Understanding multiple structure formation in laser-produced plasmas on Ag in various gases

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Introduction

With the continuous progress of high-power pulsed laser sources, the study of laser-matter interactions has become the core of developing new technologies and applications [1]. The ns-laser-matter interaction involves a complex blend of phenomena such as absorption of the laser beam by the target, heating followed by phase transformation, atoms and particle vaporization and ejection, plasma formation, expansion and interaction with the latter part of the laser pulse and gaseous atmosphere and chemical reactions in the expanding plasma. In the past years laser ablation has been successfully used for pulsed laser deposition (PLD) of thin films, nanoparticles and clusters production, or material sampling for elemental analysis [2–4]. For techniques like PLD, the deposition process usually takes place in an inert or reactive gas, which makes understanding the dynamics of the ablated material rather difficult. The presence of the background gas can be either explained by balancing the chemical reactions needed for the production of oxide, nitride or carbide films or for controlling the kinetic energy of the ablated particles at the impact with the gas molecules as well as surface of the growing film in order to avoid defects formation during deposition. In this paper we report on the dynamics of transient plasmas generated by ns-laser ablation of Ag in various ambient atmospheres. The floating probe measurements revealed the presence of a multi-peak current structure which is strongly dependent on the measurement angle and the background pressure.

Experimental Set-up

The plasma was produced in a vacuum chamber using a Continuum Surelite III Nd:YAG Laser (λ = 266 nm, E_n = 81 mJ, f = 10 Hz). The laser beam was focused on the rotating 6 mm thick Ag target (99.99% purity) at a fluence equal of 3.8 J/cm^2. The target to substrate and to Langmuir probe (LP) distances were 50 mm and 37 mm, respectively. The base pressure in the

vacuum chamber was 4×10^{-6} Pa, and the plasma investigations were performed under a residual vacuum of 5×10^{-5} Pa, and at 5×10^{-2}, 5×10^{-1}, 2, 5, and 10 Pa N_2, Ar and O_2 pressure. Before starting the investigation, the LP was shielded from the plasma area and the silver target was laser irradiated for 2 minutes (that corresponds 1200 pulses) to *in situ* clean its surface. The signal from LP was measured by acquiring the voltage signal across a load resistor with help of a digital phosphor screen oscilloscope Tektronix DPO 4140. The synchronization was realized by a fast silicon photodiode Thorlabs FDS100. The chronoamperometric measurements were performed at floating potential as well as at biased potential in the range from –20 V to +20 V.

Results and discussions

A novel way of investigating the plasma flow is the use of non-biased probe approach. When the probe not biased there are no accelerating fields in the collection region that would select charged particles with certain thermal energies. As the particles arrive unperturbed onto the probe surface, one can collect the plasma electrons and ions based on their arrival time and thus their kinetic energies. In Figure 1 (a) we represented charge particle temporal traces measured on the main expansion direction (0° – along the main expansion axis, which is perpendicular to target surgace) in ultra-high vacuum (10^{-5} Pa) conditions. We notice complex multi-maxima temporal distribution with contribution from both charges. The electronic current has two maxima attributed to two groups of electrons in the plasma, expanding with velocities of 70 km/s and 40 km/s while ions are split in three groups which move with relatively lower velocities of 23 km/s, 8 km/s and 2 km/s. The values are in line with the differences in the particle charge and mass as seen in the framework of an electrostatic ablation mechanism. The multiple structuring of plasma has been previously observed for other irradiation conditions [5,6] for single element target ablation being explained as a hydrodynamic effect induced by the interactions between the ejected particle and the background gas or induced by the different ablation mechanisms. The presence of a transient double layer in the framework of an electrostatic ablation mechanism requires the presence of different groups on ions expanding with different velocities. Similar data was reported in [7] where doubly ionized ions were traveling twice as fast as single ionized ions, a clear signature of an electrostatic mechanism. In this work the split is observed in the electron cloud as well and is an important feature of nanosecond laser produced plasmas: the structuring in multiple requires the formation of complete plasma structures containing electrons, ions atoms and photons. Figure 1 (b) displays the charge density velocity distribution in vacuum and for various N_2 atmosphere. By changing the pressure, we noticed a shift in the ionic maxima towards smaller expansion velocities, result correlated with the energy losses through collision and the overall confinement of the plasma.

Also, a transition from three-peak to two-peak distribution is seen as the plasma is confined. The addition of different gases could also lead to different outcomes: Ar gas is more susceptible to ionization and induces another ionic structure in the plasma flow affecting the properties of the thin film, while introducing reactive gases leads to molecule formation and intense chemical reaction within the plasma volume.

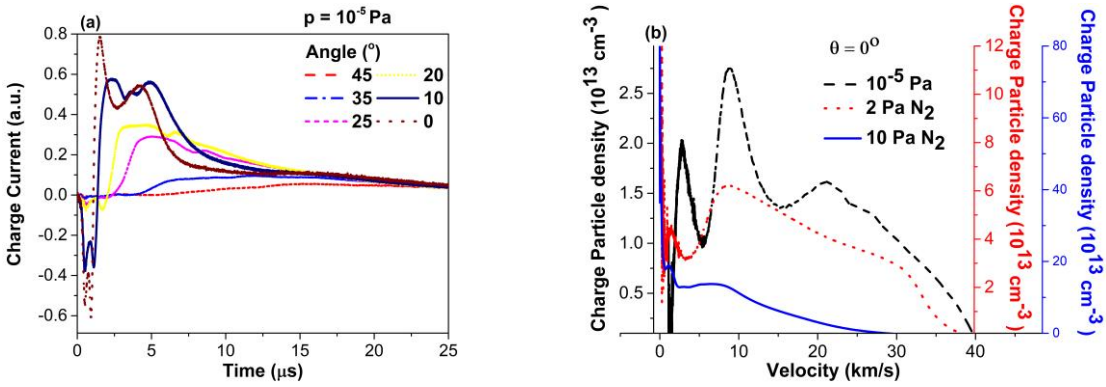

Figure 1. Angular distribution of the floating current trace recorded at 5×10^{-5} Pa (a) and charge density velocity distribution for various N_2 atmospheres (b)

Time-resolved LP analysis were performed on the main expansion axis (0°). When investigating the temporal distribution of electron temperature in high vacuum conditions (5×10^{-5} Pa) we noticed two different slopes in the logarithmic representation of the electronic part of I-V curve (Figure 2 (a)). This is a signature of two generations of hot and cold electrons. The result is in line with the two-temperature model which often lacks a strong empirical support from LP measurements.

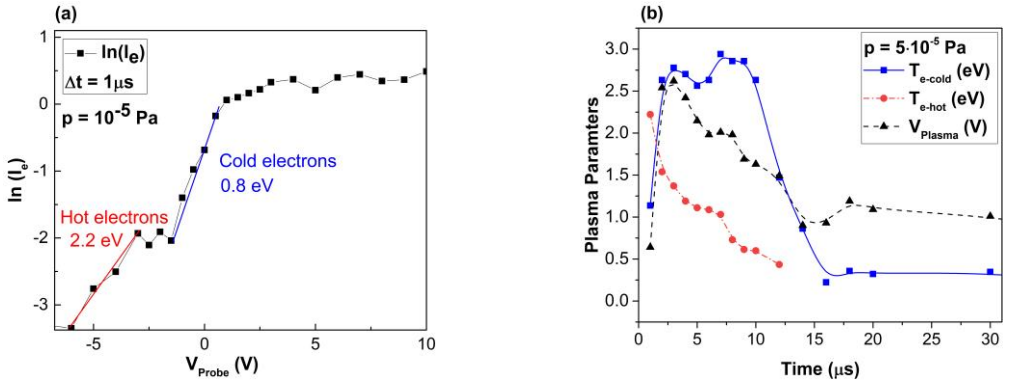

Figure 2. Logarithmic representation of the I-V characteristic of an Ag plasma at 1 μs and the temporal evolution of T_e (hot and cold) and V_{Plasma} calculated at 0°

The $T_{e\text{-}hot}$ decreases exponentially and is not observable at long evolution times (> 12μs) (Figure 2 (b) – red curve). $T_{e\text{-}cold}$ has an increase with a three-peak evolution, each peak describing one of the three plasma structures seen via floating current measurements (Figure 2 (c) – blue curve). The plasma potential follows a similar evolution as that of the cold electrons

which means that in this temporal regime the cold electron group is dominant (Figure 2 (c) – black curve).

Conclusions

Angular and time-resolved measurements were performed by implementing the LP technique for the investigations of transient plasmas generated by ns-laser ablation of Ag in various background gases. An attempt to understand the inner mechanism of ablation plasma dynamics by controlling the ionic energy distribution during the deposition process was reported. For each investigated pressure a time-resolved analysis was performed on the main expansion axis which allowed the determination of a wide series of plasma parameters and their temporal evolution. The use of non-biased probe allowed us to observe multiple structuring of the Ag laser produced plasmas and to reconstruct the ions energy distribution. The structuring is seen in the ionic part of the collected current and confirmed by the fast electrons distribution which matched the ionic one. Each structure characterizes an ionization state of the Ag ions, discussed in the framework of multiple double layer formation during plasma expansion.

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