Double layer formation in the expanding region of an inductively coupled electronegative plasma

N. Plihon,[a] C. S. Corr, and P. Chabert
Laboratoire de Physique et Technologie des Plasmas, UMR 7648, Ecole Polytechnique, 91128 Palaiseau, France

Double-layers (DLs) were observed in the expanding region of an inductively coupled plasma with Ar–SF$_6$ gas mixtures. No DL was observed in pure argon or SF$_6$ fractions below a few percent. They exist over a wide range of power and pressure although they are only stable for a small window of electronegativity (typically between 8% and 13% of SF$_6$ at 1 mTorr), becoming unstable at higher electronegativity. They seem to be formed at the boundary between the source tube and the diffusion chamber and act as an internal boundary [the amplitude being roughly $1.5(kT_e/e)$] between a high electron density, high electron temperature, low electronegativity plasma upstream (in the source), and a low electron density, low electron temperature, high electronegativity plasma downstream. © *2005 American Institute of Physics.* [DOI: 10.1063/1.1869533]

Double layers (DLs) have been studied over the past decades theoretically, numerically and experimentally (see Ref. 1 and references therein). The largest part of the literature treats the case of electropositive plasmas, however, DLs were also found in electronegative plasmas.[2–5] More recently, Charles and co-workers[6,7] have observed a current-free DL in the expanding region of a helicon wave excited plasma at very low pressures (typically less than a mTorr). A strongly diverging static magnetic field seemed to be required in order to reach the conditions for DL formation. Their system also had an abrupt change in radius at the boundary between the source and the diffusion chambers, which could possibly be a source of DL formation.[8]

In this letter we show that a DL can be formed in a system that has a similar geometry to that of Charles and co-workers, but without the use of a static diverging magnetic field. However, this was possible only with a minimum percentage of an electronegative gas (namely SF$_6$) added to argon. Moreover, this electronegative DL was mostly unstable. It was found to be stable for a very narrow range of SF$_6$ mixtures.

The reactor is shown schematically in Fig. 1. It was originally designed to operate in the helicon regime. For the work presented in this letter, the system was operated without a static magnetic field, i.e., in the inductive mode. The reactor consists of a source chamber sitting on top of a 32 cm diameter, 26 cm long aluminum diffusion chamber. Hence, there is an expanding plasma underneath the source region. The source is a 15 cm outer diameter, 30 cm long and 0.9 cm thick pyrex cylinder surrounded by a double saddle field type helicon antenna.[9] The fan-cooled antenna is powered through a close-coupled L-type matching network by an rf power supply operating at 13.56 MHz and capable of delivering up to 2 kW forward power. The input power was recorded as the difference between the forward and reflected powers. The pyrex cylinder is housed in an aluminum cylinder of 20 cm diameter and 30 cm long. A metal grid attached to the other end of the source tube confines the plasma from a turbomolecular pump that routinely achieves base pressures of

10^{-6} mbar. The partial gas pressures of Ar and SF$_6$ are determined by controlling the flows.

All measurements reported here were made along the revolution axis (z axis) of the discharge. Two types of electrostatic probes were used for measurements. The first is a nickel planar probe with guard ring biased at the same potential as the probe, to measure the real saturated positive ion current. The diameter of the collecting area was 4 mm and the diameter of the outer ring was 8 mm. The second is a passively compensated Langmuir probe (LP),[10] of 0.25 mm diameter and 6 mm long platinum wire tip. The LP was used to find the plasma potential, electron densities and electron temperature from measurements of the probe $I(V)$ characteristics using a Smartsoft data acquisition system.[11] The electronegativity, $\alpha = n_-/n_e$, and consequently the ion densities (electro-neutrality $n_+ = n_- + n_e$ was assumed), were measured according to the double-probe technique described in Ref. 12. This technique, which relies on the theory developed in Ref. 3, allows to deduce α from the ratio of the cylindrical probe current at the plasma potential to the positive ion saturation current measured by the planar probe, $R = I(V_p)/I_{sat+}$.
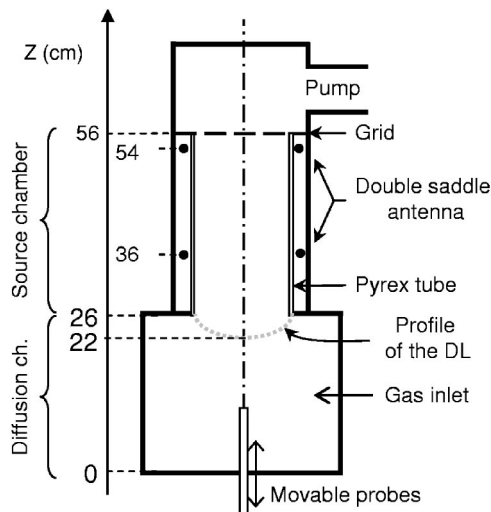

FIG. 1. Schematic of the experimental setup.

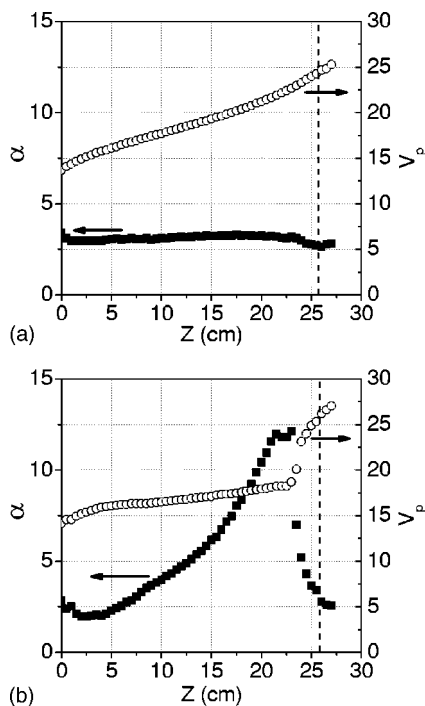[a] Electronic mail: plihon@lptp.polytechnique.fr

FIG. 2. Spatial evolution of the plasma potential, V_p, and the electronegativity, $\alpha = n_-/n_e$, in (a) the no DL case (6% SF_6 mixture), and (b) the DL case (9% SF_6 mixture), at 1 mTorr, 600 W.

FIG. 3. Spatial evolution of the particles densities and electron temperature for a 9% SF_6 mixture at 1 mTorr, 600 W.

The technique requires an estimation of the ratio of the electron temperature to the negative ion temperature $\gamma = T_e/T_-$ and the positive ion mass m_+, both difficult to measure in the gas mixture studied here. We chose $\gamma = 15$, as is commonly thought to be a reasonable value in low pressure electronegative discharges, and $m_+ = 40$ since (i) Ar^+ may be dominant since we used small percentages of SF_6 in argon; (ii) we expect a fairly high dissociation degree of SF_6 and, therefore, SF_x^+ ions with $x \ll 6$ (low mass ions). As a consequence of these estimations, the absolute values of α should be regarded as indicative. However, we believe that spatial gradients of α, or relative variations with operating conditions (pressure, power, mixture) are correctly captured by the technique.

Stable DLs are accessible for pressures from 0.3 to 10 mTorr when carefully adjusting the SF_6 concentration. The minimum power required to obtain a stationary DL increases with increasing pressure, with no upper limit observed (at 1 mTorr, DLs are observed above 200 W; at 10 mTorr, above 1400 W).

All results presented here are for a gas pressure of 1 mTorr and an input power of 600 W. Figure 2(a) shows the axial evolution of the plasma potential and the electronegativity for a SF_6 concentration of 6%. The dashed line represents the position of the interface between the source and the diffusion chamber. The plasma potential decreases continuously from the source to the diffusion chamber, as expected for an expanding plasma which exhibits a gradient in the electron density, while the electronegativity remains roughly constant along the axis. There is evidently no DL. For these conditions of pressure and power, the transition towards the formation of the DL is observed to occur at about 8% SF_6 concentration; with no DLs observed in pure argon or for SF_6 concentrations below 8%. Above this concentration, 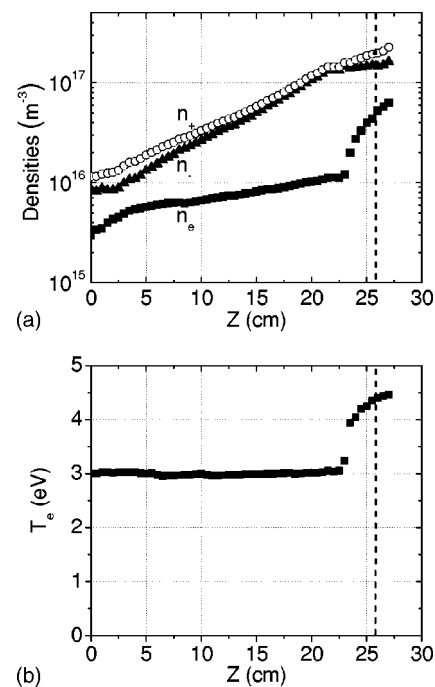the plasma potential and particles gradients are drastically changed as shown in Fig. 2(b). The plasma potential presents a sharp drop at around $z = 22$ cm on the axis, that is about 4 cm below the interface between the two chambers. The potential difference between the source chamber and the diffusion chamber seems to be at least 10 V, although the sharp drop seems to be around 5 V [which is $1.5(kT_e/e)$ with the downstream electron temperature]. This sharp drop is preceded by a strong but smoother gradient that resembles a pre-sheath. From visual observation, it seems that the DL has a spherical shape that is attached to the boundary between the source and the diffusion chamber and that expands into the diffusion chamber (refer to the dashed gray line on Fig. 1).

The electronegativity is also profoundly affected. It presents a sharp maximum at the DL position, with a slow decay downstream (below the DL in the diffusion chamber) and a much faster decay upstream. The variations of α are directly related to the change in the electron density, as shown in Fig. 3(a). The electron density is strongly affected by the sudden drop in potential, whereas both the positive and negative ion densities seem to decrease continuously from the source to the diffusion chamber. The electron temperature changes significantly when crossing the DL. The DL acts as an internal boundary (or sheath), which separates two plasmas; a high electron density, high electron temperature, low electronegativity plasma upstream, and a low electron density, low electron temperature, high electronegativity plasma downstream.

As the SF_6 concentration is increased, the upstream plasma moves further into the diffusion chamber (for a 11% SF_6 mixture, the spherical shape of the DL being more elongated, the position of the DL on the axis is $z = 18$ cm) and the plasma potential drop becomes less abrupt, gradually replaced by a larger region of strong gradient of potential upstream, before entering the DL itself. The downstream plasma potential remains mostly constant at about 15 V.

Downstream and upstream of the DL, the electrons remain in Boltzmann equilibrium, with temperatures given by

the slope of $\ln(n_e)$ as a function of V_p being 3.2 eV downstream and 4.5 eV upstream, which is very close to the temperatures (from LP processing) given in Fig. 3(b). On the contrary, negative ions are far from Boltzmann equilibrium, and are present both sides of the DL. Since they cannot cross it from upstream to downstream (their temperature is much too small), they must be created downstream, i.e., the attachment rate must be strong in this region. This may be due to the relatively low electron temperature, and also to higher neutral gas density because of colder neutral gas (inductive discharges are known to produce significant gas heating near the coil). The negative ions created in the big buffer region downstream from the DL would then be accelerated toward the source through the DL. Unlike attachment, ionization is probably mainly located in the source region where the the electron temperature is high. Hence, positive ions are mainly produced in the source region and are accelerated downstream through the DL. The DL is, therefore, crossed by two ion streams in opposite directions.

The origin of the DL formation remains unclear. From our data and from visual observation, we can postulate that the DL is formed at the boundary between the two chambers and diffuses in the diffusion chamber, as proposed in earlier work for electropositive gases.[8] However, this geometric feature is not sufficient to explain our observations since we did not observe the DL in pure argon. We can postulate that the SF_6 addition has two main effects that contributes to the DL formation. First, the positive ions will more easily reach the ion sound limitation (a necessary condition to form a DL) since it is well known that the ion sound speed is lowered in electronegative plasmas.[13] Second, the attachment process is a very efficient loss term for electrons during the plasma expansion, which makes steeper n_e gradients and, therefore, higher potential gradients. This effect may be compared to the strongly divergent magnetic field used by Charles and co-workers,[6] which also acts as a loss process for electrons during the expansion.

For the typical conditions considered so far (1 mTorr and 600 W), the DLs were stable between 8% and 13% SF_6 fractions. Above 13%, the DL becomes unstable, and periodic oscillations of the charged particle densities, plasma potential and electron temperature are observed. It seems that the unstable regime is characterized by a periodic formation and propagation of a double layer. This instability has strong similarities with the downstream instabilities observed and modeled by Tuszewski and co-workers.[14,15] However, the association between the downstream instabilities and a DL was not clearly established by these authors. This issue will be treated in a separate publication.

We have observed double-layer formation in the expanding region of an inductively coupled electronegative plasma. The DL's were not observed in pure argon. They are stable for a small window of electronegativity and become unstable at higher electronegativity. They seem to have a spherical shape and be formed at the boundary between the source and the diffusion chambers. They act as an internal boundary between a high electron density, high electron temperature, low electronegativity plasma upstream, and a low electron density, low electron temperature, high electronegativity plasma downstream. They exist in a wide range of pressure and power and without a strongly divergent magnetic field, which can be seen as an advantage for space plasma propulsion. However, the voltage drop in the DL is about three times smaller than the DL's described by Charles and co-workers.[6]

[1]M. Raadu, Phys. Rep. **178**, 25 (1989).
[2]I. Kouznetsov, A. Lichtenberg and M. Lieberman, J. Appl. Phys. **86**, 4142 (1999).
[3]T. Sheridan, P. Chabert, and R. Boswell, Plasma Sources Sci. Technol. **8**, 457 (1999).
[4]T. Sheridan, N. Braithwaite, and R. Boswell, Phys. Plasmas **6**, 4375 (1999).
[5]R. Merlino and J. Loomis, Phys. Fluids B **2**, 2865 (1990).
[6]C. Charles and R. Boswell, Appl. Phys. Lett. **82**, 1356 (2003).
[7]C. Charles and R. Boswell, Phys. Plasmas **11**, 1706 (2004).
[8]J. Andrews and J. Allen, Proc. R. Soc. London, Ser. A **320**, 459 (1971).
[9]R. Boswell, Phys. Lett. **33A**, 457 (1970).
[10]A. Cantin and R. Gagne, Appl. Phys. Lett. **30**, 31 (1977).
[11]M. Hopkins and W. Graham, Rev. Sci. Instrum. **57**, 2210 (1986).
[12]P. Chabert, T. Sheridan, R. Boswell, and J. Perrin, Plasma Sources Sci. Technol. **8**, 561 (1999).
[13]N. Braithwaite and J. Allen, J. Phys. D **21**, 1733 (1988).
[14]M. Tuszewski and S. P. Gary, Phys. Plasmas **10**, 539 (2003).
[15]M. Tuszewski, R. R. White, and G. A. Wurden, Plasma Sources Sci. Technol. **12**, 396 (2003).