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Surface Tension in Plasmas Related to Double Layer Formation

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Abstract

Self-organized space charge configurations bordered by electric double layers appear in plasma as the result of the transition into a state characterized by local minimum of the free energy. Considering the self-assemblage process of such a complex well-confined space-charge configuration in plasma, known by the name of ball of fire, as a nucleation process, it becomes possible to define an equivalent surface tension for the double layer that covers the core of the ball of fire and to make some predictions for its surface tension coefficient and capacitance.

Keywords:

self-organization, ball of fire, double layer, nucleation, surface tension

1. Introduction

An external constraint (*e.g.* an electric field) applied on a gaseous conductor, acting on a large number of particles can, in appropriate conditions, give birth to new phenomena enjoying self-organization related to long-range correlations [1-4]. In other words, the constraint leads to a "new state of the same matter", more ordered than the former one, following a "non-equilibrium phase transition" [5]. The term of "non-equilibrium phase transition" is used here concerning self-organization phenomena leading to the appearance of ordered space charge arrangements. Such a complex space charge arrangement bordered by an electrical double layer (DL) (Fig.1), known as ball of fire (BF), is self-assembled in a plasma diode in front of the anode when the external constraint surpasses a critical value. After its appearance the BF behaves as a structure whose self-consistence and stability are related to surface tension. These BF properties are determined by a self-consistent DL, present at its border. The DL, acting as a membrane characterized by a certain surface tension, is able to balance the kinetic pressure of electrons, positive ions and neutrals from the well located volume delimited by it.

Because of the resemblances between the behavior of BF and a liquid drop we may see BF as a result of a nucleation process. More precisely, we will consider the stationary stable BF a "droplet" of a new ordered phase, spontaneously formed in a uniform non-equilibrium plasma for a critical value of the control parameter (*i.e.* the potential applied on the anode) and, consequently,

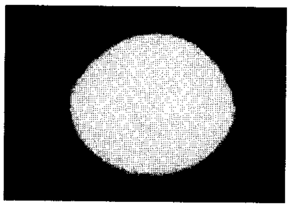

Fig. 1 Photograph of a stationary stable ball of fire.

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we will define an equivalent surface tension for the DL that surrounds its core.

The aim of this paper is to estimate the surface tension coefficient and the specific capacitance (*i.e.* the capacitance per unit of surface) associated with the self-consistent DL that surrounds the core of the stationary stable BF.

2. Why Should Double Layers Have Surface Tension?

Based on the studies on laser-plasma interaction, Hora asserted that the existence of electric fields at the boundary between two plasmas is an argument for considering the surface tension concept in order to analyze the stability of the boundary region [6,7]. He introduced the surface tension for characterizing the DL as an interface between two plasmas with different properties. Contrary to surface tension in liquids, the cohesion forces work as long as the current ensures the flux of electrons necessary for the continuous self-assemblage of the DL.

There are many arguments for considering BF as the result of a phase transition (*i.e.* nucleation) process. Both BF and the liquid drop appear spontaneously and coexist with the phase from which they arose; both of them are protected from the surroundings by a quasi-spherical ''membrane'' that is essential in maintaining them. After Lenard [8] the liquid drop membrane is in fact an electric DL, like in BF's case. Moreover, in the case of the anode-electrolyte interface [9], the DL theory was applied first to the theoretical analysis of the properties of interfaces (*e.g.* surface tension, differential capacitance). In the case of a liquid-liquid vapors interface the forces responsible for the surface tension are directed towards the liquid interior, because the liquid has a grater density than its vapors. This situation is formally encountered in the BF's case. The electrostatic forces acting on electrons are directed towards the inner positive sheet of the DL that covers the core of BF. We have considered the forces acting on electrons because they are responsible for the self-assemblage of BF, as well as for the ionization processes inside the core that make the core density to be greater than that of the outside plasma. The experiments in which BFs arise reveal a hysteresis phenomenon [3] for which appearance the DLs are responsible. Because the hysteresis phenomenon is also present in nucleation processes [10], for which appearance surface tension is responsible, this is another reason for associating a surface tension to the DL that covers the core of BF.

3. Surface Tension of Plasma Double Layers

It is experimentally proved that self-organized structures of BF-type exist in spite of electrons' recombinations with positive ions. This proves that a dynamical equilibrium is established between the processes of generation and accumulation and respectively recombination and diffusion of the two kinds of charges. More specifically, those electrons from the surrounding plasma that have enough kinetic energy to surpass the negative part of the DL will be accelerated in the DL's electric field, so they will reach the core with sufficient energy for producing neutrals' ionization. After the ionization processes a part of electrons' kinetic energy remains as thermal energy. As a result of thermal diffusion these will leave the core. A part of the reminders will recombine with the positive ions, ensuring in this way the neutrals' concentration required for maintaining a constant ionization rate. Under such conditions, the net positive charge in the BF's core will remain constant. When the number of recombinations in unit time interval will be equal to the number of ionizations in the same time interval, we may consider BF in a stationary stable state.

In collisional plasma at relatively high pressures, DL becomes able to sustain its own existence only by internal processes, or with other words, DL becomes self-consistent. This happens when the potential drop across it reaches the ionization potential V_i of the working gas [3].

In agreement with experimental results [3,4,11], in a very simple model, we will consider the stationary stable BF as a spherical structure, electrically neutral as a whole, being composed by a core with radius R, uniformly charged in volume with positive charge and covered by a DL having thickness δ. The external part of the self-consistent DL is negatively charged with the charge (-Q) uniformly distributed on the surface [see Fig. 2(a)]. In the theory of electricity, a DL is defined as a structure composed by two adjacent space charge layers with the same superficial charge density, but with opposite signs [12,13]. From this we may conclude that the total charges from each layer, disregarding the sign, are not equal, excepting the planar case. It follows that the inner positive layer has a smaller total charge than the outer negative one, meaning that the positive charge difference is located in the core of BF. The above assumption is supported by experimental results [4,11], which clearly show that in the core the positive ions' concentration is greater than in the outside plasma.

Considering the above said we can analyze the space charge configuration of a stationary stable BF with the methods of electrostatics. Under these circumstances, using Gauss law, the potential drop on the self-consistent DL will be:

$$V_i = \frac{Q}{4\pi\varepsilon_0 R} \cdot \frac{\delta}{R+\delta} \qquad (1)$$

where ε_0 is the vacuum permitivity, $Q = en_+ \cdot 4\pi R^3/3$ is the total electric charge of the BF's core, n_+ being its charge concentration and e the electron charge. As a result of the above mentioned dynamical equilibrium we can consider n_+ having a constant value. The radial distribution of the potential over BF [Fig. 2(b)] is similar to that obtained from experiments [4,11].

From Eq. (1) it will follow the dependence of the DL's thickness δ on the core radius R:

$$\delta = \frac{RR^{*2}}{R^2 - R^{*2}} \qquad (2)$$

where R^* has the expression

$$R^* = \sqrt{\frac{3\varepsilon_0 V_i}{e n_+}} \qquad (3)$$

Since δ must have a positive and finite value it results [see Eq. (2)] that $R > R^*$. This means that R^* is a critical value of the BF's core radius R.

Because the surface tension of a quasi-gaseous object such as BF may only result owing to electric charges [14], we have calculated the electrostatic potential energy of the DL:

$$W_p = \frac{1}{2}\rho_s 4\pi R^2 V_i = 2\pi\varepsilon_0 V_i^2 \frac{\left(R^2 - R^{*2}\right)^2}{RR^{*2}} \qquad (4)$$

where $\rho_s = Q/[4\pi(R+\delta)^2]$ is the surface charge density of the DL.

In order to evaluate the surface tension coefficient σ associated with the DL that covers the BF's core we have estimated the change of the free energy for the increase with ΔS of the BF's surface without changing its volume. This change of the free energy in isothermal conditions is just the potential energy variation of the DL:

$$\Delta W_p = \sigma \Delta S \qquad (5)$$

Consequently, the potential energy variation when the surface of the BF increases under the above conditions can be expressed as:

$$\Delta W_p = \frac{\varepsilon_0 V_i^2}{2} \frac{\left(R^2 - R^{*2}\right)\left(3R^2 + R^{*2}\right)}{2R^{*2}R^3} \Delta S \qquad (6)$$

From (5) and (6) it follows that:

$$\sigma = \frac{\varepsilon_0 V_i^2}{2} \frac{\left(R^2 - R^{*2}\right)\left(3R^2 + R^{*2}\right)}{2R^{*2}R^3} \qquad (7)$$

This result sustains the above observation, which states that the DL exists only for $R > R^*$. For $R = R^*$ the surface tension coefficient becomes zero making the DL to lose its role of "membrane" that maintains the BF existence.

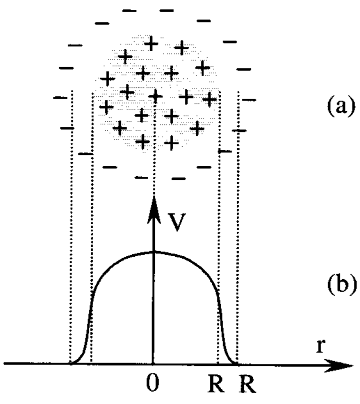

Fig. 2 (a) Ball of fire schematic representation with the core charge uniformly distributed in volume. R is the core raduis and δ the DL thickness; (b) Potential profile of the stationary stable ball of fire.

4. Specific Capacitance of Plasma Double Layers

As experimentally proved (Fig.3) between the current and the potential of a gaseous conductor containing a BF there exist a phase difference showing

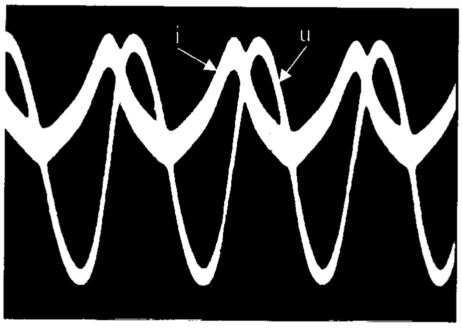

Fig. 3 Phase difference between $i(t)$ and $u(t)$ for a plasma diode in which a ball of fire is formed.

that the DL which border the BF plays a capacitive role. This allows us to compute the specific capacitance of the DL. For this purpose let us apply an ac tension over the dc one between the anode and the cathode of the plasma diode, after the stable BF's formation:

$$u(t) = U_0 \cos \omega t \quad (8)$$

where U_0 is the ac tension amplitude. The frequency $\omega/2\pi$ and the amplitude of the applied ac tension must have such low values that the current distribution inside the DL, as well as outside it, to be the same as in the case of a constant electric field.

Because the DL is a capacitive element of circuit, between the tension and the current there will be a phase difference φ, such that:

$$i(t) = I_0 \cos(\omega t + \varphi) \quad (9)$$

where I_0 is the ac current amplitude.

The instantaneous power will be:

$$p(t) = U_0 I_0 (\cos^2 \omega t \cos\varphi - \sin \omega t \cos \omega t \sin \varphi) \quad (10)$$

The first part of Eq. (10) is the power dissipated by Joule effect. The second represents the supplementary power accumulated into the DL structure, due to the superposition of the ac tension over the dc one:

$$\begin{aligned} p_{acc}(t) &= U_0 I_0 \sin \omega t \cos \omega t \sin \varphi \\ &= U_0 I_0 \sin \varphi \frac{\mathrm{d}}{\mathrm{d}t}\left(\frac{\sin^2 \omega t}{2\omega}\right) = \frac{\mathrm{d}w_{acc}}{\mathrm{d}t} \end{aligned} \quad (11)$$

From Eq. (11) we get the additional energy wacc accumulated in the DL structure:

$$w_{acc} = U_0 I_0 \sin \varphi \frac{\sin^2 \omega t}{2\omega} \quad (12)$$

The averaged energy over a period will be:

$$W_{acc} = \frac{U_0 I_0 \sin \varphi}{4\omega} \quad (13)$$

so, the specific capacitance of the self-consistent DL that covers the BF's core will have the expression:

$$C = \frac{2}{V_i^2} \cdot \frac{W_{acc}}{4\pi R^2} = \frac{U_0 I_0 \sin \varphi}{8\pi\omega R^2 V_i^2} \quad (14)$$

The electrostatic model used above for the surface tension coefficient evaluation also offers the possibility to find the specific capacitance of the DL:

$$C = \frac{\rho_s}{V_i} = \varepsilon_0 \cdot \frac{\left(R^2 - R^{*2}\right)^2}{R^{*2} R^3} \quad (15)$$

It is a matter of experimental measurements to prove the convergence of these two expressions for the specific capacitance of a quasi-spherical DL, this being the subject of a forthcoming paper.

5. Conclusions

In the phase transition theory [15] it is stated that the approaching of the critical point is accompanied by the thickness growing of the superficial layer between the two phases taken into consideration. Also, at the critical point, the surface tension goes to zero. In the present case the critical point corresponds to $R = R^*$ because the decreasing of the core radius towards R^* makes the DL's thickness to approach an extremely large value (the DL practically vanishes at this core radius). The superficial tension coefficient, as well as the specific capacitance go to zero when R tends to R^*.

We notice that all the above mathematical evaluations, in spite of their simplicity, are valid for a stationary stable BF. The essential novelty of this paper is that BF is considered as a result of a phase transition produced in plasma after self-organization. Consequently, it is stressed that the DL that covers the core of the BF plays the role of a "membrane" that splits the plasma in two regions with different characteristics.

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