Diagnostics of plasma plume produced by laser ablation using ICCD imaging and transient electrical probe technique

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Abstract. The dynamics of transient plasmas generated by high-fluence nanosecond laser ablation has been investigated by means of fast ICCD imaging and electrical probes in transient regime. Measurements have been carried out on plasmas produced in vacuum (5×10^{-6} Torr residual pressure) by a pulsed Excimer laser (20 ns, XeCl, λ = 308 nm) irradiating stainless steel targets. Two plasma expansion velocities were estimated, one from probe measurements and another from recorded ICCD images. Electron plasma temperature (0.1 – 0.3 eV) and density (1 – 5×10^{11} cm^{-3}) were measured by electrical probes for a laser radiation power density of 6×10^8 W/cm^2.

1. Introduction

Nanosecond laser ablation is a complex process, which implies interactions between laser radiation and target materials [1], between a laser beam and an ablation plasma plume [2,3], and, finally, between ablation cloud particles [4,5]. Experimental methods, theoretical models and numerical simulations were used to describe the laser-target interaction and the dynamic of the particles ejected and the plasma plume formed after laser irradiation on a solid target [6–8]. The early stage of the ablation process and the space and time dynamics of the involved species were investigated, so that useful information were obtained for improving the technology for thin layer deposition of materials under special conditions of purity.

In the current paper experimental results are presented and discussed on the dynamic properties of the plasma plume produced by so called laser ablation of a metallic target. The diagnostic techniques used were fast ICCD imaging and electrical probes in transient regime. The ICCD images can register the instantaneous spatial distribution of the light emission of the plasma plume at different moments of its temporal evolution. Plasma parameters as electron density and temperature were measured using electrical probes. The light intensity profiles recorded by the ICCD camera and the electrical probe measurements allow identifying the two stages of the plasma plume expansion with different velocities.

2. Experimental set-up and measuring circuits

The experimental set-up with the ICCD measuring circuit is schematically shown in figure 1. The laser radiation of a 20 ns excimer pulsed laser (XeCl, λ = 308 nm, 10 Hz frequency) beam was focused by an f = 18 cm lens into a stainless steel target placed in a cylindrical vacuum chamber (20 cm in diameter and 15 cm in height). A vacuum system (preliminary and turbomolecular pump)
pumped down the chamber to a $5 \times 10^{-6}$ Torr residual pressure. Due to the technical parameters of the laser design and of the optical system used, the laser beam was focused onto a line with length of 1.5 mm and thickness of 0.3 mm (an area of 0.45 mm$^2$). Before each experiment, the laser beam energy has been measured with a Joulemeter. The energy usually used was 50–55 mJ/pulse, which leads to a typical laser intensity of $\sim 6 \times 10^8$ W/cm$^2$.

Both the formation and the dynamic of the plasma plume have been studied by means of a Hamamatsu digital camera (model C848-O5G) coupled with an image intensifier (model 9546-03) placed perpendicular to the plasma expansion direction. A signal generator was used as a master, which may control the delay between different measuring channels and to trigger the laser and the ICCD. Thus, the ICCD can acquire the total light emitted by plasma plume at different time instants during the temporal evolution of the plasma plume.

Figure 1 shows schematically the experimental set-up with the probe measuring circuit. The heated cylindrical probe was a tungsten wire of 0.23 mm in diameter and 3 mm in length. The probe was movable both radially (from 1.5 cm to 4 cm away from the target surface) and azimuthally ($\theta$ ranged from 10 degree to 170 degree).

The time evolution of the probe current for a constant bias of the probe was recorded by a Tektronix TD2500 oscilloscope and then transferred to a PC for further analysis. The bias voltage of the probe was changed in the range form $-10$V to $+10$V with respect to the target and the vacuum chamber. These probe signals recorded along time were used to reconstruct the probe characteristic at a certain moment of the time evolution of the plasma plume. The total time required for recording the signals needed for the reconstruction of a probe characteristic was about 30 minutes.

A double probe system was also used to detect and to measure the internal electric field within the plasma plume. Each probe of the system was made from tungsten wire of 0.23 mm in diameter and 3 mm in length. The time evolution of the potential for each probe was also recorded using the Tektronix TD2500 oscilloscope and then transferred to a PC for analysis.
3. Results and discussion

3.1. ICCD imaging

The spatial distribution of the light emission produced by the plasma plume at different instants of time of the plasma plume evolution was found from the ICCD images. Each image was obtained for a separated laser shot. Because of that, special attention has been paid to keep similar conditions for the laser ablation process at each laser shot. In this view, an equal energy of 55 mJ was used for each laser pulse. Moreover, by a continuously target spinning around the axis each shot send the laser beam on a fresh target surface. Each picture was taken for a gate time duration of 200 ns. A set of such ICCD images taken at certain moments of the plasma plume is presented in figure 3. Because the images were recorded with different setting of ICCD gain, current, and exposure time, these images show only a qualitative evolution of the plasma plume.

![Figure 3. Temporal evolution of the plasma plume (ICCD gate time – 200 ns). Different images were recorded for successive laser pulses of equal energy (55 mJ/pulse).](image)

![Figure 4. Spatial light intensity profile (1 μs after laser shot).](image)

![Figure 5. Dependence of the distance \( r_m \) given by the maximum value of light intensity versus time.](image)

The pictures from figure 3 were used to find the spatial profile of the plasma plume light intensity. Thus, using free software (ImageJ), the spatial profile on the direction normal to the target surface corresponding to the ablation point was plotted. Such an example of a light intensity profile is presented in figure 4 for an image recorded at 1 μs after the laser shot. The distance \( r = 0 \) cm
represents the surface of the target. The expansion velocity of the plasma plume was calculated as the slope of the line which depicts the distance from the target surface where the maximum value of light intensity (distance \( r_m \) in the figure 4) is obtained versus the time of the recorded image. This dependence is presented in figure 5. It was found that the expansion velocity of the plasma plume is about \( 4.9 \times 10^3 \) m/s.

3.2. Probe measurements

The use of probe measurements in the diagnostic of the ablation plasma plume was previously reported [9–13]. First, experiments and testing of the probe method have been made taking into account the contamination process due to a possible thin layer deposition of the ablated metal onto the probe surface.

First, the probe was exposed for 30 minutes to the plasma plume. Then, the probe characteristic was reconstructed from the signals recorded (figure 6, dashed line). The dashed dot line in figure 6 represents the probe characteristic after a 90 minute total exposure of the probe to the plasma plume. It is clear the effect of the contamination due to a deposition of a thin layer of the target material on the probe surface.

Then, to avoid probe contamination, a heated probe was used. The probe characteristic for the heated probe is represented in figure 6 – full line. The probe was heated to a temperature at which deposition of the ablated material on the probe surface was not possible but taking care that the probe has not become a thermoemissive one. The latter fact can be proved by keeping the ionic saturation current at the same value as that of an uncontaminated cold probe.

![Figure 6. Cold probe versus heated probe characteristics.](image)

![Figure 7. Time dependence of the probe signal for various biasing voltage \((r = 3 \text{ cm and } \theta = 90^\circ)\).](image)

A large difference between the cold contaminated probe and the heated probe characteristics can be observed. It shows that measurements performed with contamination of the cold probe might lead to untrue plasma parameters, while the heated probe provides reliable data on plasma parameters.

A typical time-dependence of the probe signal for various biasing voltage \((r = 3 \text{ cm and } \theta = 90^\circ)\) is shown in figure 7. At certain values of time, the probe characteristics were reconstructed and plasma parameters were calculated. The electron temperature, \( T_e \), and the electron density, \( n_e \), were estimated using the standard procedure of processing the probe characteristic [14]. Time dependence of the electron temperature and of the electron density for various values of the \( \theta \) angle are shown in figure 8 a) and b), respectively.

Moreover, the probe characteristics show the presence of two groups of electrons within the plasma plume. One group of electrons had the temperature in the range of 0.1 eV to 0.3 eV. The other group
of electrons could not always be measured (due to the small number of points of the characteristic) and the temperature ranged from 1 eV up to about 4 eV. The density of the slow electrons ranged from $1 \times 10^{11}$ cm$^{-3}$ to $5 \times 10^{11}$ cm$^{-3}$ and decreases in time, while the second group is more than one order of magnitude lower than the density of the slow electrons.

![Figure 8. Time dependence of $T_e$ (a) and $n_e$ (b) for various $\theta$ ($r = 3$ cm and a laser intensity of $6 \times 10^8$ W/cm$^2$).](image)

Another velocity estimation of the spatial expansion of the plasma plume was obtained from the time evolution of the ion saturation current of the probe registered at a different position, $r$, along the axis of the plasma plume. It can be seen that with increasing the $r$ distance measured from the target (starting with 1.7 cm up to 4 cm) the maximum of the ionic probe signal is moving from early values of time toward later values of time (figure 9). Again a remarkable linear dependence is obtained when the distance $r$ is plotted versus time $t_m$ when the ionic probe signal has a maximum value. This dependence is shown in figure 10. The estimated value of the plasma plume expansion velocity is around $13 \times 10^3$ m/s, which is more than twice the expansion velocity estimated from the light intensities recorded with the ICCD.

![Figure 9. Ionic signals for various radial positions](image)

![Figure 10. Estimation of plasma expansion velocity from ionic part of probe signals](image)
Using the double probe instead, potential differences between the probe tips at various distances from target surface were recorded (figure 11). From the dependence of the distance at which the probe signal was recorded versus the time-value \( t_m \) that correspond to the maximum value of the potential difference, the plasma plume expansion velocity can be also estimated from the slope of the dependence. This dependence is shown in figure 12. The estimated value of the plasma expansion velocity is around \(10 \times 10^3\) m/s which is in good agreement with the one estimated from the ionic probe signal and is twice the one estimated from ICCD images.

The presence of two different values of the plasma expansion velocity can be explained by the formation of two plasma structures. This process has already been observed in studies on targets as Cu [12,13], Al [15,16] or graphite [17]. The two structures exhibit distinct dynamics and velocities, usually in the range of \(10^4\) m/s for the first structure and \(10^3\) m/s for the second one. The first structure, with a high speed, is mainly formed by plasma charged particles, while the second one is mainly formed by neutrals [16]. Consequently, probe measurements allow measuring the velocity of the first structure of the expanding plume (with velocity in the range of \(10^4\) m/s), while using the light intensity recorded by ICCD images the expansion velocity of the second structure can be estimated (with the velocity around \(10^3\) m/s).

**Figure 11.** Double probe difference potential for various radial positions.

**Figure 12.** Plasma expansion velocity calculated from double probe measurements.

**4. Conclusion**

The transient plasmas generated by high-fluence nanosecond laser ablation of stainless steel targets have been investigated by means of fast ICCD imaging and by electrical probes in transient regime. The electron temperature and density calculated from the probe characteristics recorded by electric probes in transient regime were in the range of 0.1 eV to 0.3 eV and \(1 – 5 \times 10^{11}\) cm\(^{-3}\) respectively.

The expansion velocity estimated from the total light intensity recorded by ICCD imaging is correlated with a second slower structure, mainly formed by neutral particles, which appears in the dynamics of plasma plume and has the value around \(5 \times 10^3\) m/s.

From the probe measurements a plasma expansion velocity was estimated that is correlated with a first faster structure, mainly formed by charged particles, and has a value around \(1 \times 10^4\) m/s.

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**References**

[1] Peterlongo A, Miotello A and Kelly R 1994 *Phys. Rev. E* 50 4746
[2] Jordan R and Lunney J G 1998 *Appl. Surf. Sci.* 127–129 968
[3] Chang J J and Warner B E 1996 Appl. Phys. Lett. 69 473
[4] Amoruso S, Sambri A, Vitiello M and Wang X 2006 Appl. Surf. Sci. 252 4712
[5] Amoruso S, Sambri A and Wang X 2006 J. Appl. Phys. 100 013302
[6] Hermann J, Boulmer-Leborgne C and Hong D J. 1998 Appl. Phys. 83 691
[7] Amoruso S, Bruzzese R, Velotta R, Spinelli N, Vitiello M and Wang X 2005 Appl. Surf. Sci. 248 45
[8] Amoruso S, Armenante M, Berardi V, Bruzzese R, Pica G and Velotta R 1996 Appl. Surf. Sci. 106 507
[9] Lunney J G, Doggett B and Kaufman Y 2007 Journal of Physics: Conference Series 59 470
[10] Hansen T N, Schou J and Lunney J G 1999 Appl. Phys. A (Suppl.) 69 S601-4
[11] Hendron J M, Mahony C M O, Morrow T and Graham W G 1997 J. Appl. Phys. 81 2131
[12] Gurlui S, Sanduloviciu M, 2006 AIP Conf. Proc. 812 279
[13] Gurlui S, Sanduloviciu M, Strat M, Strat G, Mihean C, Ziskind M and Focsa C 2006 J. Optoelectron. Adv. Mat. 8 148
[14] Swift J D, Shwar M J R 1970 Electrical probe for plasma diagnostics (ILLFE-Book)
[15] Gurlui S, Agop M, Nica P, Ziskind M and Focsa C 2008 Phys. Rev. E in press
[16] Ursu C, Gurlui S, Focsa C, Popa G 2008 Nucl. Instrum. Meth. B in press
[17] Bulgakova N M, Bulgakov A V and Bobrenok O F 2000 Phys. Rev. E 62 5624