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# Dynamic space charge structures in high fluence laser ablation plumes

S. GURLUI*, M. SANDULOVICIU, M. STRAT, G. STRAT[a], C. MIHESAN[b], M. ZISKIND[b], C. FOCSA[b]
*Al. I. Cuza University of Iasi, Faculty of Physics, B-dul Carol I Nr 11, Iasi, RO-6600, Romania*
[a]*Gh. Asachi Technical University of Iasi, Department of Physics, B-dul Carol I Nr 10, Iasi, RO-6600, Romania*
[b]*Laboratoire de Physique des Lasers, Atomes et Molécules (UMR 8523), Centre d'Etudes et de Recherches Lasers et Applications (FR 2416), Université des Sciences et Technologies de Lille, 59655 Villeneuve d'Ascq cedex, France*

The formation and dynamics of the space charge structures have been studied by means of multiple Langmuir probes in a laser ablation plume produced by high fluence ns laser ablation of a copper target. Based on the analysis of the temporal evolution and angular distribution of ionic densities, a schematic view of space charge structure dynamics is proposed. The observation of periodic oscillations in the MHz range suggests the presence of a self-organization process.

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## 1. Introduction

There is a high current interest in understanding the fundamentals of the complex pulsed laser desorption/ablation phenomena, due to their importance in a wide range of applications as pulsed laser deposition (PLD), nanoparticle formation and growth, chemical analysis etc. [1,2]. At high laser pulse energy, the plume of ablated materials appears as a nearly spherical plasma blob [3-5] that has its origin in the non-equilibrium hot plasma produced at the impact point of the laser beam on the solid target.

The self-assembly process and the dynamics of double layer space charges have been investigated during the last years in plasma diodes. A phenomenological model essentially based on a new scenario of self-organization suggested by plasma experiments has been published [10, 11]. We have shown [20] that the scenario of self-organization explaining the emergence of complex space charge configuration in gaseous conductors (plasma) is potentially able to explain the behavior of the multifunctional systems as those used in micro- and nano – technologies. In laser ablation experiments some different processes are involved, including light absorption, evaporation, transient gas dynamics, radiation transport, condensation, ionization and recombination. All these processes are rather complex and require further investigation.

In this paper we investigate the total ionic current at different time moments measured by a set of Langmuir probes circularly placed in front of the plasma plume produced by nanosecond Nd:YAG laser ablation of a copper target. The obtained experimental results emphasize the presence of a moving electrical double layer originating at the border of the self-assembled plasma blob following the laser pulse.

As we show in the following, for understanding these processes in the high fluence regime, one must take into consideration a succession of phenomena: (i) the production of a local hot non-equilibrium plasma whose temperature is so high that a "gas anode" appears due to the difference between the thermal diffusivity of electrons and positive ions; (ii) the presence in the next evolution phase of inelastic collisions between the electrons and the gas atoms produced after acceleration towards the gas anode. Both phenomena are premises for the presence of self-organization [8].

## 2. Experimental details

The experiments have been performed in a stainless steel vacuum chamber ($10^{-7}$ Torr) evacuated by means of a 450 l/s turbomolecular pump (Pfeiffer). The second harmonic (532 nm) of a 10 ns Nd:YAG laser (Quantel Brillant) has been focused with a 25 cm lens on the target (copper) surface placed under vacuum on a X-Y-Z micrometric stage. The estimated spot diameter at the impact point has been ~100 µm. The laser energy usually employed was 45-50 mJ/ pulse; this leads to a typical laser intensity of ~ 50 GW/cm$^2$.

The total ionic current extracted from the high potential side of the nearly spherical space charge structure was measured by means of ten cylindrical probes biased at negatively voltage (-30 V, stabilized dc power source). The probes (numbered anticlockwise from 1 to 10) were positioned in a circular array around the impact spot of the laser beam at a distance of 40 mm. The angular separation between two consecutive probes is constant; the probes in the extreme positions form a 60$^\circ$ angle with the normal to the target.

The 10 transitory signals have been simultaneously recorded by three 500 MHz digital oscilloscopes (Le Croy) and transferred to a PC for further analysis (Lab View environment). A schematic view of the employed experimental setup is given in Fig. 1.

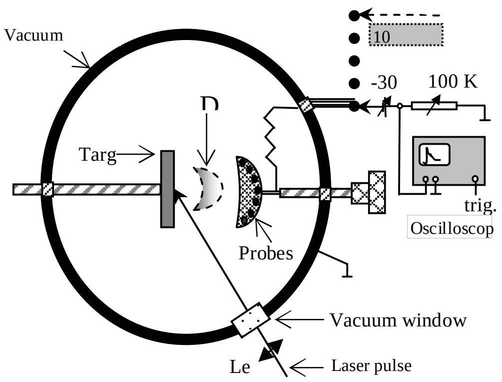



*Fig. 1. Experimental set-up.*

### 3. Results and discussion

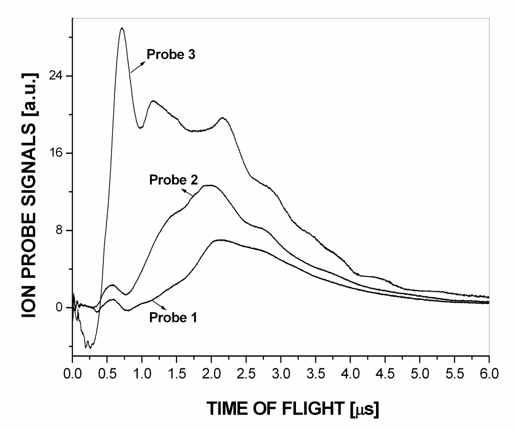
The transient ionic currents measured by the Langmuir probes are displayed in Fig. 2. When analyzing these time of flight (TOF) profiles we notice the following:

(1) Multi-peak ionic density signals are recorded by all the probes; the amplitudes of these signals depend on the probe position with respect to normal to the target surface.

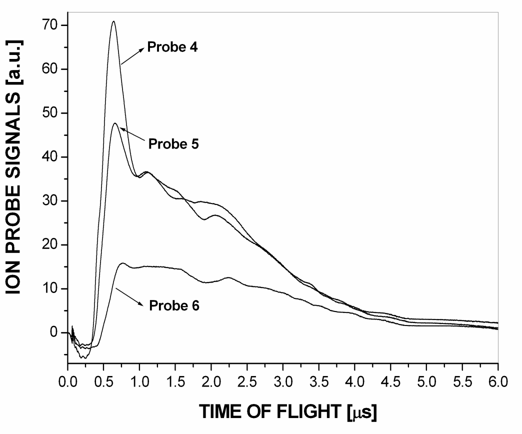
(2) All the TOF profiles emphasize two components: a fast component obtained for all the probes at a flying time of 0.7 μs (the time of flight profile obtained from probe 7 has a shoulder at 0.7 μs) and a multi-peak slower component. The high and sharp maximum corresponds to a group of ions located in the high potential side of the double layer; the velocity of this group, derived from present data, is therefore of $\approx 5.4\times10^4$ m/s.

(3) The signals corresponding to the fast component obtained by the probes 3-6 are much more intense than those of the 1, 2, 7-9 probes.

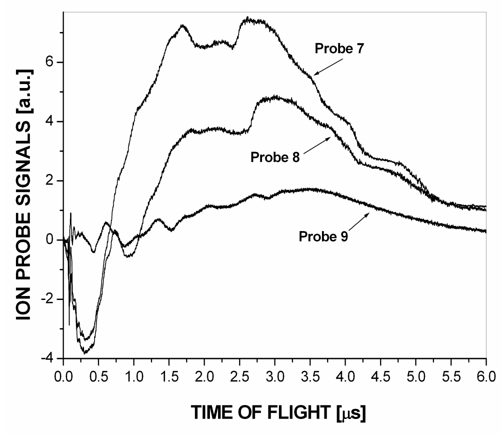
In a recent work [18] we evidenced the existence of periodic variations in the total ionic densities recorded by a Langmuir probe placed at 3 cm distance on the normal to the copper target. We can now extend these studies to longer distances from the target (e.g. 4 cm in the present work) and also undertake an angular investigation (up to ± 60° off the normal to the target) of plasma plume dynamics.




a)




b)




c)

*Fig. 2. Transient (time-of-flight) signals recorded by Langmuir probes in a plasma plume created by 532 nm laser ablation of a Cu target. The distance from target to probes is 40 mm.*

From the TOF signals in Fig. 2 we can state that the variation of the copper ion density "propagates" with high speeds in a narrow conic space region, close to the normal to the target surface (Fig. 2a, 2b). This "forward peaking" effect is confirmed by the angular analysis presented below; it results in $\cos^n \theta$ distributions with high n (of the order of 10).

The plasma ablation process produced by the laser pulse is followed by the appearance of a well localized space charge of positive ions that propagates away from the plume (at lower speed) localized in a much larger conic space region. In the case of our measurements, the total angular opening available is about 120°. Considering similar phenomena observed in plasma diodes [8], we can attribute the presence of the space charge of positive ions to an electrical double layer originating at the border of the plume.

We can see that the ionic current distribution is function of the direction in the plume extension space and has periodic oscillations (with frequency in the range of MHz; the number of periods observed increases with the energy of the laser pulse). The ionic current oscillations can be attributed to the observed plume splitting process [6] related to a periodic shelling-off process of the double layer from the border of the plume.

Similar phenomena observed in plasma devices revealed that, for surviving a certain span, the plume, that actually represents a complex space charge configuration, performs a rhythmic exchange of matter and energy with the surrounding plasma created by the laser pulse.

Based on the analysis of the data obtained in this paper, Fig. 3 gives a schematic representation of the proposed multiple double layer structures in a laser ablation plume.

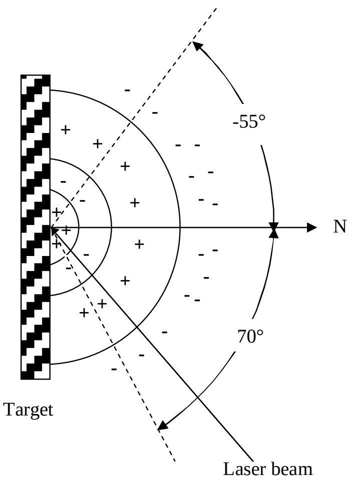



*Fig. 3. Schematic representation of the multiple double layer geometry.*

The maximum ion current of each Langmuir probe has been measured and the angular distribution obtained is displayed in Fig. 4. The angular distribution $I(\theta)$ of ablated particles is often expressed as [12]:

$$I(\theta) = I(\theta_0)\cos^n(\theta - \theta_0) \qquad (1)$$

where $n$ is an exponent and $\theta_0$ represents the plume axis displacement with respect to the normal to the target. Typically, $n$ varies between 5 and 13 [14-17].

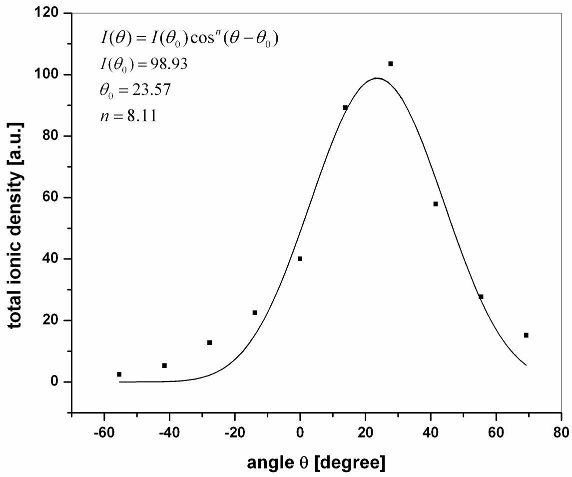



*Fig. 4. Angular distribution of the maximum ionic current. The solid line is the fit of experimental data to Eq. (1).*

By fitting our experimental data to Eq. (1) we found the exponent value $n \approx 8$, which is in very good agreement with the previous results obtained in similar configurations [14-17].

We remark that the maximum of the ionic density is not on the normal to the target, but displaced by an angle of ~20° (i.e. the ionic current is maximum for the probes 4 and 5, whereas probe 6 is on the normal to the target). A possible explanation would be the presence of a deep crater in the target, formed by the repeated action of the intense laser pulses. This may lead to an ejection direction that is different from the normal to the target or to a crater shape that would block part of the ejection plume. Preliminary optical microscopy observations seem to confirm this hypothesis, but further work is necessary in order to get a definitive answer.

## 4. Conclusions

In this paper we have studied the dynamics of the double layers obtained in laser ablation plumes using an experimental setup based on multiple Langmuir probes. The temporal evolution and the angular distribution of the ionic densities have been registered and analyzed. This allowed the emergence of a schematic view of space charge dynamics in the plasma plume. Moreover, the observation of MHz oscillations led to the hypothesis of the presence of a self-organization process.

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* Corresponding author: sgurlui@uaic.ro