Optical characterization of plasmas produced by concurrent application of laser and high voltage pulses

A Alonso¹, H Sobral², A Robledo-Martinez³ M Peña-Gomar¹

¹Universidad Michoacana de San Nicolás de Hidalgo. Avenida Francisco J. Múgica s/n, Ciudad Universitaria, C. P. 58030, Morelia Mich., Mexico
²Instituto de Ciencias Aplicadas y Tecnología, Universidad Nacional Autónoma de México (ICAT-UNAM), Apartado Postal 70-186, Ciudad de México, 04510, México.
³Departamento de Ciencias Básicas, Universidad Autónoma Metropolitana, Av. San Pablo 180, Azcapotzalco, Ciudad de México, 02200, Mexico.

e-mail: martin.sobral@ccadet.unam.mx

Abstract. Laser Induced Breakdown Spectroscopy (LIBS) is a simple, powerful analytic technique that is limited by a relatively low sensibility to detect traces. To improve LIBS sensitivity an option often used consists in applying a second source of excitation such as an electric discharge. This work’s goal is to investigate the interaction of a laser-produced plasma on an aluminum target with a self-triggered electric discharge. The plasma dynamics has been investigated using shadowgraphy, fast photography and the application of narrow band-pass filters to follow the evolution of the ionic and neutral species. Results show that within the initial microseconds, the spatial extent of the laser-produced plasma increases due to the presence of a low density region induced by the spark channel. Furthermore, it was found that the electric arc mainly re-excite the ionic species while the neutral ones remains unaffected.

1. Introduction.
Laser Induced Breakdown Spectroscopy (LIBS) is a powerful analytic technique used in the metallurgical industry, archeology, nuclear industry, forensic studies, ambient and cultural applications and many others. Its main advantages stem from its portability, in situ capability, and high spatial and temporal resolution [1,2]. However, the main drawback of the technique is its relatively low trace detection sensitivity which is of the order of one part per million, compared to other established techniques such as Inductively Coupled Plasma. Thus, to enhance LIBS sensitivity a second excitation source such as a second laser pulse, microwave, hollow cathode lamp and electric discharge have been used [3-6]. The later configuration has the advantage of a relative low cost and easy instrumentation. Furthermore, in the work of Belkov et al. [7] it has been reported lower detection limits of traces using Spark Discharge Laser Ablation (SD-LA) than using the double pulse scheme. On the other hand, Chen et al. [8] reported a negligible enhancement of the LIBS sensibility usig SD-LA, probably due to the large delay between the laser pulse and the discharge employed, which was of the order of tens of microseconds. In a previous work our group obtained an enhancement of up to one order of magnitude using an unpolar electric discharge as the second
excitation source [9]. Recently we further enhanced the previous obtained values controlling the delay at which the electric pulse is applied [10].

The aim of this work is to investigate the physical mechanism which results in an enhancement of the limit of detection of traces of the LIBS technique using a high voltage pulse. Therefore, the dynamic of plasma interactions has been investigated with fast photography of the whole light and of selected bands using interferential filters to monitor the spatial distribution of the ablated species [11]. In order to improve the understanding of the plasma dynamics shadowgraphy was employed to visualize the interplay between the plasma and the arc [12].

2. Methodology and experimental setup.

Figure 1 shows the schematics of the experimental setup employed to investigate the evolution of the plasma interaction using shadowgraphy and fast photography.

The ablation laser was a Nd:YAG (Surelite III, from Continuum) delivering 5-ns width Gaussian pulses at 1064 nm and was operated at a 1 Hz repetition rate. The output energy was 20 mJ per pulse and the beam was focused by a 10 cm plano-convex lens onto the target producing a spot diameter of ~120 μm hence, the ablation laser fluence was 180 J cm⁻². The ablated sample was a polished aluminum slab from a commercial aluminum 6463 alloy containing 98% Al, 0.8% Mg, 0.5% Si, 0.2% Cu, 0.2% Fe, plus traces of other elements. The target was mounted onto a translation stage to adjust the focusing distance and to shift the target for each run of data collection. Each acquisition consisted of 20 shots on the same spot before moving to a new position. All experiments were performed in ambient air.

The re-excitation electric pulse is similar to that reported in a previous work [9]. Briefly, a high voltage power supply (model 205 B, from Bertan) giving an output of 14 kV charges, through a 10 MΩ resistor, a 50-m length of 50-Ω RG58 cable which acts as a capacitor. The cable output is connected to a stainless-steel cylindrical rod 10 mm in diameter with hemispherical tip that acts as the anode of the spark-gap. The aluminum target acts as the cathode and it was grounded through a 50 Ω resistance. The distance between the electrode and the target was about 6 mm with the axis of the rod at an angle of ~30°. The current was monitored by means of a Rogowski coil on the low-voltage side of the circuit, and was connected to a digital oscilloscope (DPO 4104B from Tektronix). The laser and diagnostic system were synchronized through an 8-channel pulse/delay generator DG (575-8C, from Berkeley Nucleonics).

Shadowgraphy measurements were performed expanding and collimating the output of a continuous diode laser emitting at 532 nm with 50 mW power. Its beam illuminates the plasma plume in a direction that is perpendicular to the path of the pulsed laser as shown in Figure 1. The plume produces a shadow which is proportional to the second derivative of the plasma’s refractive index. The image is collected by the ICCD camera at different delays and with a variable gate width from 20 ns to
1 µs, depending on the acquisition delay. The camera was synchronized with the laser emission by means of a pulse/delay generator (Berkeley Nucleonics, 575-8C).

For fast photography technique the plasma plume was directly imaged on ICCD camera at different delays after laser onset. Besides, two band pass filters (10 nm full width at half maximum) centered at 307 and 500 nm were employed to investigate the spatial distribution of the neutral aluminium and ionic nitrogen transition, respectively. The first filter monitors several transition in the 308-309 nm region corresponding to Al I and the later the N II line at 500.5 nm.

3. Results and discussion.

3.1. Electric circuit characteristics

Figure 2 shows the signals of the laser light, the electrical current and the ICCD’s gate recorded by the oscilloscope in a representative test. The laser light was always taken as the time reference: \( t=0 \). Figure 2 shows the current profile obtained after the integration of the Rogowski coil’s signal. The delay between the beginning of the laser pulse and the electric spark was estimated to be about 200 ns as can be seen from the figure.

![Figure 2](image-url)

**Figure 2.** Temporal sequence of the laser light, current and the camera gate. In this instance the fast camera took the photograph at \( t=700 \) ns with an exposure of 10 ns

The technique to obtain a unipolar, square current pulse is based in the use of a 50-m length of RG58 cable which works as a transmission line. When the laser beam hits the target, it generates an ablation plasma that leads to the electrical breakdown of the gap. As the speed of propagation on this cable is \( 2 \times 10^8 \) m/s the single transit time is \( \tau = 250 \) ns; therefore, a current pulse of total length \( 2\tau = 500 \) ns is obtained at the load, as can be seen in the middle frame of Fig. 2. The RG58 has a characteristic impedance of 50 Ω and consequently the resistor connected to the target (the cathode) must have the same value, in order to avoid reflections.
3.2. Shadowgraphy.

![Shadowgraphy images](image)

**Figure 3.** Dynamic evolution of the plasmas using shadowgraphy images. Top row: laser ablation; lower row: Spark Discharge Laser Ablation. Indicated above each frame is the ICCD gate delay after laser onset.

Figure 3 shows a sequence of images obtained with the shadowgraphy technique comparing the evolution of a laser ablation plasma (LA) and spark discharge triggered by laser ablation (SD-LA).

As it can be expected, at early times, shadowgrams corresponding to both experimental schemes show basically the same plume shape. At around 200 ns after laser ignition, the discharge channel already bridges the gap between the plume and the anode. The ablation plasma grows approximately in a radial direction. For SD-LA the arc axis crosses the center of the plasma. The arc has a cylindrical symmetry and it expands away from its axis as time elapses. After the extinction of the electric discharge, at around 800 ns, both plasmas continue expanding and cooling as can be seen in the right-hand side panels of Fig 3. The expansion of the LA plasma is confined by the atmospheric pressure ending with a flattened shape as can be observed at 90 µs. On the other hand, for the SD-LA the ablation plasma expands in a turbulent manner to a larger size compared to the single pulse configuration, probably due to the lower density region produced by the electric discharge.

3.3. Fast photography.

Fast photography results complemented the shadowgraphy analysis giving an additional tool to explain the observed emission enhancement [9,10]. Figure 4 shows a sequence of images obtained at a fixed charging voltage of 14 kV and variable camera delay. The gate width of the ICCD employed was 10 ns for all images.
Figure 4. Sequence obtained using fast photography. Top row: photos taken without filter; middle row: using a band pass filter centered at 307 nm (Al I); bottom row: using a filter for N II at 500 nm.

The figure has three rows: the upper one correspond to images of the unfiltered light emitted by the plasma. The middle and lower rows show the images obtained using two interferential filters that preferentially transmit the emission of the neutral aluminum and ionic nitrogen. At the earliest reported time, the spatial distribution of ionic and neutral species is quite similar. Here it is expected that the ionic species will concentrate near the plasma core and that the aluminium neutral emission be located all around the plasma borders. After the electric discharge ignition, at 210 ns, the fully completed arc can be seen in the three rows. The observed emission in the spatial region of the arc channel with the neutral aluminium filter is due to the Bremsstrahlung radiation passing the 10 nm bandwidth filter. At later times in the top row, around the microsecond scale, the light from the discharge channel starts to vanish and by 10 µs it is only possible to see the ablation plume.

We did not observe a major change effect of the arc on shape, intensity or temporal duration of the neutral aluminium emission. For that reason, is possible to conclude the arc has a negligible influence on the neutral species as was previously observed [9]. The photos taken using the N II filter shows that the whole discharge has atmospheric ionic species that transfer their energy to the plume and excite ionic species in the ablation plasma, a sign that reheating is taking place. Finally, at longer times, the neutral species fill all the unfiltered plume volume while the ionic species are limited to a zone near the target as can be seen in the lower row at 11 µs.

4. Conclusions
In this work we investigated the dynamics of a laser produced plasma (LA) onto an aluminium target and the interaction of this laser ablation plume with a fast, square high-voltage discharge (SD-LA). The evolution of produced plasmas was analyzed using shadowgraphy, fast photography of the whole light and narrow-bandwidth filtered light. Shadowgraphy results of SD-LA scheme shows, in the microsecond time scale, that the laser plume becomes turbulent faster compared to the single pulse experiment; this is due to the interaction of LA with the electric discharge. Furthermore, LA expands
due to the low air density produced by the spark discharge channel. Fast photography using filters, shows that the electric discharge has no appreciable effect on neutral aluminium species and that most of the electric energy is transferred to ions. Finally, for longer times, ionic emission is only found near the target ablation spot, while neutral species occupy a larger physical region.

Acknowledgments

This work was supported by the National Autonomous University of Mexico (DGAPA-UNAM: IG100918) and the Universidad Michoacana de San Nicolás de Hidalgo (CIC-UMSNH).

References

[1] Noll R 2012 *Laser-Induced Breakdown Spectroscopy, Fundamentals and Applications* (Berlin: Springer-Verlag).
[2] Musazzi S and Perini U 2014 *Laser-induced Breakdown Spectroscopy Theory and Applications* (Berlin Heidelberg: Springer-Verlag).
[3] Aziz A, Brekaert J A, Laqua K, Leis F 1984 A study of direct analysis of solid samples using spark ablation combined with excitation in an inductively coupled plasma *Spectrochim. Acta B* 39 1091–1103.
[4] Dittrich K and Wennrich RLaser vaporization in atomic spectroscopy 1984 *Prog. Analzyt. Atom. Spectrosc.* 7 139–198.
[5] Lewis C, Doorn S K, Wayne D M, King F L, Majidi V 2000 Characterization of a pulsed glow discharge laser ablation system using optical emission *App. Spectroscopy* 54 1236–1244.
[6] Liu Y, Baudelet M, Richardson M 2010 Elemental analysis by microwave-assisted laser induced breakdown spectroscopy: evaluation on ceramics *J. Anal. At. Spectr.* 25 1316–1323.
[7] Belkov M V, Burakov V S, De Giacomo A, Kiris V V, Raikov S N, Tarasenko N V 2009 Comparison of two laser-induced breakdown spectroscopy techniques for total carbon measurement in soils *Spectrochim. Acta B* 64 899–904.
[8] Chen Y, Zhang Q, Li G, Li R, J. Zhou 2010 Laser ignition assisted spark-induced breakdown spectroscopy for the ultra-sensitive detection of trace mercury ions in aqueous solutions *J. Anal. At. Spectrom.* 25 1969–1973.
[9] Sobral H and Robledo-Martinez A 2016 Signal enhancement in laser-induced breakdown spectroscopy using fast square-pulse discharges *Spectrochim. B* 124 67–73.
[10] Robledo-Martinez A, Sobral H, Garcia-Villarreal A 2018 Effect of applied voltage and interpulse delay in spark-assisted LIBS *Spectrochim. B* 144 7–14.
[11] Sanginés R and Sobral H 2013 Time resolved study of the emission enhancement mechanisms in orthogonal double-pulse laser-induced breakdown spectroscopy *Spectrochim. B* 88 150–155.
[12] Sanginés R and Sobral H 2011 Two-color interferometry and shadowgraphy characterization of an orthogonal double-pulse laser ablation J.App. Phys. 110. 033301-1-6.