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Determination of recombination length of a non-equilibrium plasma produced by laser ablation

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Summary. — An experimental study of the laser ablation produced plasma evolution is necessary for its deeper understanding, since plasma expansion has both spatially and temporally varying characteristics. We irradiated a Cu target with a KrF laser beam. A small Faraday cup array and an axial Faraday cup were used as diagnostic systems, in order to study the spatial variation in the total charge carried by plasma ions. Charge loss during the plasma expansion was observed, which was attributed to the charged species recombination. This occurred upstream to the critical distance where the plasma density is high enough. Downstream the critical distance the plasma particles collisions were negligible and the ion charge remained frozen. In these experiments it was observed that the critical distance for charge recombination was a function of laser fluence.

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1. – Introduction

The interaction of laser radiation with metallic targets is a very useful tool to create easily plasma plumes consisting of highly concentrated electrons, ions and neutral particles. The plasma evolves in adiabatic expansion predominantly along the direction of the normal to the target [1]. The characteristics of the expanding plasma, *i.e.* the plasma density as well as the charge-state abundances, are dependent on the target distance and they exhibit the highest values near the source [2-5].

The aim of this work is devoted to the characterization of ion current changes resulting from the plasma expansion into the vacuum for the development of a laser ion source where it is necessary to balance, in appropriate way, the position of the ion extraction gap in order to achieve the highest intensity of the extracted ion beam.

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Fig. 1. – Experimental set-up. W: quartz windows; V_c : bias voltage.

Near to the irradiated target the three-body recombination process lowers the average charge state of the expanding ions [1,2]. Starting from the recombination rate, which is proportional to $\approx q^3 n_e^2 T_e^{-9/2}$ (q is the ion charge state, n_e is the electron density and T_e is the electron temperature), and making some assumptions on the plasma behaviour [3] (among which an adiabatic spherical expansion with a well-defined dependence of the ion velocity, n_e and T_e on the distance from the target) it is possible to estimate the critical distance from the target, $L_{\rm CR}$, by the following equation

(1)
$$L_{\rm CR} \approx T_{\rm in}^{13/12} \nu^{14/6} \tau_{\rm L}^{13/6} / n_{\rm CR}^{8/18} d^{8/6},$$

where $T_{\rm in}$ is the initial electron temperature of the plasma cloud at the end of the heating stage, ν is the ion velocity, $\tau_{\rm L}$ is the laser pulse duration, $n_{\rm CR}$ is the critical electron density over which the plasma kernel is shielded from the laser radiation and d is the laser spot diameter. After $L_{\rm CR}$, the initial sharp rise of recombination losses is changed to a slow descend. Roudskoy also estimated that a typical value of $L_{\rm CR}$ can reach a few meters for a 10.6 μ m laser wavelength (CO₂) and can be only a few centimetres for a 1.06 μ m laser wavelength (Nd:glass). One can deduce from this large range of $L_{\rm CR}$ that any comparison of different laser ion sources could be misleading without a direct measurement of ion charge losses during the plasma expansion into the vacuum system.

For distances from the target larger than $L_{\rm CR}$, the "freezing" of charge states dominates because the recombination process rate in the expanding laser plasma slows down quickly on the distance. For this reason the aim of this work is to study the plasma flux changes during the plasma expansion in a vacuum.

2. – Experimental apparatus and results

The laser-produced plasma is created in a stainless-steel vacuum chamber (10^{-6} mbar) by focusing an excimer laser pulse (KrF, 248 nm wavelength and 20 ns pulse duration) up to irradiances of the order of 10^9 W/cm^2 onto a plane copper target. The focusing lens has a focal length of 15 cm and the laser spot is about 1 mm^2 . The laser light strikes the target at about 70° with respect to the target normal. The target chamber is axially connected to an 8 cm in diameter drift tube to study the plasma evolution at various distances. Figure 1 presents a sketch of the interaction chamber. Two diagnostic equipments are used: a system of 13 small Faraday cups able to measure the plasma

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Fig. 2. – Diagnostic system made up of 13 small Faraday cups of $4.5 \,\mathrm{mm}$ in diameter used for the plasma angular distribution measurement.

angular distribution and one movable axial Faraday cup to record the distance-charge dependence utilized to control the presence of the recombination processes. In the former set-up the 13 Faraday cups lie in a line perpendicular to the target normal. Each collector has a diameter of 4.5 mm and the distance between the centers of each other is 5.5 mm (see fig. 2). In the second set-up the cup has a diameter of 10 mm. All Faraday cups were biased at a negative voltage of -500 V, in order to collect positive ions.

The experiment was performed at different laser intensities of $4 \cdot 10^8$, $14 \cdot 10^8$ and $34 \cdot 10^8 \text{ W/cm}^2$.

Due to the near-surface collisions amongst the emitted particles, the laser-produced plasma shape is forward peaked and the density distribution of charged or neutral particles in a plane parallel to the target surface is well described by the following equation [6]:

(2)
$$\frac{\mathrm{d}Q}{\mathrm{d}S} = \frac{A}{L^2} \cos^p \phi,$$

where A is a coefficient of proportionality, L is the target-cup distance, S is the perpendicular surface at L distance, p is an empirical parameter related to the plasma plume shape, and $0 \le \phi \le \pi/2$ is the zenithal angle respect to the target normal with the origin in the laser spot. If the recombination processes could be neglected, one can rewrite eq. (2) as follows:

(3)
$$\frac{\mathrm{d}Q}{\mathrm{d}\Omega} = A\cos^{p-3}\phi,$$

where $d\Omega = \sin \phi d\phi d\theta$ is the differential solid angle and $0 \le \theta < 2\pi$ is the azimuthal angle in the target surface-plane.

The charge distributions were measured at a fixed distance of 6 cm from the target. To determine the p parameter for the different laser irradiances, the charge was collected by each of the 13 Faraday cups. Considering the cup diameter and fitting the recorded



Fig. 3. – Probability distribution of the charge per unit zenithal angle ϕ at $4 \cdot 10^8$, $14 \cdot 10^8$ and $34 \cdot 10^8 \text{ W/cm}^2$.

distributions by eq. (2), we derived the p values of 9.7, 15.9 and 16.8 for the intensities of 0.4, 1.4 and $3.4 \,\mathrm{GW/cm^2}$, respectively. In fig. 3 the charge probability distribution, $dQ/Qd\phi$, is shown for every p parameter and it is possible to observe that the ion charge increases at small angles with increasing laser intensity and the plasma plume becomes narrower around its axis.



Fig. 4. – Total ion charge in the plasma plume, $Q_{\rm T}$, vs. the distance, L, from the target.

We also used an axial Faraday cup with a radius, R, of 5 mm to study the distance dependence of the total ion charge. The experimental data were collected for target-cup distances up to 70 cm. The collected charge is only a part of the total charge produced, because of the small solid angle subtended by the Faraday cup surface. To evaluate the charge losses due to the recombination effects in the expanding plasma, we assume that the plasma expands in self-similar way. It implies that the plasma shape and, thus, the p parameter are constant during the expansion at fixed laser energy. Therefore, we attributed at every set of data, measured at a given laser irradiance, a p value constant independently of the distance. Taking into account what said above, it is possible to derive from eq. (2) a function which links the total ion charge in the plume, $Q_{\rm T}$, with the charge, $Q_{\rm C}$, collected by means of the Faraday cup at a distance L:

(4)
$$Q_{\rm T} = \frac{Q_{\rm C}}{1 - \cos^{p-2}\varphi} = \frac{Q_{\rm C}}{1 - (\frac{L^2}{L^2 + R^2})^{\frac{p-2}{2}}},$$

where φ is the Faraday cup acceptance angle (tg $\varphi = L/R$). Figure 4 shows the trend of the total charge for the three different laser irradiances recovered applying eq. (4) to the experimental data. In the absence of recombination processes, $Q_{\rm T}$ is independent of the distance. All the three curves of fig. 4 show a constant $Q_{\rm T}$ value for larger distances and an increasing one for shorter distances. This is the evidence that the recombination processes take place below distances of the order of 100 mm, 200 mm and 300 mm for 0.4, 1.4 and 3.4 GW/cm², respectively. Such an order of distances was estimated as the distance from which the variation of the experimental point values is higher than 5% (our experimental error) of the average value of the three furthest points, which are assumed to be in regime of recombination absence.

3. – Conclusion

This paper describes variations in the charge carried by Cu ions during the expansion of UV laser-produced plasma into the vacuum by TOF spectrometry, in the laser irradiance range 10^8-10^9 W/cm². The ion charge, recorded at different distances from the target and at different laser irradiances, made possible the estimation of the upper limit of the critical distance, which was in accordance with Roudskoy's theory. Beyond this upper value, which increases as the focused laser intensity increases, the "freezing" of the ion charge states occurs. The estimation of the critical distance is very important in the extraction of ions from the laser-produced plasma.

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