TOF ION SPECTRA DECONVOLUTION FOR LASER-GENERATED PLASMAS

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Abstract

A study of different targets (Fe, Ti, Ni, Al_2O_3) ablation, in vacuum, by using a ns Nd:YAG laser radiation, 1064 nm and 532 nm (second harmonic) wavelengths, is reported. Laser pulse with high intensity generates a plasma at the target surface, with high non-isotropic emission of neutral and ion species, mainly emitted along the normal to the target surface.

Time of flight (TOF) measurements are performed by using an ion collector consisting of a collimated Faraday cup placed along the normal to the target surface and an Ion Energy Analyzer (IEA) detector. The TOF spectra are converted as a function of the ions velocity and they are deconvolved for the various ion charge states by using the "Coulomb-Boltzmann shifted" function approach through the "Peakfit" mathematical code.

The fit of the experimental distribution data permits to estimate the equivalent plasma temperature and the average energy shift of the distributions as a function of the ion charge state. This energy shift leads to the evaluation of the electric field producing the ion acceleration inside the plasma.

INTRODUCTION

A pulsed laser beam irradiating solid targets placed in vacuum induces thermal processes leading to material sublimation and ionization effects on the expanding vapour. The laser-generated plasma can be investigated by different techniques, such as optical spectroscopy, mass spectrometry, Langmuir probe, charge collectors, etc. [1].

The pulsed laser ablation (PLA) technique is employed for several purposes: ion generation, deposition of thin films, X-ray production, ion implantation, etc...

A special interest of PLA concerns the high-temperature plasma production where high charge state ions are involved. The extraction of the ions from the plasma makes it possible to inject them into special ion sources of ion accelerators. This process permits to increase the ionization efficiency and the ion extraction current of the traditional ion sources, as recently developed at INFN-LNS of Catania permitting to inject ions into an electron cyclotron resonance (ECR) system [2].

PLA is also used to grow thin films in vacuum, as coverage of different substrates. The film properties, such as stoichiometry, roughness, grain size, crystallinity and porosity, can be modified on the basis of the used laser characteristics (wavelength, pulse intensity and width, etc.), substrate nature, irradiation environment conditions, etc. [3].

The technique is useful in many scientific fields, such as microelectronics, chemistry, biomedicine and metallurgy.

Our investigations concern the ns-laser ablation in vacuum of different targets and the evaluation of the plasma temperature and equivalent acceleration voltage with time of flight (TOF) measurements. The experimental ion velocity distributions follow Boltzmann functions and the deconvolution process through the Peakfit mathematical code permits to determine such plasma parameters.

EXPERIMENTAL SETUP

A Nd:YAG laser operating at the INFN-LNS of Catania, with 1064 nm (first harmonic), 9 ns pulse width and 1 to 900 mJ pulse energy was used. It works both at 30 Hz repetition rate and in single shot mode. The laser beam is focused, through a 50 cm focal lens placed in air, on the target placed inside a vacuum chamber at 10^{-6} mbar pressure. The laser light through a thin glass window hits the target with an incidence angle of 45°. The laser spot has 0.7 mm^2 dimension, on the target surface, at 0° incidence angle.

A cylindrical electrostatic ion energy analyzer (IEA) is mounted at 45° along the normal to the target surface [4]. The IEA contains a special ion collector (ICR) at the input (60 cm distance from the target), having a high transmission factor, and an electron multiplier placed behind the deflector plates, at 1.5 m total distance from the target. Both IC and IEA detectors permit to perform ion time-of-flight (TOF) measurements and their spectra were converted as a function of the ions velocity in order to perform the fit with the Coulomb-Boltzmann-shifted functions.

A Nd:YAG laser operating at 532 nm (second harmonic) with 3 ns pulse duration, maximum energy of 150 mJ, at single pulse or repetition rate mode (10 Hz), was employed at the Plasma Physics Laboratory of the Messina University to irradiate a target placed inside a high vacuum chamber (10^{-6} mbar), in which the laser beam interacts with the target surface with a laser spot of 1 mm^2 dimension at 0° incidence angle. The incident laser beam, focused through a lens placed in air (70 cm focal length), crosses a glass window and hits the target surface. The incidence angle of the laser beam was 45°.

A special mass quadrupole spectrometer, the Hiden Electrostatic Quadrupole Plasma (EQP) 300, was employed to monitor the particles ejected from the plasma with a mass ranging between 1 and 300 amu. It was placed along the normal to the target surface, i.e. at 45° angle with respect to the incidence laser beam.

An ion collector (IC), consisting of a Faraday cup with electron suppression grid, was employed in time-of-flight (TOF) configuration in order to measure the mean ion kinetic energy. IC spectra were converted as a function of the ions velocity and they were deconvolved in different Coulomb-Boltzmann-shifted distributions, for the different ion charge states; this procedure was obtained through the Peakfit code [5].

Fig. 1 shows a photo of the INFN-LNS experimental set-up (a) and a photo of the experimental set-up at the Messina University (b).

RESULTS

Fig. 2a (bottom) shows a typical IEA spectrum obtained irradiating the Fe target at 1064 nm wavelength and 335 mJ laser pulse energy and detecting ions with 100 V plate bias, corresponding to a filtering of E/z = 1 keV/charge state. The negative peaks indicate that six charge states, identifiable through the different TOFs [6], are produced and that the TOF time decreases with the charge state, i.e. the ion velocity increases with the z charge. On the top of Fig. 2a the ICR spectrum relative to the ion yield detected along the IEA direction is shown. This spectrum represents the ion yield emitted at 0° angle with respect to the normal direction to the target surface. The photo-peak start signal, for both spectra, occurs at zero time. Converting the TOF spectra in velocity spectra, as reported in a previous work [7], the ICR experimental data appears so as reported in Fig. 2b. In this figure the experimental ion velocity distribution is indicated with open points and, assuming the Fe ions to be detected from charge state 1+ up to charge state 6+, in agreement with the IEA measurements, a deconvolution process can be approached. This mathematical calculation shows the single ion contributions to the final convolution of detected ion yields. The deconvolved peaks have been obtained assuming that their shape follows a "Coulomb-Boltzmann-shifted" function:

$$f(v) = A\left(\frac{m}{2\pi K_B T}\right)^{\frac{3}{2}} v^3 \exp\left[-\left(\frac{m}{2K_B T}\right)(v - v_K - v_c)^2\right]$$

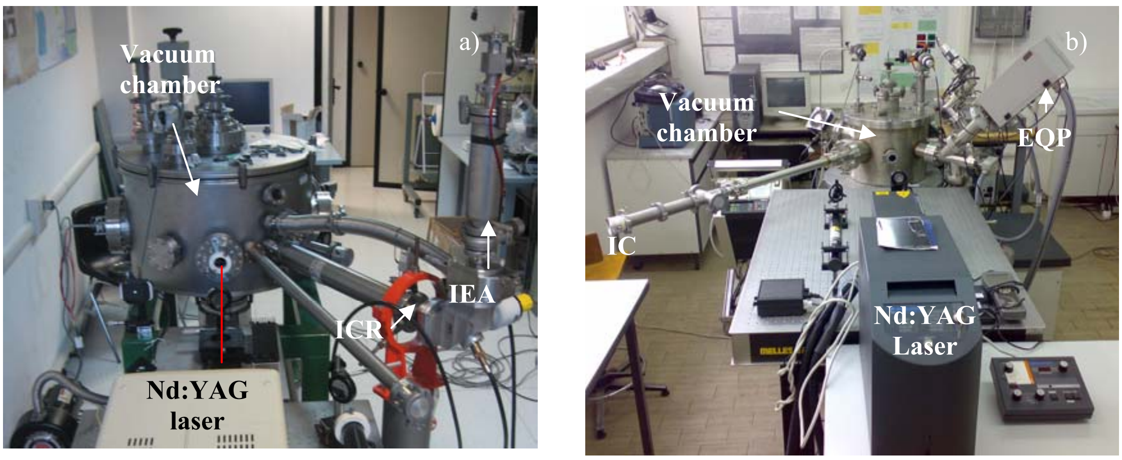

Fig. 1. Photo of the INFN-LNS (a) and of the Messina University (b) experimental set-ups.

where A is a normalization constant, m is the ion mass, k_B is the Boltzmann constant, T is the ion temperature, v is the total velocity along the normal direction to the target surface, v_k is the adiabatic expansion velocity and v_c is the Coulomb velocity [8].

The fitting parameters of the experimental data determine the plasma equivalent temperature and the different components of the ion velocity. The equivalent temperature obtained with a laser pulse energy of 335 mJ is $K_BT = 46$ eV.

The regular shift of the ion velocity distributions, observable in Fig. 2b, indicates that an equivalent ion acceleration voltage V_0, of the order of 90 V, is generated inside the non-equilibrium plasma.

Typical TOF spectra for Al_2O_3 irradiation at 532 nm are shown in Fig. 3a for low and high laser fluence irradiation, respectively.

a)

b)

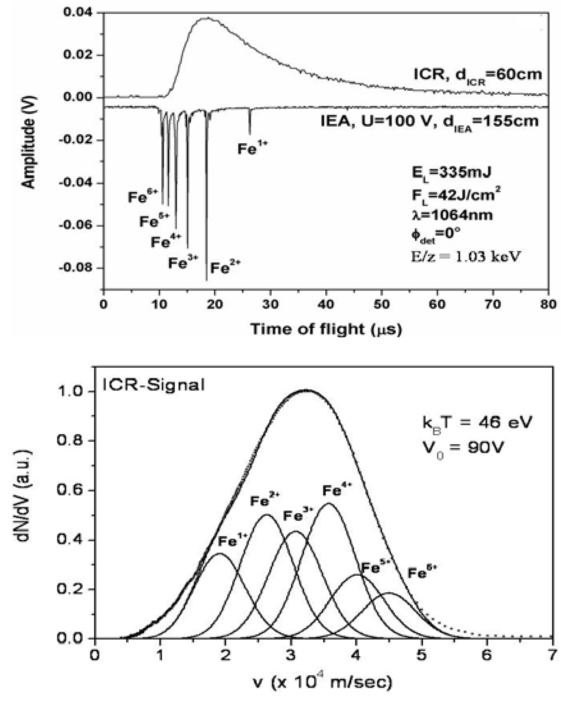

Fig. 2. A typical IEA spectrum (bottom) and ICR spectrum (top) (a) and the experimental ion velocity distribution (b).

Fig. 3a shows that, at a fixed target-IC distance of 72 cm, the time-of-flight separation between aluminium and oxygen ions is evident at the laser energy of 150 mJ. This separation disappears when the laser energy is reduced to 25 mJ because the signal yields decrease strongly.

The IC spectrum obtained ablating the alumina target at 150 mJ laser energy is reported in Fig. 3b in terms of ion velocity distributions (full lines) obtained by a deconvolution process of the IC signal (dots curve).

The IC measurements are in agreement with ion energy distributions of the multiple ionized species, according to the energy shift given by the Coulomb-Boltzmann-shifted theory. The regularity of the shift with the charge state confirms the existence of an equivalent voltage developed in the plasma, which accelerates the ions towards the target normal direction [9]. The experimental voltage value is 510 V at a laser pulse energy of 150 mJ. The fit of the experimental results with the Coulomb-Boltzmann-shifted function indicated also an equivalent plasma temperature of 15 eV for Al_2O_3 plasma, for high laser fluence.

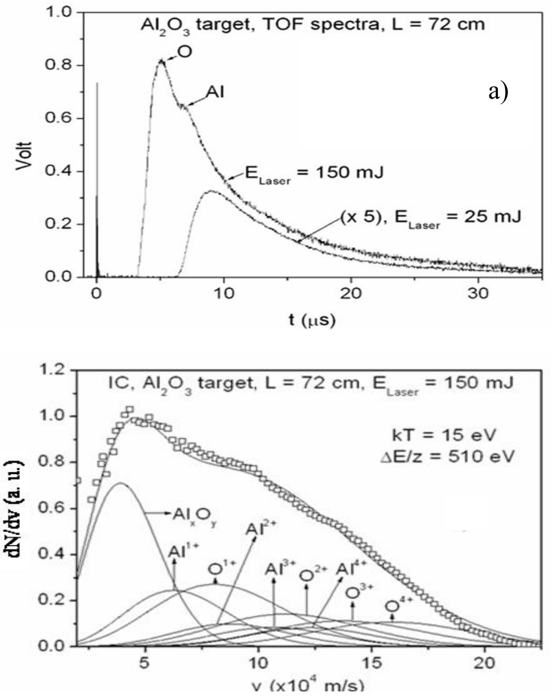

Fig. 3. Typical Al_2O_3 TOF spectra at 532 nm for high and low laser fluence irradiation (a) and the ion velocity distribution at 150 mJ laser energy (b).

The deconvolution process has been made by using the elements detected with the MQS, as shown in Fig. 4, that reports in detail a comparison

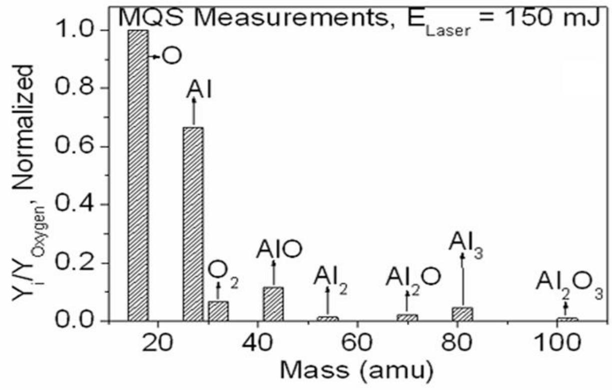

Fig. 4. A comparison of the MQS signal peaks yield relative to that one

of the MQS signal peaks yield relative to that one of the oxygen (16 amu) for the different detected elements. High peak yields of the masses 16 (O), 27 (Al), 43 (AlO) and 81 (Al_3 atom group) are detected. The Al_2O_3 peak yield at 102 amu is lower than the ones of the other emitted species.

DISCUSSION AND CONCLUSIONS

The time of flight (TOF) measurements performed by using the ion collectors gave TOF spectra, that have been converted as a function of the ions velocity and deconvolved for the various ion charge states by using the "Coulomb-Boltzmann shifted" function approach through the "Peakfit" code.

The fit of the experimental distribution data permits to estimate the equivalent plasma temperature and the average energy shift of the distributions as a function of the ion charge state. The regularity of the energy shift with the charge state for the plasma ions detected by the ion collectors indicates that they are submitted to a Coulomb acceleration due to a high electrical field self-generated in the non-equilibrium plasma. The most confirmed model supposes that the electric field is due to a space-charge separation between the fast electrons and the slow ion clouds, respectively, ejected from the target along the normal direction.

The calculation of this field is influenced by many parameters such as the laser irradiation conditions, the used electron temperature and density and the LTE conditions, only approximated due to the non-equilibrium involved phenomena. Moreover, the temperature and the density depend strongly on the time and on the space and, consequently, the Debye length too; thus the electric field is not well localized in time and in space at this stage.

Further investigations are in progress to evaluate in detail this possible kinetic mechanism of electric field generation [10].

A work is in progress to explain the great difference between the metallic and ceramic ion equivalent acceleration voltage values at the same laser irradiation conditions.

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