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Long-range oscillations of a Carbon laser-produced plasma recorded by an electrostatic energy analyzer

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Abstract:

Understanding peculiar dynamics in transient laser-produced plasmas is of great importance for a number of applications as energetic ion production, ion implantation, laser micro-machining etc. Continuing a series of papers on the electrical and optical characterization of laser ablation plasma plumes, we report here the first observation of oscillations in the ionic and electronic currents at long distance (17.5 cm) from the ablation target. An electrostatic energy analyzer was used to finely probe the dynamics of charge carriers in a Carbon plasma generated in vacuum by KrF excimer laser ablation of a graphite target. A short-time Fourier transform data processing approach was applied for the first time to extract valuable information from the recorded currents. This method not only reveals different frequencies for two distinct expanding plasma structures, but also unveils different trends in their temporal evolution. These new experimental results corroborate previous studies supporting a scenario based on the plasma

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structuring in multiple double layers. A discussion with respect to an alternative scenario based on low-frequency sheath instabilities is also presented.

Keywords: ionic current oscillations; excimer laser ablation; carbon plasma; electrostatic energy analyzer.

1. Introduction

Detailed characterization of the complex charged particles dynamics in transient plasmas generated by laser ablation of solid targets bears a high current interest, not only for fundamental reasons, but also due to the high number of potential applications, such as ion implantation,^{1,2} nanoprocessing,³ biomedicine, or surface cleaning and patterning.⁴ In this framework, production of (multiply-charged, high-energy) ions with well-defined properties attracts particular attention: for instance, the availability of various charge states allows controlling the ion implantation depth profile. For instance, as described in Ref. 5, when a uniform Al concentration gradient is required for the p-type doping of SiC, implantation is conducted with singly-charged Al ions with energies ranging from tens to hundreds of keV. The characterization of the laser-generated ions consists in measuring their charge state, number density and angular distribution, energy or velocity distributions. These measurements usually imply combinations of electrical time-of-flight methods, using simple Langmuir probes, Faraday cups, electrostatic energy analyzers or mass spectrometers.⁶⁻⁹ At low to moderate laser ablation fluence, the transient ionic current is typically recorded for tens of μs , regardless to the laser pulse duration (fs, ps or ns). Depending on the ablation process parameters, the ionic current time-of-flight profile may exhibit a multi-peak structure, which is commonly attributed to various charge states or ion nature (in multiple-element plasmas). However, for particular experimental conditions, several authors¹⁰⁻¹⁴ linked the observed multiple-peak structure to

more exotic phenomena, as plume splitting, charge separation in double (multiple) layers, plasma oscillations, or two (multiple) plasma temperatures.

For more than a decade, our groups have tracked the occurrence of these peculiar phenomena in transient plasmas generated by laser ablation in vacuum of various targets, from simple metals¹⁵ to more complex compositions.^{16,17} These contributions were recently summarized in a review paper¹⁸ and a book chapter,¹⁹ therefore only some brief outlines useful for the context of the present study will be reminded here. In a series of early works,^{15,18,20–23} an oscillatory structure of the current recorded by a Langmuir probe was observed for plasma plumes generated by ns laser ablation of Cu or Al targets in vacuum. By measuring the potential variation on the ablation target,²⁰ oscillations of similar (MHz) frequencies were recorded. Later on, by investigating the expansion of a fs-laser produced plasma with a Langmuir probe, the existence of two types (frequencies) of oscillations was shown²⁴ and related to two distinct plasma structures: a fast one consisting of highly charged particles ejected at the early stages of the ablation, and a subsequent colder tail of thermalized particles emitted from the target at later times. However, most of these studies were limited to short distances (typically < 2 cm) from the target surface. A fade-out of the multi-peak/oscillatory features was observed at longer distances^{12,13} and the pronounced decrease of the current intensities when moving away from the target required the use of high (10 M Ω) input impedance (which may by itself preclude the observation of high-frequency phenomena).¹¹

Continuing a series of papers^{25–30} on the investigation of the peculiar dynamics of carbon plasmas generated by excimer laser ablation of graphite targets, we report here for the first time the observation of oscillations in the ionic current recorded with an electrostatic energy analyzer at long (17.5 cm) distance from the target. An original signal processing approach based on the short-time Fourier transform³¹ allows unveiling fine details on the temporal evolution of the oscillation frequencies.

2. Experimental details

The carbon plasma was generated by ablating a pyrolytic graphite target in vacuum (10^{-4} Pa residual pressure) with a KrF excimer laser beam (20 ns, 248 nm, 5 Hz, 264 mJ/pulse) focused at 45° incidence by a spherical lens (50 cm focal length). By changing the lens to target distance (called LP in the following) the laser fluence was varied to obtain the values of 2.1 J/cm^2 for $LP = 45 \text{ cm}$, 1.2 J/cm^2 for $LP = 42 \text{ cm}$, and 1 J/cm^2 for $LP = 40 \text{ cm}$. For the sake of simplicity, only LP will be further specified.

Electrical investigation of the expanding plasma plume was performed with the help of an electrostatic energy analyzer (EEA – see schematic representation on the left side of Fig. 1a), placed at $L = 0.175 \text{ m}$ along the normal to the target surface. It consists in three 5-mm diameter parallel grids of 81% open area (20 μm diameter tungsten wires, 200 μm pitch), placed in the front of a Faraday cup (FC, Kimball Physics). The entrance grid narrows the ion collecting area to achieve a better spatial resolution. A second 9.5 mm diameter aperture is placed at the entrance of the FC and the device total length is 75 mm.

The first grid (exposed directly to the plasma) was grounded (denoted as GG) for a minimal electrical perturbation. The second one, having the role of a suppression grid (SG), was negatively biased at $V_{SG} = -9 \text{ V}$, for completely repelling the electrons. The last one is a retarding grid (RG), which was positively biased in the range of $V_{RG} = +(0\div 120) \text{ V}$ (using Hameg HM8143 stabilized power supplies), to select different portions of the ions energy distribution. A $V_{FC} = -9 \text{ V}$ voltage was applied on the FC for repelling the possible secondary electrons, and therefore to record only the selected ions. The electrical signals were monitored by using the 50Ω input impedance of a 2 GHz oscilloscope (LeCroy).

3. Results and discussion

3.1. Current oscillation regimes in correspondence with fast and slow plasma

structures

The experimentally recorded FC current extends over about 30 microseconds and it generally displays a smooth temporal trace of single-peak aspect.²⁵ However, above an RG bias threshold, the recorded time-of-flight profile splits into (at least) two components. This threshold was found to vary with the laser fluence ($V_{RG\ Th1} = +24\text{V}$ for $LP = 40$ cm, $V_{RG\ Th1} \cong +32$ V for $LP = 42$ cm, and $V_{RG\ Th1} \cong +35$ V for $LP = 45$ cm), which proves³⁰ that the splitting phenomenon is intrinsic to the plasma plume, and not induced by the EEA. Fig. 1a displays an example of the splitting for $LP=42$ cm and three different RG biases. In agreement with our previous works,^{18,32} the first peak is attributed to C_{fast}^{2+} ions, while the second one is given by the C_{fast}^{1+} ion, both belonging to the so-called “fast plasma structure”. They are followed by a hidden peak belonging to C_{slow}^{1+} ions, *i.e.* to the “slow plasma structure”; this can be better evidenced using a signal deconvolution procedure as in Refs. 6 and 7. The complete separation of the two current peaks is achieved at biasing values which are also LP (fluence) dependent: $V_{RG\ Th2} = +38$ V, $V_{RG\ Th2} = +55\text{V}$ and $V_{RG\ Th2} = +62\text{V}$ for $LP = 40$ cm, $LP = 42$ cm, and $LP = 45$ cm, respectively. At higher RG biases, the occurrence of oscillations in the recorded current is evidenced (Fig. 1a), their amplitude increasing with V_{RG} .

At a closer look, in the temporal ranges between 8 μs and 9 μs and between 16 μs and 17 μs (see the insets of Fig. 1a) we can actually observe different oscillation frequencies, which can be assigned to C_{fast}^{1+} and C_{slow}^{1+} ions (see above). In the attempt to precisely measure these two frequencies, some difficulties may arise from the lack of constant amplitude, even when subtracting the continuous (smoothed) signal from the total recorded current. Even in these circumstances, we fitted the oscillatory part with the usual sine function of constant amplitude, which generally gave low values of adjusted R-square (less than 0.5), but allowed calculation

of the oscillation period with a reasonable accuracy (better than 1%). The results are plotted in Fig. 1b, and the average frequency of 12.29 MHz for the fast plasma structure results, with low fluctuations around it. No clear evidence of the frequency dependence on the retarding grid voltage can be inferred; the observed fluctuations might rather originate in small imperfections of the target surface which possibly perturb the focusing conditions, *i.e.* the ejection of highly charged ions. On the other hand, the slow plasma structure oscillates with a frequency which linearly increases with V_{RG} , from about 3 MHz at $V_{RG} = +70$ V to 5 MHz at $V_{RG} = +100$ V.

Similar results (given in Supplementary Material) were obtained for $LP=40$ cm and $LP=45$ cm. For lower laser fluence ($LP=40$ cm) the FC signal diminishes, as expected, while the peaks separation and the subsequent oscillations occur above $V_{RG Th2} = +38$ V, *i.e.* at a lower threshold than the one for $LP=42$ cm. This can be easily explained through the decrease in the ion density number and expansion velocities.³⁰ Similar values for the oscillation frequencies for the fast and the slow plasma structures were also obtained. Conversely, at higher fluence ($LP=45$ cm) the oscillations occur above the increased threshold value $V_{RG Th2} = +62$ V, since the ions are more energetic and in higher concentration. The fast plasma structure also oscillates with similar frequency, while the linear increase for the slow structure is again observed.

The sine function used above returns reasonable values for the frequency, but high error bars on amplitudes and reveals itself to be non-adequate for fitting the early parts of some particular datasets. However, it allows extracting two general concluding remarks: i) the existence of different oscillation frequencies in particular time ranges, of about 12 MHz for the fast plasma structure and about 4 MHz for the signal tail; ii) a linear increase with the retarding grid voltage of the oscillation frequency for the slow plasma structure. The first conclusion is a confirmation of our previous data obtained in different experimental conditions:²⁴ by placing a Langmuir probe at several millimetres from an aluminium target that was ablated by a fs-laser, we observed oscillations of average frequencies $\nu=8.5$ MHz and $\nu'=6.5$ MHz. Moreover, for Cu

targets the probe current initially oscillates with a frequency of $\nu=10.5$ MHz, while later on it becomes $\nu'=6.4$ MHz. As said above, the present work is the first to evidence oscillations at long distance from the target surface, therefore the time range of charged particle detection is also extended. This allowed us recording a more significant number of oscillations, having frequencies of the same order (MHz) of magnitude. Regarding to the linear increase of the slow structure oscillation frequency with the RG biasing, which manifests for high fluences, a possible physical explanation comes from the local increase of plasma ion density in the retarding electric field. This increases the plasma conductivity and, according with the theoretical model from Nica *et al.*,²⁴ the oscillation frequency. We note that in Ref. 24 a weak dependence on the metallic target biasing was also observed.

3.2. Short-time Fourier transform reveals oscillation frequency temporal evolution

The fast Fourier transform was previously applied for decomposing the signal oscillations of the Langmuir probe placed at several millimeters from the target surface.²⁴ At larger distance, *i.e.* for the present measurements, the collector temporal trace extends on a broader time domain, allowing us to record a higher number of oscillations. In this context, the use of the short-time Fourier transform (STFT) can reveal the variation of the oscillation frequency with time. The method is based on dividing the recorded signal into shorter time-windows and then separately computing the Fourier transform on each of them. We used a sampling interval of 1 ns, a Gaussian-type window of 256 points length, and 200 points overlapping.

In Fig. 2, in correspondence with the temporal traces of the FC ion current, the resulting STFT amplitudes are given as contour plots, for various retarding grid V_{RG} voltages in the range 70 - 110 V. The two oscillation frequencies manifest themselves more significantly when increasing the bias (as also shown in Fig. 1a), having values similar to the ones derived in Fig.

1b. Thus, we observe that the structure containing C_{fast}^{1+} ions oscillates between 10 μs and 20 μs , while the slow part containing C_{slow}^{1+} ions exhibits periodic features between 20 μs and 30 μs .

In comparison with the usual fast Fourier transform, using STFT allowed to observe more detailed features. Thus, the time-dependences of the frequencies show different trends: significantly decreasing for the fast part (*e.g.* in Fig. 2d from 12.5 MHz at 10 μs , to 10 MHz at 20 μs) and slowly increasing for the slow one (*e.g.* in Fig. 2d from 6 MHz at 20 μs , to 7 MHz at 30 μs). We note that similar results were found also for $LP=40$ and $LP=45$ cm, but with a lower accuracy (see Fig. S2, Supplementary Material). The physical mechanisms behind these behaviors (observed here for the first time) are not clear and they should be further theoretically and experimentally investigated. Here we can only speculate on a possible correlation with the time-dependence of the corresponding ion current region (*i.e.* the ion number density): decreasing with time for the C_{fast}^{1+} ions and slowly increasing for C_{slow}^{1+} .

3.3. Correlation between SG, RG and FC recorded currents

To get more insight on the origin of the oscillations, the signals of the suppression and the retarding grids were simultaneously recorded with the FC temporal traces. They are exemplified in Fig. 3a for $LP=42$ cm and $V_{RG} = (+70, +80, +90)$ V. We observed that the periodic structures occur in all recorded traces, with even higher amplitudes for SG and RG than for FC. This is an important observation, which allows us to consolidate the conclusion that the oscillations are intrinsic to the plasma plume and not induced by the EEA. The SG signal follows mainly the same structure as the FC one, with the exception of small differences in the early part. From their comparison, the FC temporal trace is slightly reduced, as a consequence of the fast ions repelling by the RG. For the RG signal, the multi-peak aspect is more clearly revealed, due to the contribution of different plasma parts and types of charges. At the beginning

it exhibits a negative current due to the excess of fast (escaping) electrons that were not completely repelled ahead by the suppression grid. This is followed by a positive part mainly consisting in slow ions, where the electrons are almost absent.

Fig. 3b displays the evolution of the SG, RG and FC total collected charge (temporally integrated current) with the V_{RG} bias. The decreasing trend of the FC integrated signal allows us to infer a shifted Maxwell-Boltzmann energy distribution: using the interpretation given by Küdyan,³³ the derivative with respect to the RG voltage leads to the energy distribution function on the plume axis (to be addressed in a separate paper). The total charge collected by the SG (Fig. 3b) is slowly influenced by V_{RG} , and only starting with about +50 V, possibly due the ions repelled by the RG. Finally, the current recorded by the RG is negative at early times and later on becomes positive, leading to a time-integrated null value (with some fluctuations, always staying below the total charge collected by SG and FC, Fig. 3c). Similar results are found for $LP=40$ and $LP=45$ cm (Fig. S3, Supplementary Material).

3.4. Discussion

The experimental results reported above bring additional support for a plume dynamics scenario built by our group over the last decade. This scenario involves splitting of the plume in fast and slow components and its structuring in multiple double-layers, in intimate relationship with the presence of two-temperature electrons. The occurrence of oscillations is seen as a genuine property of these peculiar plasma (see Ref. 18 and references therein). We note however that an alternative scenario has been proposed by another group,³⁴⁻³⁶ which assigns the MHz-range oscillations to low-frequency sheath instability. In this alternative scenario, the detected instabilities (oscillations) are associated with the interaction of the plasma flow (including an energetic ion component) with the electrostatic field of the analyzer, as the flow of energetic ions gets reflected inside the sheath of the probe. This space-charge effect

induced the perturbations in the probe potential and consequently the “disruption” of the incoming energetic ion flow, leading to oscillations (or rather multi-peak structure) in the electronic saturation current recorded by the probe.³⁴⁻³⁶

Although this alternative scenario seems very appealing for explaining the MHz oscillations in our work, we do believe that these oscillations are genuine manifestations of the charge separation in the plasma, rather than consequences of the plasma flow interaction with the probe sheath. This is based on several arguments. First, as described in Refs. 34-36, the sheath instability leads to oscillations only in the electronic saturation current, as the positive ions higher inertia is the basis itself of the instability (which vanishes when the Ar ions are partly replaced by lighter H, as described in Ref. 36). In our case, the oscillations are observed in vacuum for both electronic and ionic currents (see SG, RG, FC signals in the current paper) and even for the optical signal recorded by a photomultiplier tube (see Ref. 18). Although observed for a rather light ion (C) here, these oscillations occur also for much heavier elements (e.g. W, see Ref. 18). Moreover, no theoretical attempt is proposed in Refs. 34-36 to account for the recorded sheath instability frequencies. Our group has provided consistent efforts to offer a theoretical calculation of the observed frequencies (Ref. 18 and references therein). We have proposed an original fractal hydrodynamic model to explain plasma evolution at different stages,^{15,27} and current oscillations were discussed in the frame of a damped plasma oscillator in the collector electric field.^{23,24} Although not perfect nor universal, the experiment-theory agreement appeared as satisfactory at least for a number of elements^{15,23} (Al, Cu) and was confirmed by other groups in different experimental conditions.^{12,13,37} The new data presented in this paper, where oscillations are recorded for the first time at long distance from the target, should trigger further theoretical developments to complete the two-temperature multiple-double layer scenario. In these developments, a combination of the damped plasma oscillator and the energetic ions reflection in the sheath above a threshold bias can be interesting, as the

space-charge effects in the vicinity of the reflection point (*i.e.* retarding grid in our case) may indeed distort the electrostatic potential and produce some additional deceleration of the incoming ions. A theoretical modeling of the reflected ions and potential profile joint dynamics is therefore on our list of future investigations, with the benefit of using the current data for benchmarking.

4. Conclusions

We investigated the oscillations of the ion current recorded by an electrostatic energy analyzer (EEA) used to characterize the carbon plasma produced by ablating a graphite target with a KrF excimer laser under vacuum. The temporal traces reveal different frequencies for the early and late parts, which were assigned to the fast and slow expanding plasma structures, containing different types of ions. The fast plasma structure oscillates with an average frequency of ~ 12 MHz regardless of the retarding voltage, while for the slow structure the oscillation frequency exhibits a linear increase with V_{RG} , from 3 MHz to 5 MHz.

The short time Fourier transform (STFT) was successfully applied to the recorded signals to unveil the temporal variation of the oscillation frequency. The resulting STFT amplitudes confirm that the two oscillation frequencies manifest more significantly when increasing the retarding bias. The time-evolution of the frequency shows different trends for the two plasma structures: decreasing for the fast one and slowly increasing for the slow one.

By investigating the correlation between the currents recorded by the different electrodes of the EEA, we observed that the periodic structures appear in all recorded traces, allowing us to conclude that the oscillatory behavior is an intrinsic manifestation of the plasma plume dynamics, related to the formation of multiple electric double layers. This was discussed with respect to an alternative low-frequency sheath instability scenario involving the reflection of energetic ions,³⁴⁻³⁶ which can be also included in future joint theoretical developments using

as input the experimental data recorded here (at long distance from the target surface) to fully describe the oscillatory phenomena in connection with plasma parameters and to predict oscillation frequencies for a wide class of target materials.

Supplementary Material

The oscillations in the ionic and electronic currents of the carbon excimer laser-produced plasma recorded by the electrostatic energy analyzer are presented in supplementary material for two more additional laser fluences.

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Author's contributions

P. Nica and C. Ursu equally contributed to conceptualization, investigation and original draft preparation. C. Focsa contributed to discussion on results interpretation and to the original and revised manuscript writing.

Data Availability

The data that support the findings of this study are available from the corresponding author upon request.

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Figure captions

Figure 1. a) FC recorded signals for $LP = 42$ cm at various retarding grid voltages and the oscillatory part in different time-ranges (insets); a schematic representation of the EEA is given on the left side. b) Oscillation frequency evolution with retarding grid voltage (V_{RG}) for the fast and slow plasma structures (error bars $<1\%$ hidden by the size of the symbols).

Figure 2. Contour plots of STFT amplitudes and collector current for various retarding grid voltages: a) $VRG = +70V$, b) $VRG = +80V$, c) $VRG = +100V$, and d) $VRG = +110V$.

Figure 3. a) SG, RG and FC recorded currents for various retarding grid voltages ($V_{RG} = +70$ V, $+80$ V, $+90$ V) and b) the corresponding total collected charge *vs.* RG bias.















