \( \Psi_s \), corresponds to the second maximum of \( Q(\Psi) \) whereas the first maximum corresponds to \( \Psi_f \) (see Figs. 1 and 2). The potential profile in the ion–ion plasma is rather flat (defined by ion temperature), so that the electron density and the ionization rate in this plasma must be almost uniform. At larger \( \alpha \) the double layer disappears and the potential drop in the plasma becomes of the order of electron temperature. This case corresponds to curve 3 in Fig. 2.

Finally, the wall potential \( \Psi_w \) is set up to equalize the electron production and loss rates. In low pressure discharges, \( \Psi_w \) must exceed the ionization potential of the atoms. Since the potential at the plasma sheath boundary \( \Psi_s \) is reduced by the presence of negative ions (see Fig. 4), the potential drop in the sheath becomes larger. The presence of a double layer modifies the energy distribution of positive ions escaping the plasma which then becomes double peaked. The lower energy peak corresponds to ions formed in the electron–ion plasma and accelerated by the sheath field. The higher energy peak corresponds to ions formed in the ion–ion plasma and accelerated by the double layer and the sheath fields.

Pulsed-power discharges: In pulsed-power discharges, the power sustaining the plasma is modulated (e.g., square-wave modulation) with a certain frequency and duty cycle. Some predictions on the temporal dynamics of ion–ion plasmas in pulsed-power discharges can be made based upon the steady-state solutions obtained above. In particular, two key quantities, \( \alpha \) and \( \delta \) define the spatial distribution of plasma parameters. Under favorable conditions, a double layer may form during the power-on fraction of the cycle. During the power-off fraction of the cycle, high energy electrons continue to escape to the wall, but the ionization is effectively switched off. While the decaying plasma is continually depleted of high energy electrons, both \( \delta \) and \( \alpha \) keep decreasing due to electron losses to the wall and attachment to molecules. At some point in the afterglow, the double layer must disappear as \( \delta < 10.8 \) or \( \alpha < \alpha_0 \) (see Fig. 3). Sufficiently late in the afterglow, the negative ion density begins to exceed the electron density in the entire discharge volume, and a virtually electron-free plasma is formed. At this time it becomes possible to extract negative ions out of the plasma.

An important difference exists between the properties of ion-ion plasmas in the collisional and near-collisionless regimes. Although collisional electronegative plasmas also tend to separate into ion-ion and electron–ion regions, quasineutrality remains throughout the plasma and the potential profile remains smooth. In our case, the quasineutrality is violated in the double layer which is formed within the plasma under certain conditions. This double layer has much in common with the double layer observed in a two-electron-temperature plasma. Such direct consequences of the double layer as an abrupt potential drop within the plasma and the appearance of two peaks in the energy distribution of positive ions extracted out of the plasma should be detectable experimentally.

In summary, an ion–ion plasma can coexist with an electron–ion plasma in the same vessel. The boundary between the two plasmas may be smooth or abrupt. In the latter case, a double layer separates the two plasmas. Conditions favoring the formation of such a double layer have been presented in this letter. Depending on conditions, potential oscillations corresponding to multiple double layers may also form. Relevant theories that have been developed for electronegative plasmas with bi-Maxwellian electron energy distributions can be applied directly to electronegative plasmas.

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