Anomalously High Near-Wall Sheath Potential Drop in a Plasma with Nonlocal Fast Electrons

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It is demonstrated for the first time that the presence of a small number of fast, nonlocal electrons can dramatically change the thickness of and electric field in the near-wall sheath. Even if the density of the nonlocal fast group, n_f, is much less than the density of the bulk electrons, n_b ($n_f \sim 10^{-5} n_b$), the near-wall potential can increase dramatically resulting in a comparable increase in the sheath thickness. Because of this low fractional density, the average energy (electron temperature T_e) of all electrons is little changed from that of the bulk, yet the near-wall potential drop can increase to tens of T_e/e. More importantly, due to the nonlocal nature of this group of electrons, the near-wall sheath potential is found to be independent of T_e and is determined only by the energy of the fast group.

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The principles of plasma-boundary sheath formation are important for understanding fundamental plasma properties [1] and for applications where the plasma is confined to a finite volume or flows around a rigid body. It is especially important for near-wall processes such as plasma technology and plasma processing [2]. However, despite efforts by numerous researchers (see, for example, a small sample of very recent works [3–6]), this problem is not well understood and some surprising effects have been observed [7].

In this Letter we discuss the sheath formation in plasmas with nonlocal fast electrons which, because of their low density, cannot significantly influence the average electron energy and ambipolar plasma fields but can have a substantial effect on the plasma sheath. It is shown here that these electrons can lead to a dramatic increase in the thickness and potential of the sheath in, for example, postdischarge, or similarly, plasmas with internal or external sources of fast electrons.

It is well known that electrons escape the plasma much faster than ions and charge the wall surface negatively. To maintain plasma quasineutrality, a layer of positive space charge in the vicinity of the boundary surface is formed, which leads to an equalizing of the electron and ion fluxes ($j_e = j_i$) to the surface. In a traditional local approximation, the electron density profile at retarding (negative) potential $\varphi(\vec{r}, t)$ and for a Maxwellian electron energy distribution function (EEDF) with electron temperature T_e takes the form

$$n_e(\vec{r}, t) = n_0 \exp[-e\varphi(\vec{r}, t)/T_e], \quad (1)$$

where e is the electron charge and n_0 corresponds to the plasma density at $\varphi = 0$. The electron flux to a surface is equal to

$$j_{eb} = j_{eT} \exp(-e\Delta\Phi/T_e), \quad (2)$$

where the chaotic flux at the plasma-sheath edge is $j_{eT} = n_s \bar{v}_e = n_s\sqrt{T_e/2\pi m}$, and m is the electron mass, n_s is the plasma density at the sheath edge, and $\Delta\Phi = (\varphi_w - \varphi_s)$ is the potential drop in the sheath (φ_w is the wall potential and φ_s is the sheath edge potential). Near the surface, electrons are repelled and the Boltzmann distribution (1) is invariant with respect to the choice of reference point, where $\varphi = 0$, and $n_e = n_0$. Therefore, in the above it is possible to replace n_s by n_0, so that $j_{eT} = n_0\sqrt{T_e/2\pi m}$, and to replace $\Delta\Phi$ by φ.

The ion flux depends on various parameters of the quasineutral plasma, is not significantly altered by the negative $\Delta\Phi$ value, and is referred to as the ion saturation current. This flux corresponds to the sum of the diffusion and conductivity fluxes which, for uniform T_e and T_i, equals the ambipolar flux

$$j_i = -D_i dn/dr + b_i E_a n = -D_a dn/dr, \quad (3)$$

where D_i is the ion diffusion coefficient, D_a is the ambipolar diffusion coefficient, b_i is the ion mobility, and E_a is the ambipolar electric field. The density gradient is evaluated in the region where the plasma is quasineutral and collisional, but close enough to the wall so that the particle fluxes are equal.

The size of the near-wall sheath is governed by the local Debye radius r_D (and may be many r_D [8]) and is collisionless at low pressure, i.e., thin with respect to ion mean free path λ_i. The separation into plasma and sheath is, to some extent, a matter of convention since at a distance of order λ_i from the plasma boundary, the charge separation (and the field strength) smoothly increases towards the surface. This region is often referred to as the presheath. The directed ion velocity u_i continuously increases towards the wall up to the ion sound speed c_s ($u_i = c_s$) at some point near the wall. For $r_D < \lambda_i$ the quasineutral approximation ($n_e = n_i$) becomes invalid near this point, the electric field diverges, the transition to the sheath occurs, and n_e decreases rapidly toward the wall. This point is usually adopted as the location of the plasma-sheath boundary and is normally derived from the Bohm criteria (see, for example, [2,8,9] for details), which for $T_e \gg T_i$ and ions of mass M has the simple form

$$u_i = \sqrt{T_e/M}. \tag{4}$$

For a Maxwellian EEDF, the potential drop in the near-wall sheath, which corresponds to the absence of a net current through the surface, can be found from (2) and (4) to be

$$|\Delta\Phi_b| = \frac{T_e}{2e}\ln\left(\frac{M}{2\pi m}\right) \approx 5\frac{T_e}{e} \qquad (M = 40). \tag{5}$$

In most weakly ionized plasmas, the EEDF is non-Maxwellian. Inelastic collisions cause the high energy tail of the EEDF to be depleted relative to a Maxwellian. The escape of fast electrons to the walls further depletes the EEDF tail. The latter process is efficient for distances less than the electron energy relaxation length λ_ε. At low pressure, when the plasma dimension L is less than λ_ε, the EEDF is nonlocal (see [10] for details), and the EEDF tail is depleted throughout the entire plasma volume. For example, in an atomic gas with an inelastic threshold ε^*, for electrons in the energy range $\varepsilon < \varepsilon^*$ (typically, the majority), $\lambda_\varepsilon = \lambda_e\sqrt{M/m} > 100\lambda_e$, where λ_e is the electron mean free path. Therefore the inequality $\lambda_\varepsilon \gg L$ holds up to relatively high pressures, $pL < 5 \sim 10$ torr cm. In the case of a nonlocal EEDF, the electron current density to the negatively biased wall, j_w, is transported only by the high energy part of electron population. This differs from the results of the fluid approach since the concept of mean directed velocity fails at distances less than the EEDF relaxation length λ_ε [8,10,11]. Electrons with total energy $\varepsilon = w + e\varphi(\vec{r})$ (kinetic plus potential energy in electric field) lower than $e\Delta\Phi$ are *trapped* and at these distances do not contribute to the electron current to the wall. The current in the vicinity of a surface is transported in the form of a free diffusive flux of fast, *untrapped* electrons with $\varepsilon > e\Delta\Phi$ at a constant value of ε. For a full description of the flux of untrapped electrons to the walls, a loss cone should be taken into account [12]. This mechanism cannot be taken into account by a fluid approach. Note that a somewhat similar phenomenon of self-trapping of negative ions, in a plasma containing an electronegative gas, has been described in Ref. [13].

The ratio of the density of trapped electrons ($n_{e<}$) to the density of free electrons ($n_{e>}$) can, in general, be estimated from the condition that the ambipolar flux of ions must equal the free diffusion flux of electrons ($n_i D_a = n_{e>} D_{e>}$). Since the number of higher energy free electrons is always small [$n_{e>} \approx \sqrt{T_e/T_i}\sqrt{m/M}(\lambda_i/\lambda_a)n_{e<} < 10^{-3}n_{e<}$], the addition of even a small number of fast electrons ($n_{ef} \approx \sqrt{T_e/\varepsilon_f}n_{e>}$) with energies $\varepsilon_f \gg T_e$ can dramatically change the characteristics of the plasma with a nonlocal EEDF. The last statement is the crux of this Letter and the results presented here differ markedly from those obtained within the framework of a fluid model. This includes fluid models with beam electrons [14] and for plasmas with a bi-Maxwellian EEDF [15].

Additional fast electrons can be created by various plasma-chemical volume processes with the involvement of long-living excited states of atoms and molecules, negative ions, photoionization, etc., or externally injected. Examples of volume processes are pooling reactions of metastable atoms and molecules which lead to ionization

$$A^* + A^* \rightarrow A^+ + A + e_f, \tag{6}$$

superelastic collisions of slow electrons with metastables

$$A^* + e \rightarrow A + e_f, \tag{7}$$

or associative detachment of electrons from negative ions

$$A^- + A \rightarrow A_2 + e_f. \tag{8}$$

The excess energy in reactions (6)–(8) can produce fast electrons, e_f, with energies that can greatly exceed the average electron energy. For example, for Ar metastables (with energy $\varepsilon^* = 11.55$ eV), the energy of fast electrons $\varepsilon_f \approx 7.3$ eV for (6) and $\varepsilon_f \approx 11.55$ eV for (7); for O_2 molecules $\varepsilon_f \approx 3.6$ eV for (8). At the same time, in an afterglow plasma T_e can be 0.1 eV.

Nonlocal fast electrons with $\varepsilon_f > e\varphi_b$ are produced in the volume by source terms ΣI_j from reactions (6)–(8) and are lost to the walls. In this case, the nonlocal EEDF of the fast electrons is narrowly distributed around the production energy. Their flux to the plasma boundary with area S can be found from their creation rate as

$$j_{ef} = \int_V \Sigma I_j dV/S. \tag{9}$$

If the source terms I_j are known, Eq. (9) allows one to readily calculate the flux j_{ef}. For example, for reaction (6), $I_b = \beta_b N_m^2$, where the rate coefficient $\beta_b \approx 10^{-9}$ cm^3/s, and N_m is the density of metastable atoms. In the presence of fast electrons, if $j_{ef} < j_i$, the zero current condition is of the form

$$j_i = j_{eb} + j_{ef}, \tag{10}$$

where j_{eb}, j_i, and j_{ef} are defined according to Eqs. (2), (3), and (9). Equation (10) determines the potential drop in the sheath as a function of the fast electron flux j_{ef}, giving

$$|\Delta\Phi| = |\Delta\Phi_b| + \frac{T_e}{e}\ln\frac{j_i}{j_i - j_{ef}}. \tag{11}$$

Therefore, the presence of j_{ef} causes the near-wall potential drop to increase, compared with the value from Eq. (5), which is calculated under the assumption of only bulk electrons, i.e., when j_{ef} is negligible. This increase may be large (many times $|\Delta\Phi_b|$), even if the density of fast electrons is much less than the density of bulk electrons (say, 10^{-5} times). When $j_{ef} > j_i$ in order to maintain quasineutrality, part of the $j_{ef} - j_i$ flux of fast electrons must be self-trapped, i.e., the walls will acquire a potential of the order of ε_f/e [16–18]. The near-wall potential drop becomes approximately equal to the energy of the fast group. The electron energy distributions from reactions (6)–(8) have widths of only a few tenths of an eV (due to the large λ_ε) [16,17]. Therefore the wall potential in this regime deviates from ε_f/e by only a few tenths of a volt and, for simplicity, we neglect this difference.

The presence of fast electrons should be taken into account for calculations of sheath thickness, potential, and electron and ion densities in the plasma, presheath, and sheath. When $\varepsilon_f n_f \ll n_b T_e$, the density of fast electrons in the quasineutral plasma and presheath is negligible, and need not be taken into account in most types of calculations. Therefore, the analysis of the plasma and presheath can be done as in Refs. [1,2] for a plasma with a Maxwellian EEDF. In this case the Bohm criterion will be the same as in the plasma without fast electrons (i.e., in a plasma with a Maxwellian EEDF with electron temperature T_e). If ($j_{ef} > j_i$), we cannot use the simple Boltzmann relationship for electron density and should use nonlocal plasma kinetics [8,10]. We can conclude that during the transition between the two regimes the sheath thickness changes from a thin sheath (a few r_D) to a thick sheath (a few tens of r_D).

To illustrate this point, Fig. 1 shows the calculated near-wall sheath thickness in an argon postdischarge afterglow plasma with electron temperature $T_e = 0.1$ eV, which is typical for the afterglow [19]. Here, we define the sheath sickness in accordance with Refs. [20,21] and use the formula from Ref. [21],

$$h_{sh} = [(\sqrt{2}/3)X^{3/2} + 2\sqrt{2}X^{1/2}]r_D, \qquad (12)$$

for the collisionless sheath, where $X = \sqrt{1 + 2|\Delta\Phi|/T_e} - 2$. In this calculation, only the fast electrons arising from reaction (6) have been taken into account. It can be seen that the transition between the two regimes (thin and thick sheath) occurs when $j_{ef} = j_i$ and increases rapidly with respect to that ratio. This sharp increase is connected to the near-complete absence of electrons within the energy interval from a few T_e (say, 0.5 eV) to about 7 eV.

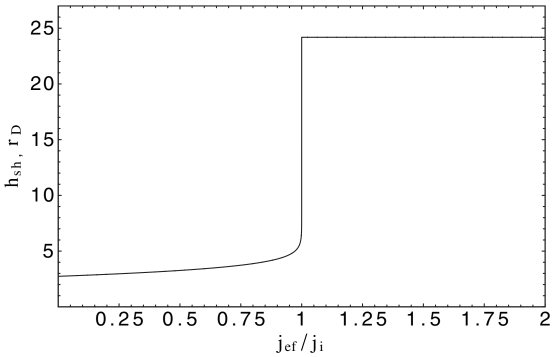

FIG. 1. The thickness of the near-wall sheath (in r_D units) calculated for argon afterglow plasma. $T_e = 0.1$ eV; $\varepsilon_f = 7.3$ eV.

In a steady state self-sustained glow discharge with typical T_e, the wall potential is greater than the fast electron energy, i.e., $e\Phi > \varepsilon_f$ [see Eq. (5)], so fast electrons created in reactions (6)–(8) are usually trapped in the volume and do not contribute to the wall flux. In contrast, in a postdischarge plasma T_e falls rapidly with time so the potential drop [Eq. (5)] also decreases rapidly and is small compared to the energy ε_f of created fast electrons (6)–(8). As the densities of long-lived excited states, which determine the sources I_j in (9), decrease slowly, at some time in the afterglow the flux (9) will equal the ambipolar ion flux (3). Equation (11) becomes invalid for $j_{ef} \geq f_i$, and, as shown above, the walls will acquire a potential of order of ε_f. When this occurs, the wall potential, φ_b, will increase from $|\varphi_{bT_e}| \sim 0.1$–$0.3$ V to the anomalously large $|\varphi_{bf}| = \varepsilon_f/e \sim 3$–$10$ V. The trapped electrons cannot escape to the wall and are cooled in the plasma volume by collisions with atoms and bulk electrons (heating the latter) forming a steplike EEDF in the low energy direction (see [16] for detail). The reason for this steplike EEDF can be explained as follows. This portion of the volume-generated fast electrons are trapped and cannot escape to the walls. They relax by electron-electron and elastic electron-atom collisions, e.g., by processes with small energy losses, creating a continuous electron spectra in the region $\varepsilon < \varepsilon_f$.

Thus, both of the above cases (low and high $|\Delta\Phi|$) can be realized in the process of plasma decay in the nonlocal regime: initially, $j_{ef} < j_i$ and $e\Delta\Phi \ll \varepsilon_f$, while at later times, when the bulk electrons cool to low temperatures, $j_{ef} > j_i$ and $e\Delta\Phi \approx \varepsilon_f$. As an example, Fig. 2 shows the results of the potential measurements, φ_w, in a xenon afterglow at a gas pressure of $p = 0.2$ torr, dc current in

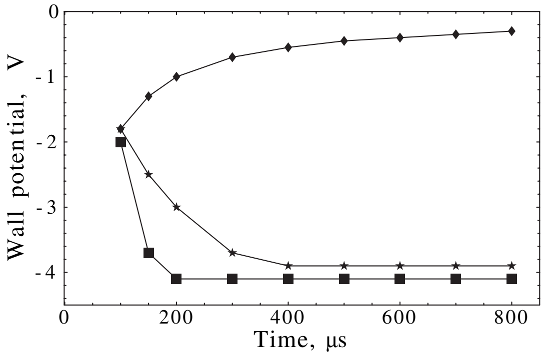

FIG. 2. Near-wall potential drop in Xe afterglow plasma. Pressure is 0.2 torr. Measurements (stars), calculation with formula (5) (diamonds) and calculations taking into account fast electrons, and the resulting anomalous potential jump (boxes).

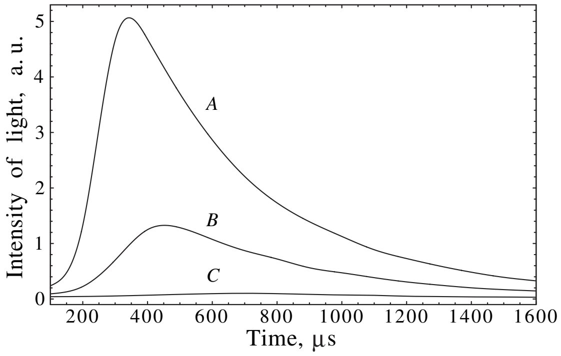

FIG. 3. Intensity of the Ar 420.1 nm spectral line in the afterglow. Pulse duration is 100 μs and repetition frequency is 500 Hz. Ar pressure is 15 mtorr (curve A), 20 mtorr (curve B), and 10 mtorr (curve C).

the pulse $i = 5$ mA, current duration 80 μs, and 1 kHz repetition rate in a glass tube of radius $R = 1.75$ cm. A molybdenum ring was inserted into a section of the tube which allowed measurements of the wall potential. A probe in the vicinity of the wall allowed measurements of the plasma potential (a second derivative method was used [22]). It could be clearly seen that a transition from a free diffusion regime ($j_{ef} < j_i$) to a regime with an anomalous large wall potential ($j_{ef} > j_i$) occurred over a period of time in the afterglow (sometime between 100 and 200 μs of the afterglow). Calculations using Eq. (5) do not work in this case, while calculations which take into account fast electrons are in good agreement with experiment [see Eq. (11) and the text below it].

Trapped fast electrons can lead to excitation from metastable levels and significantly increase the intensity of spectral line emission in the afterglow. These effects have been experimentally investigated in the postdischarge of a 100% power-modulated rf plasma in argon. The experimental apparatus has been previously described in detail [23] and the more recent plasma excitation experiment is described in [18]. To show the effect of a gradual increase in the wall potential, measurements of the time dependence of the plasma potential have been made for different gas pressures. The transition between regimes of free flight and partial trapping has been observed over a pressure range of 5 to 20 mtorr. During this transition, the wall potential increases from a few tenths of a volt to several volts, respectively. At the same time, as the pressure increases to 20 mTorr, a significant increase in the intensity of spectral lines is also observed. These correspond to transitions between the argon $3p^54p$ and $3p^54s$ levels, which is consistent with stepwise excitation from metastable levels (see Fig. 3). The presence of this emission, along with the observed increase in wall potential, clearly indicates the presence of fast electrons.

In summary, a small density of nonlocal fast electrons can result in a large change in the near-wall potential. This change is not predicted by local plasma models.

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