Study of nanosecond resolved spectra and comparison between single and double pulse in linear configuration of aluminum plasmas


Centro de Lasers e Aplicações, Instituto de Pesquisas Energéticas e Nucleares, Caixa Postal 11049, 05422-970 São Paulo-SP, Brazil
mamaral@ipen.br

Abstract

Laser induced plasma spectroscopy is a very interesting technique that can be employed to characterize metallic materials by generating plasma due to the laser interaction to the samples. Some improvements of the technique are needed in order to obtain full correspondence between the sample characteristics and plasma generated during the ablation. Comparison between single and double-pulse (collinear configuration) was made in this work for laser induced plasma spectroscopy of high purity aluminum samples using a laser system with wavelength of 800 nm, 480 µJ of energy pulse (measured near the laser output), 1 kHz of repetition rate and 50fs FWMH. The emission from the plasma was collected by a lens and dispersed on a monochromator whose exit slit was connected to a photomultiplier. The spectra were recorded by the lock-in technique (time integrated spectra) as well as the box-car technique (time resolved spectra). Temperature and density were acquired using Boltzmann Plot and Stark broadening treatment, respectively. The collinear configuration with first pulse (374 µJ) more energetic than second (106 µJ) presents a temperature and density (49000 ± 36000 K and 1.36 ± 0.09 10^{17} cm^{-3} respectively) greater than single pulse (370 µJ measured after passing the optical path through mirrors and focusing lens) and double-pulse with first pulse (42 µJ) less energetic than second (308 µJ) that has 11400 ± 900 K and 8.7 ± 0.5 10^{16} cm^{-3} and 8100 ± 200 K and 6.6 ± 0.3 10^{16} cm^{-3}. This behavior was attributed to the pre-ablative effect when the first pulse is more energetic than second. The study of emission lifetime of Al I 394.4 nm line and resolved spectra of plasma temporal evolution with delay times from 60 ns to 200 ns were used to chose the best delay to acquire the plasma information, the best delay acquired was 120 ns delay time due the maximum emission and spectra resolution.

Introduction

Many studies have been made using LIBS (Laser Induced Breakdown Spectroscopy) for analysis of aluminum alloys. Some papers investigate the ionic and atomic density of plasma [1] as well as the influence of laser parameters on the plasma characteristics [2; 3]. A comparison between LIBS and other quantitative analysis methods are also an interesting subject for the consolidation of LIBS as an analytical method [4].

Some groups have reported comparison between single and double-pulse LIBS configuration [5-9] and they argue that enhancement of signal in double pulse configuration was observed. The aim of the present work was compare single and double-pulse in collinear configuration and study the plasma temporal evolution using a photomultiplier to record nanosecond resolved spectra from the plasma.

In order to determine the plasma parameters we use Boltzmann plot (equation 1) [10] for evaluating temperature and Stark broadening (equation 2) for electron density [6].
\[
\ln \left( \frac{N \lambda}{gA} \right) = C - \frac{E}{kT}
\]

Equation 1

where \(N\) is the observed transition intensity, \(\lambda\) is the transition wavelength, \(g\) is the statistical weight of upper level, \(A\) is the transition probability, \(C\) is a constant, \(E\) is the energy of upper level, \(k\) is de Boltzmann constant and \(T\) is the temperature.

\[W_{tot} = w(T) \left( \frac{n_e}{10^{16}} \right)\]

Equation 2

where \(W_{tot}\) is the half width of experimental line, \(w(T)\) is the tabled Stark parameter [11] as a temperature function and finally \(n_e\) is the electron density.

Experimental setup

A 50 fs -300 mW pulse from a KLM Ti:Sapphire laser (Mira Coherent) was used as seeder to an amplifier (Odin – Quantronix) pumped by a Q-switched 2\(\omega\) Nd:YLF laser (Falcon 527 -Quantronix). The output pulse from the amplifier with 800nm, 480\(\mu\)J, 1kHz and 50fs was used to generate the plasma in samples. After breakdown, the plasma emission was collected with a Galilean telescope and injected into a 0.5m monochromator connected to a photomultiplier (Hamamatsu); the schematic experimental setup is shown in Figure 1. The pulse energy was measured after passing the optical path through mirrors and focusing lens. For double pulse the measured energy was distributed to each pulse using oscilloscope signal for calibration.

In the first part of this work, the plasma was generated focusing the laser over the sample surface. The sample was mounted in a rotator to permit that the crater generated by the laser was not so deep, avoiding decrease of the plasma signal, and to guarantee that we collect light from the same position in the plasma. The photomultiplier signal was connected to a box-car with 5ns of integration time gate, the sample point (delay time from pulse to acquisition) was varied from 60ns to 200ns (in intervals of 20ns) to permit the temporal resolved spectra acquisition.

The second part was the comparison between single and double pulse in the collinear configuration. In the collinear configuration the incidence is perpendicular to the sample in the two laser pulses (Figure 2). Obtaining the two pulses was made by a misalignment of Pockels cell, with this, the pulse picker permits more than one pulse passing through, and the interpulse separation was of 13ns. Two different pulse configurations were possible, first pulse with 374\(\mu\)J and second with 106\(\mu\)J or first pulse with 42\(\mu\)J and second with 308\(\mu\)J. The single pulse energy was 370\(\mu\)J. The aluminum sample was again placed on a rotator for the same reason than before. The photomultiplier signal was connected to a Lock-in with 540\(\mu\)s integration time gate and the integrated spectra acquired.

To study the plasma emission, the data base of NIST [12] (National Institute of Standards and Technology) was used. These data give us the parameters \((E, l, g, A\) from equation 1\) needed to calculate the plasma temperature.
Results and discussion

In the first part of the experiment, temporal resolved spectra was acquired with a delay time from 60 ns to 200 ns, these spectra in the region between 390 nm and 400 nm is shown in Figure 3. These spectra show that the line emission becomes narrower when the time increases although the signal becomes weaker. The time evolution of Al I 394.4 nm line, shown in Figure 4, was also studied as an important tool in the results discussion. With the information of Al I 394.4 nm line lifetime we could get 120ns delay time as the best delay to acquire the plasma information, due to the maximum emission and spectra resolution at this time.

Figure 3: Temporal evolution the region between 390 nm and 400 nm

In the second part, the acquired spectra allowed to calculate de temperature and density for the three different configurations: single pulse, double pulse with 374µJ in the first pulse and 106µJ in the second one and double pulse with 42µJ in the first pulse and 307µJ second one. For calculating the density from Stark broadening was chosen the Al I 394.4 nm line that appears in all spectra with a good resolution, allowing a Lorentzian fit to the data. The Figure 5 and Figure 6 show the value of temperature and density calculated for these three configurations.

Figure 5: Temperature calculate for this three configuration

Figure 6: Density calculate for this three configuration

The temperature and density in collinear configuration with first pulse more energetic than second (49000±36000K and 1.36±0.09 \(10^{16}\) cm\(^{-3}\) respectively) is greater than the other configurations, single pulse (11400±900K and 8.7±0.5 \(10^{15}\) cm\(^{-3}\)) and collinear configuration with first pulse less energetic than second (8100±200K and 6.6±0.3 \(10^{15}\) cm\(^{-3}\)). This behavior is attributed to the first pulse having sufficient energy to ionize the sample and the air around the sample causing a high temperature and density; the second pulse just reheats plasma. Besides the ablative effect, the first pulse has a pre-ablative effect due to creation of air sparks.
In the other collinear configuration, when first pulse was less energetic than second, the first pulse was not sufficiently energetic to ionize the air, being sufficient only to ionize the aluminum sample. Because of its low energy ($42 \mu J$) the temperature and density is comparable to single pulse plasma temperature and density. When the second pulse reaches the plasma it causes a reheating in the plasma and is absorbed by it causing new ionizations. This reheat causes an expansion in the bulk plasma and consequently the decrease of temperature and density.

**Conclusions**

The comparison between single and double-pulse in collinear configuration was made and when the first pulse is more energetic than second the temperature and density is greater than single pulse and double-pulse with first pulse less energetic than second. It happens due to the first pulse having a pre-ablative effect.

The plasma temporal evolution study could give 120ns delay time as the best delay to acquire the plasma information due to the maximum emission and spectra resolution.

**Acknowledgments**

The authors acknowledge FAPESP (00/15135-9, 04/15965-2) and FINEP (04-1604) for financial support. One of authors Marcello Magri Amaral thanks CNPq for his fellowship under process number 135366/2006-2.

**Reference**


