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Laser-induced ablation: physics and diagnostics of ion emission

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Abstract. Pulsed lasers generating beams of different intensities may be used to produce ablation of solid targets placed in high vacuum and to generate pulsed plasma and ion acceleration. The plasma is in a non-equilibrium condition and in the first instant the particles being generated are subject to thermal interactions, to a supersonic gas expansion in vacuum and to a Coulomb acceleration due to the high electric field developed along the normal to the target surface. The ion diagnostics, based on time-of-flight technique, allow us to measure the mean ion energy, the total number of ions, as well as the ion energy and charge state distributions. The ion energy distributions may be described by the Coulomb-Boltzmann-Shifted (CBS) function, which after fitting to the experimental data may be used to determine the equivalent ion temperature and the accelerating voltage. Given the equivalent acceleration voltage and the plasma Debye length, it is possible to estimate the magnitude of the electric field developed in the plasma. Measurements of the ablation yield, plasma dimension and optical spectroscopy allow us to calculate the atomic and electronic plasma density and to evaluate the coronal plasma temperature. Some applications of the laser-induced ablation consist in the realization of laser ion sources (LIS), generation of multi-energetic ion beams by using a post-accelerating voltage, use of ultra-intense fs lasers to accelerate ions to energies of the order of tens MeV/nucleon. Other special applications include the pulsed laser deposition (PLD) of thin films, the laser ablation coupled to mass quadrupole spectrometry (LAMQS) probes, ablation of biological tissues, and generation of plasma for astrophysical and nuclear investigations.

Key words: laser-generating plasma • Boltzmann distribution • time-of-flight

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Introduction

In the last decade there had been growing interest in the field of the ion acceleration induced by the laser-generated plasmas. The laser pulses of duration 10 fs-10 ns and intensities exceeding 10^{10} W/cm^2, interacting with solid targets placed in vacuum, generate ionized vapor in a non-equilibrium condition, that has peculiar properties: high ion and electron temperature, high density, high ionization states, and high directive ion emission along the normal to the target surface. The fast electron emission and the slower ion emission, in fact, produce an intense electric field directed along the normal to the target surface. The kinetic energy of the ions emitted from the plasma generally has a Boltzmann distribution with a mean energy proportional to the laser intensity; it ranges between about 100 eV/charge state up to about 10 MeV/charge state, depending on the laser pulse intensity [3, 12].

Many plasma parameters increase with the laser intensity I, and the laser light wavelength, λ, according to a scaling law involving the square root of $I\lambda^2$ [3].

At low laser intensities ($\sim 10^{10}$ W/cm^2) the ablation of a thick target has a yield of the order of

1 μg/pulse, the fractional ionization is low, the maximum charge state is about 10⁺, the ion energy is of the order of 100 eV/charge state, the ion temperature about 100 eV and the maximum electron density of the order of 10^{16}/cm^3 [12].

At medium laser intensities (~ 10^{15} W/cm^2) the ablation yield of thick targets is of the order of 1 μg/pulse, the fractional ionization is about 95%, the maximum charge state is about 50⁺, the ion energy is about 100 keV/charge state, the ion temperature is of the order of 10 keV and the maximum electron density is of the order of 10^{20}/cm^3 [6].

At very high laser intensities (~ 10^{19} W/cm^2), when the pulse of tens of fs duration is used and the irradiated target is thin (10–100 μm), the ablation yield depends on the laser spot diameter and the target thickness, and generally it is below 1 μg/pulse, while the fractional ionization is 100%, the maximum charge state is about 60⁺, the ion energy in the forward direction is about 1–10 MeV/charge state, the ion temperature is above 10 keV and the maximum electron density is of the order of 10^{22}/cm^3 [3].

Generally, the ion energy distributions follow a CBS distribution, i.e. the Boltzmann shape is shifted towards the high energy by increasing the ion charge state [6]. The experimental energy shift may be regular and it allows one to evaluate the acceleration voltage developed by the space charge in the plasma. By assuming that this voltage develops on a distance comparable with the plasma Debye length, it is possible to evaluate the electric field accelerating ions. Its value is of the order of tens MV/cm in the case of low laser intensities, of the order of tens GV/cm in the case of medium intensities, and even higher for the highest laser intensities [11].

The ion acceleration via laser-generated plasma finds interesting applications in many fields: the laser ion sources, post-ion acceleration, ion implantation of multi-energetic ion beams, astrophysical and nuclear applications, biomedical applications such as therapy, diagnostics and preparation of biomaterials.

Materials and methods

The main experiments had been performed using the Nd:YAG laser at the Istituto Nazionale di Fisica Nucleare-Laboratori Nazionali del Sud (INFN-LNS, Catania, Italy) and the iodine laser at the Prague Asterix Laser System (PALS) laboratory in Prague, Czech Republic. The first of those lasers may be operated at 1064, 532 and 355 nm, with 9 ns pulse duration, 1 J maximum pulse energy and intensities between 10^8 and 10^{11} W/cm^2. The second may be operated at 1315 and 438 nm, with 300 ps pulse duration and 800 J maximum pulse energy, with intensities between 10^{13} and 10^{16} W/cm^2.

In order to verify the scaling of the results as a power $I\lambda^2$ [3] we used also data at intensities higher than this limit, which we collected from the literature and compared with our measurements.

Time-of-flight (TOF) measurements had been performed, accompanied by ion collectors (IC) and an electrostatic deflector ion energy analyzer (IEA). They permit to measure the average ion energy and the ion energy and charge state distributions, respectively. Details of the IC and IEA are given in Ref. [14].

The experimental ion energy distributions were fitted with the CBS function:

$$F(t) = A\cdot\left(\frac{m}{2\pi kT}\right)^{3/2}\cdot\left(\frac{L^4}{t^5}\right)\cdot\exp\left[-\left(\frac{m}{2kT}\right)\left(\frac{L}{t}-\sqrt{\frac{\gamma kT}{m}}-\sqrt{\frac{2zeV_0}{m}}\right)^2\right] \tag{1}$$

where m is the ion mass, kT is the equivalent temperature, L is the target-IC detector distance, γ is the adiabatic coefficient, ze is the ion charge and V_0 is the equivalent acceleration voltage developed in the non-equilibrium plasma [8].

The ion plasma temperature, T_i, was measured though the CBS fit of the experimental ion energy distributions; the electronic plasma density, n_e, was measured through the evaluation of the atoms removed from the laser crater, the charge state distribution and the volume of the visible plasma observed by a fast CCD camera [2]. Figure 1 shows a typical CCD image of the plasma of tantalum ions at 1 μs exposition time (a) and the corresponding crater depth profile obtained with 820 mJ pulse of 9 ns duration, at 1064 nm, in the tantalum target (b). Assuming the fractional ionization to be 50% and the mean charge state to be 2⁺, the electron density corresponds to 1.8×10^{17} electrons/cm^3. Both

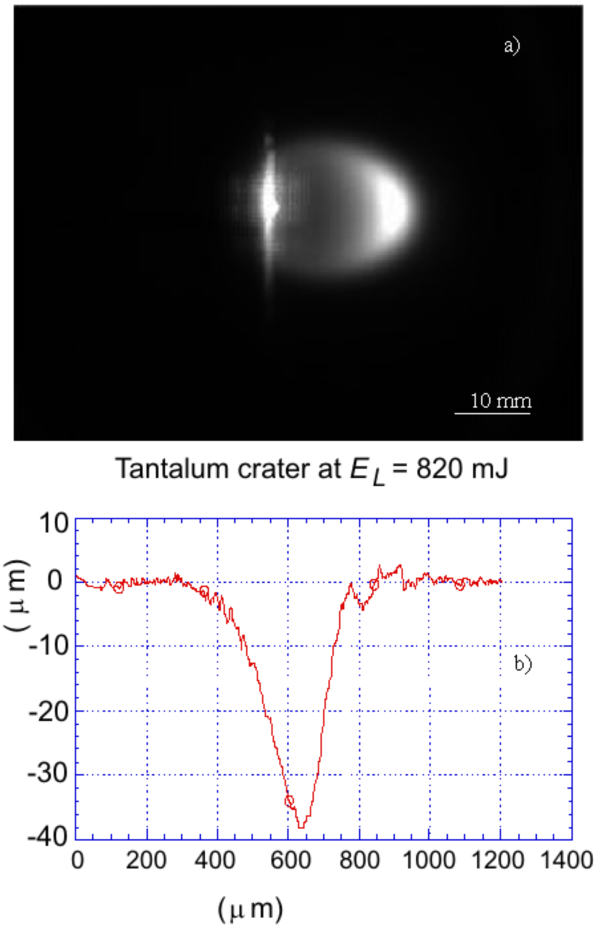

Fig. 1. Photograph of the Ta plasma expansion in vacuum (1 μs exposition time) (a) and the corresponding crater profile generated by 820 mJ, 9 ns, 1064 nm laser pulse (b).

parameters, T_i and n_e, were also measured though the optical spectroscopy by using the plasma emission lines of visible, UV and X-ray photons [2].

The mass quadrupole spectrometer (MQS), with 1–300 amu mass range, was employed to measure the elements and chemical compounds emitted from the plasma in vacuum; the surface profiler (SP) Tencor-P10, with 10 ns depth resolution and 10 μm lateral resolution, was employed to analyze the crater morphology as a function of the laser intensity; the Thomson parabola spectrometer (TPS) was employed to deflect electrostatically and magnetically the ions ejected from the plasma, in order to measure their energy, charge state and mass.

Results

Typical TOF spectra are reported in Fig. 2, as observed via IC (left) and IEA (right) detection at low (top) and high (bottom) intensity. Figures 2a and 2b refer to H^+ and C^{n+} ions ejected from the plasma produced by the laser interaction with a polyethylene target, and to Au^{n+} ions generated from the plasma produced by the laser ablation of gold, respectively, both at an intensity of 10^{10} W/cm^2. Due to the 1.3 m IC-TOF length, the proton energy is about 100 eV, and the carbon energy at the carbon peak is about 1 keV. Due to the 1.6 m IEA-TOF length, the Au^{6+} kinetic energy corresponds to about 4.6 keV. The spectra reported in Figs. 2c and 2d are refer to the ions detected from ablating Ta targets at 10^{15} W/cm^2 pulse intensity. In this case the IC and IEA TOF length was 1.6 m, the corresponding proton energy was about 2 MeV, the maximum Ta charge state was 48^+ (Fig. 2d) and the maximum Ta ion energy was about 22 MeV.

By changing the energy-to-charge ratio of the IEA spectrometer it is possible to measure the ion energy distributions, at low and high laser fluence, as reported in Fig. 3.

This figure shows that the experimental data points referring to Au (a) and Ta (b) laser ablation, at 10^{10} W/cm^2 and 10^{15} W/cm^2, respectively, may be fitted with the CBS distribution. The input parameters of the fit are the equivalent ion temperature, T_i, and the acceleration voltage V_0.

Measurements of Ta core electronic temperature gave values of the order of 100 eV and 10 keV for the 10^{10} W/cm^2 and 10^{15} W/cm^2 laser intensities, respectively, and V_0 values of the order of 700 V and 14 kV, respectively. The electronic densities in these two cases, at about 1 μs exposition time from the laser shot, are about 10^{16}/cm^3 and 10^{19}/cm^3, respectively. The corresponding values of the Debye length, λ_D, are 0.74 μm and 0.23 μm, respectively. Thus the electric field in the Ta non-equilibrium plasma, which is of the order of the ratio V_0/λ_D, assumes the values of 9.5 MV/cm and 600 MV/cm, respectively at low and at high laser intensities.

The Thomson parabola spectrometer gives information about the different contributions of the ion emis-

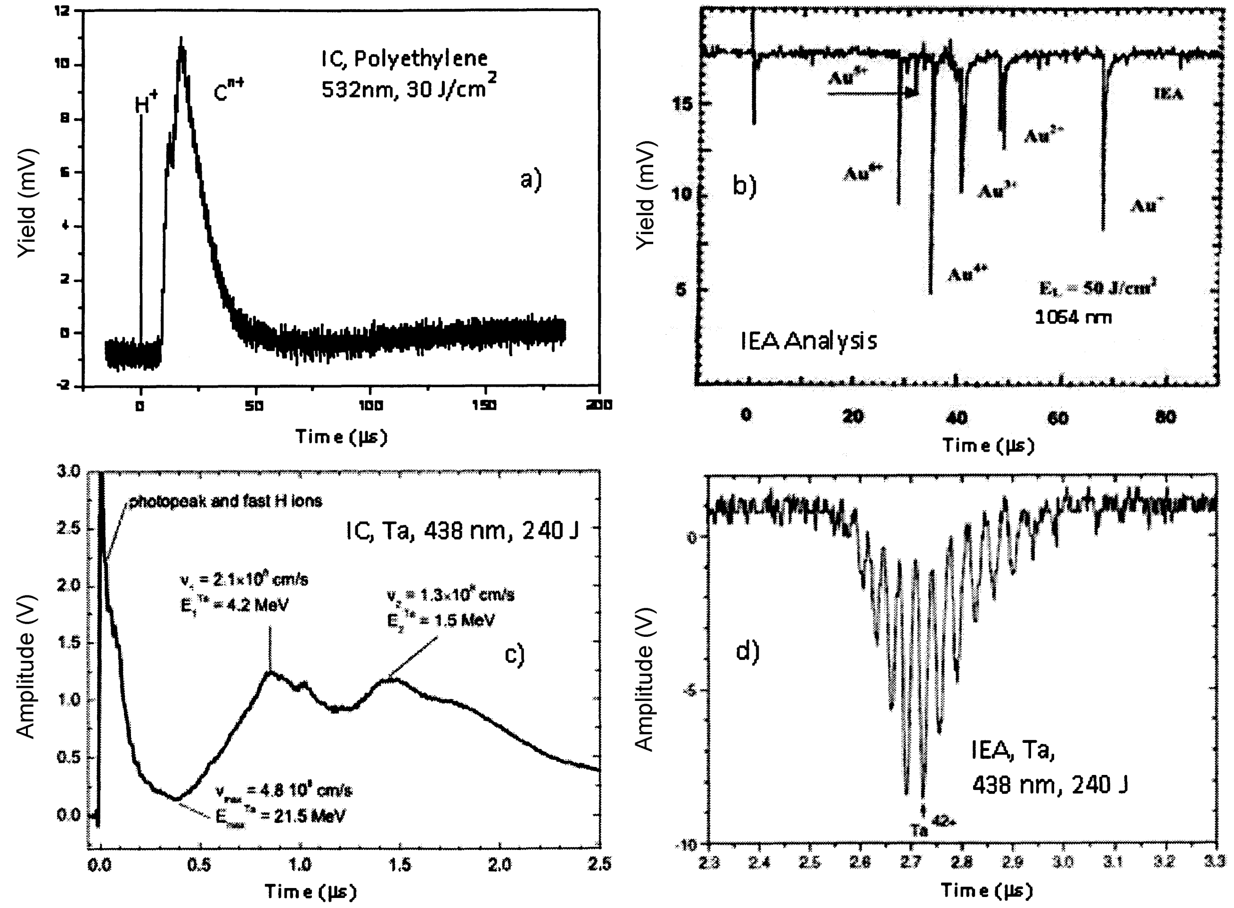

Fig. 2. IC (a) and IEA (b) spectra of the ions ejected from plasma generated at 10^{10} W/cm^2 by ablating polyethylene (a) and gold (b), and IC (c) and IEA (d) spectra of the ions ejected from plasma generated at 10^{15} W/cm^2 by ablating tantalum targets.

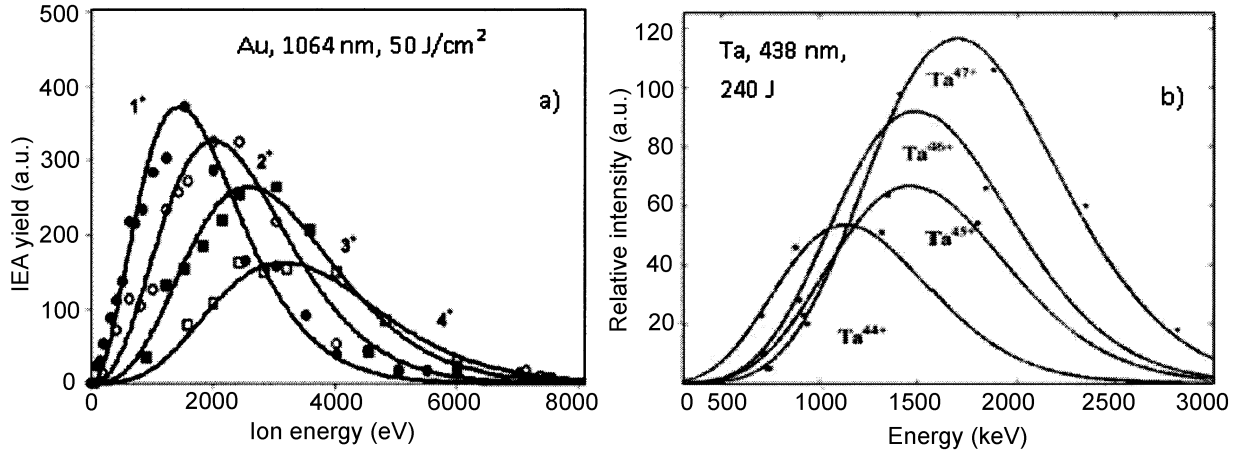

Fig. 3. Typical ion energy distributions for the Au target, at 10^{10} W/cm^2 (a) and the Ta target, at 10^{15} W/cm^2 (b).

sion from plasma in a single laser shot. Figure 4 shows a photo of the TPS's geometry of PALS laboratory (a) and a typical TPS spectrum of the protons and carbon ions ejected in the forward direction and along the normal to the target surface, from a plasma obtained by irradiating a thin (100 μm) polyethylene film at the PALS laboratory by using a laser pulse intensity of 10^{15} W/cm^2 (b). The parabolic lines, detected with a fast CCD camera placed on the focal TPS plane, correspond to different ions, charge states and ionized C_xH_y heavier groups emitted from the plasma. The maximum proton energy of about 400 keV measured with TPS was confirmed by IC measurements. The presence of carbon ions and heavy C_xH_y molecules was confirmed also by the contemporary MQS analysis of the ejected particles during the polyethylene laser ablation [13].

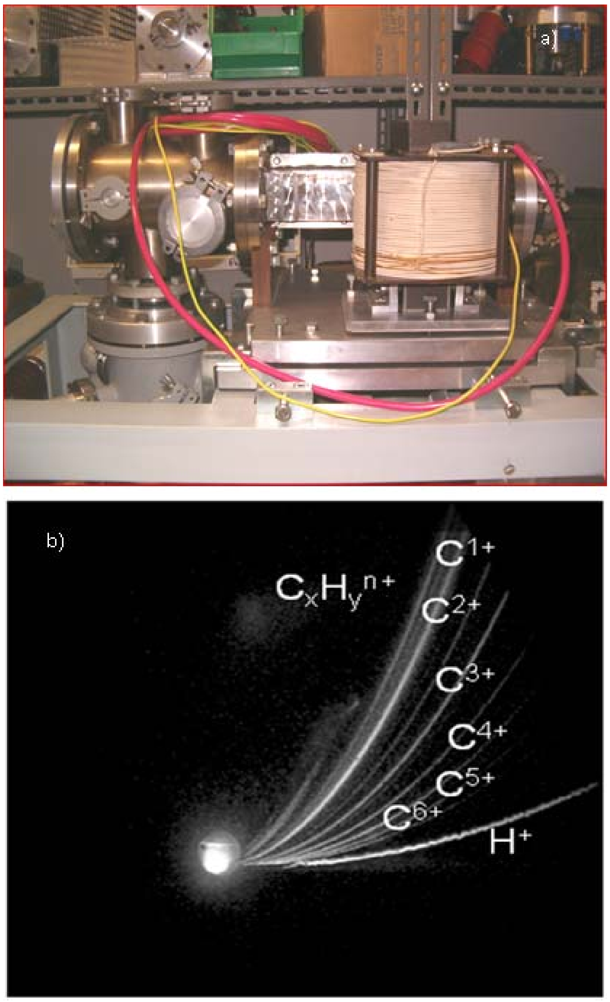

Fig. 4. Photograph of the TPS used at PALS (a) and a typical TPS spectrum of the ions ejected from the plasma generated at 10^{15} W/cm^2 (b) along the normal to the target direction, obtained by irradiating a polyethylene target.

Applications and conclusions

The applications of the laser-generated plasmas are numerous and cover several disparate fields of interest. The main applications that are being investigated at the INFN-LNS of Catania and at the Physics Department of Messina University are the following:

- Laser ion source (LIS): Their advantage is that they may generate ions from any atomic species, also from elements with high boiling point, producing high charge states and high currents. By using 1–10 Hz laser repetition rates and roto-translating thick targets it is possible to extract continuously current densities of the order of 100 mA/cm^2 [4].
- Post ion acceleration (PIA): It is a simple method to accelerate the ion species, produced by the LIS, with a kinetic energy proportional to the ion charge state. The PIA technique produces a high current, multi-energetic and multi-species ion beam. Generally such beams are obtained by employing a post-acceleration voltage of about 30–60 kV [10].
- Multi ion implanter (MII): a LIS coupled to the PIA allows for the generation of a multi-energetic ion beam, which is very useful for ion implantation in many substrates (metals, semiconductors, insulators). The chemical and physical properties of the surface (wear, hardness, wetting, roughness, chemical reactivity, optical and electrical properties, etc.) may be modified by the implanted ion doses. Ions are implanted at different depths depending on their kinetic energy and mass. Ion doses of the order of 10^{15}/cm^2 may be obtained in about 1 h with 2 Hz repetitive laser pulse irradiating roto-translating thick targets [10].
- Ultrafast ion acceleration (UIA): Terawatt lasers, generating laser pulses of tens of fs duration and intensities of the order of 10^{20} W/cm^2, may be focused

on thin hydrogenated targets to generate, in the forward direction, light ions (protons, carbons,…) of high energy, above 10 MeV/charge state. This new method of ion acceleration, so-called TNSA (transverse normal sheath acceleration), is of special interest in the fields of the new generation of particle accelerators, nuclear plasma physics and astrophysical plasma simulations [3].

- Pulse laser deposition (PLD): This application is about 30 years old. It is traditionally employed to deposit in vacuum thin and highly adherent films on different substrates. Crystal growth and special nanostructured films (diamond like, oxides, biocompatible structures, transparent thin films,…) with peculiar physical and chemical properties may be deposited on hot substrates [7].
- Laser ablation coupled to mass quadrupole spectrometer (LAMQS): A controlled low intensity laser (~ 10^8 W/cm^2) pulse with a repetition rate (1–10 Hz) removes, in high vacuum, molecular mono-layers from the surface of the target; a coupled mass quadrupole spectrometer allows to analyze the removed masses and the relative amount of the detected species. The technique may be applied to characterize the chemical composition and the isotope ratios of the sample surfaces of different nature. A special application of this technique is found in the fields of the microelectronics and archaeology [9].
- Bio-medical applications: pulsed laser may be applied in different field of bio-medicine for therapy, diagnostics and for preparation of biocompatible surfaces. Low intensity lasers are employed to induce thermal and photochemical effects in biological tissues and in biocompatible materials; high intensity lasers are employed to induce ablation and shockwave pressure on different tissues [5]. Recent there research proposals were formulated involving terawatt fs lasers, which could be used to irradiate a thin hydrogenated film in order to extract 60–100 MeV protons, useful in the field of the proton-therapy.
- Nuclear fusion applications: the pulsed lasers accelerating deuterium up to energies above 100 keV (directly by hot plasmas or indirectly by post acceleration processes) may be employed to induce D-D and D-T fusion processes with a significant value of nuclear cross-section. In special laser and target configurations this interaction could be used to ignite the fusion process, in order to create a nuclear fusion energy source [1].

In conclusion, the problems of laser-induced ablation, plasma generation, ion acceleration and the corresponding diagnostics constitute a new field at the frontier of physics, very interesting not only from the point of view of studies of the fundamental mechanisms of non-equilibrium plasma physics, but also because of useful applications in different fields of advanced research.

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