Acoustic and optical emission during laser-induced plasma formation

S. Conesa, S. Palanco, J.J. Laserna*

Department of Analytical Chemistry, Faculty of Sciences, University of Málaga, Campus de Teatinos, E29071 Málaga, Spain

Received 20 February 2004; accepted 9 June 2004
Available online 1 August 2004

Abstract
Laser ablation is widely used in laser processing and analysis of materials. The laser beam evaporates and ionizes material, creating a plasma plume that expands to variable extent and morphology depending on both the sample and its surrounding gas properties. At ambient pressure a shock wave front appears, traveling at variable velocities which are related to the own plasma formation mechanism. Plasma images as well as the acoustic spectral content of the emission within the aural perception range are related to the plasma formation and evolution dynamics. These results are discussed on the basis of different plasma expansion mechanisms.

© 2004 Elsevier B.V. All rights reserved.

Keywords: Laser-induced plasma; Plasma expansion; Plasma acoustic signal; Acoustic spectra

1. Introduction
Short-pulse laser ablation is a proven tool in the field of laser processing and analysis of materials due among others reasons to the weaken thermal side effects compared to long-pulse laser operation. The laser beam evaporates and ionizes material creating a plasma plume that expands differently depending on the laser pulse stability, surrounding atmosphere conditions, and target characteristics. These factors may induce certain fluctuations in the amount of ablated material. Such behavior is also evidenced when using laser ablation as a solid sampling method for analytical techniques such as inductively coupled plasma atomic emission spectrometry (ICP-AES) [1,2]. To further develop equipment based on laser ablation, the implementation of a system to monitor the ablation rate in real time would be of paramount importance. The energy absorbed in the sample from the laser beam is then released through a thermal mechanism. The gas surrounding the vapor plume, initially transparent, is heated and starts absorbing the laser radiation with a self-perpetuating mechanism that results in plasma propagation to the surrounding atmosphere. The energy is mostly transferred by conduction during the first stages, while radiation is the dominant mechanism in later stages. The subsequent evolution of the plasma may follow three major paths depending on the increasing laser irradiance: (i) laser-supported combustion (LSC) wave, for irradiances lower than $10^7$ W cm$^{-2}$, (ii) laser-supported detonation (LSD) wave, for irradiances up to $10^9$ W cm$^{-2}$, or (iii) laser-supported radiation (LSR) wave for irradiances beyond that level. Differences in plasma expansion arise from the different mechanisms dominating the propagation of the absorbing front into the cool, transparent atmosphere. Although these phenomena can be tracked by studying velocity, pressure, and the effect of radial expansion on the subsequent plasma evolution, it should be emphasized that no clear boundaries exist in between them [3,4].

Regardless of the dominant plasma wave mechanism, the high energy deposited on the target is released in the form of thermal (thermoelastic expansion), optical (plasma spark), and acoustic phenomena. The latter consist of a broadband acoustic wave, including an audible, characteristic popping sound owing to both the supersonic expansion of the generated shock waves, and the expansion of the plasma wave associated to it. Thus, these shock waves do carry information regarding the laser-induced ablation. If this information is proved to be both accessible and meaningful, an acoustic signal monitoring system should be considered among the candidates to study the laser-induced ablation processes. Moreover, such an acoustic based system can be designed to be rugged, inexpensive, and easy to use or even automated. Several papers have dealt with the acoustic signal generated by laser-induced plasmas; among them,
the work by Diaci and Mozina [5] focused on the energy content of the acoustic wave. In a continuation of this work, qualitative results were presented that demonstrated monitoring during excimer-laser ablation of ceramics by photono-acoustics means [6]. A few years back, the relationship between the shock wave energy and the ablation rate for the different shock wave propagation mechanism was demonstrated [7]. Recently, a practical model of acoustic wave generation was presented and found applicable to weakly ablative regimes under vacuum [8]. The spectrum of acoustic emission produced during laser welding has been also studied [9]. In a recent work by this group, the acoustic emission of laser-induced plasmas has been spectrally studied and discussed on the basis of the physical properties of the ablated material, the expansion mechanisms of the plasma and an empirical parameter representative of the transported energy [10].

In the present study, the information carried out by the acoustic wave has been further investigated, focusing in the spectral content within the aural acoustic range. The irradiances employed covered the range from $10^9$ to $10^{10}$ W cm$^{-2}$. Even though we were theoretically only covering the LSR regime, the results seem to indicate that the plasma underwent a transition from LSD to LSR. Images of the expanding plasma were also acquired in order to accomplish the study of the plume expansion from another perspective. Although there is clear limitation on the sampling rate if we try to measure down the microsecond range, the shock wave still transports enough information related to its origin—the plasma formation—once it becomes sonic. In fact, an experienced observer could distinguish the plasma formation regime just from the aural perception. Our investigation does not imply to perform measurements right during the plasma formation, but to infer phenomena occurring during plasma formation and its expansion from measurements of the air shock wave in the sonic domain.

2. Experimental

Fig. 1 shows a schematic diagram of the experimental layout employed during these experiments. A Q-switched Nd:YAG (Quantel Brilliant) laser providing pulses of 350 mJ in energy and under 5 ns in length was operated at 5 Hz. Its fundamental wavelength output was focused at normal incidence on reference aluminum samples through a plano-convex lens ($f = 100$ mm). A filter was placed within the $5\times$-expanded beam as a means of controlling the
irradiance on the sample without altering the beam profile. The spot area at the sample was approximately 0.50 mm$^2$. The acoustic emission from laser-produced plasmas was detected parallel to the sample surface with a dynamic microphone (FoneStar FDM-9058) placed at an approximate distance of 7 cm from the plasma. The microphone, the sample holder, and the sample itself were decoupled by using neoprene dampers to prevent detection of undesired vibrations and feedback loops. The output from the microphone was pre-amplified and then directed to a computer. Signals 11.6 ms in length were digitized using a computer sound card at the standard sampling rate of 44.1 kHz. The sample was observed simultaneously with an intensified CCD camera. The synchrony output of the laser Q-switch was used to trigger the acquisition. Acoustic emission corresponding to 100 laser shots was acquired and averaged for each experiment, while 25 plasma images at increasing delays were acquired at every irradiance level.

The pulse duration used in this experiment (< 5 ns) is short relative to the diffusion time across the absorption depth (around 10 ns). Thus, the process is verified in the regime of thermal confinement, but not thermoelastic confinement. It has been demonstrated that in this regime the amount of energy deposited by the laser pulse—rather than the pulse width—determines the mass removal process [11,12]. A non-thermal mechanism occurs at the irradiances contemplated in the present study, with mass removal assumed to be in the form of the mixture of vapor and micrometer sized particles.

All experimental parameters were optimized in order to get a reproducible acoustic spectrum in which significant differences could be detected at varying irradiances. The laboratory dimensions and sample location determine the frequencies (or modes) at which standing waves appear. These eigenfrequencies form a landscape of modes with distinct hills and deep valleys. On top of this, the acquisition times for the acoustic signal imposed a frequency cut-off at 86 Hz. The three-dimensional standing waves were also calculated applying the corresponding equation:

$$f(n_x, n_y, n_z) = \frac{c}{2}\left[\left(\frac{n_x}{L_x}\right)^2 + \left(\frac{n_y}{L_y}\right)^2 + \left(\frac{n_z}{L_z}\right)^2\right]^{1/2}$$

where $f$ is the sound frequency in Hz, $n_x$, $n_y$, and $n_z$ are positive integers corresponding to the different harmonics, $c$ is the speed of sound, and $L$ is the laboratory dimension considered. As the acoustic signal climbs the harmonic ladder, its intensity decreases. All possible combinations of fundamentals and second harmonics were studied for our laboratory dimensions, and all the obtained eigenfrequencies were within the range from 129 to 259 Hz. Furthermore, it is well known that standing waves cause little problems above approximately 300 Hz. Hence, this source of acoustic feedback was negligible during our study. Taking into account that great care was taken to damp the acoustic signal from the sample, it is safe to assume that only the external shock wave was responsible for the detected acoustic signal. In view of this, and after studying the spectral response of the microphone, the spectral window covering from 3 to 16 kHz was chosen for the present investigation. The mechanical roll-off in the response of the transducer for frequencies above 16
kHz acted as a low-pass filter and ensured that there was no possibility for the ultrasonic frequencies to appear in our spectra by means of aliasing artifacts below the Nyquist frequency [13].

3. Results and discussion

The laser-induced plasma (LIP) acoustic spectra from an aluminum sheet sample obtained at two different irradiances are shown in Fig. 2. Oppositely to the broadband-like spectra observed in a previous work [10], spectra in Fig. 2 are nearly featureless with the exception of the peaks at 7.62 and 11.13 kHz. These peaks match two spectral regions where the microphone exhibits a higher response and were found to be invariant to room dimensions and to sample shape, mass, and composition. In fact, the same experiments were run with a copper ring and a silicon wafer and the peaks remained at the same positions. The only difference among the different samples was the laser irradiance for which the peak at 7.62 kHz became the dominant spectral feature.
In the case of the aluminum sample in Fig. 2, the spectra corresponding to the lower irradiance \((2.6 \times 10^9 \text{ W cm}^{-2})\) exhibits a single peak at 11.13 kHz, whereas the acoustic signal at 7.62 kHz is the highest at an irradiance level of \(12.2 \times 10^9 \text{ W cm}^{-2}\). Taking into account the inherent non-linear response of microphones, it is more important to focus on the fact that most of the acoustic energy emitted within the audible range has been displaced under increasing irradiances toward lower frequencies, rather than focusing on the precise frequencies measured. These results are in good agreement with those obtained at frequencies above the aural perception [5]. A peak in the region 9–10 kHz for irradiances above \(10^{10} \text{ W cm}^{-2}\), that was absent at lower irradiance levels has been also reported [8].

In Fig. 3, the net intensities calculated by subtraction of the background from the absolute values of the 7.62 and 11.13 kHz peaks are plotted versus irradiance. Although the net signal for the 11.13 kHz peak remains approximately constant up to \(8.4 \times 10^9 \text{ W cm}^{-2}\), no clear behavior can be observed beyond that point. On the other hand, the intensity of the lower frequency peak remains constant up to \(4.7 \times 10^9 \text{ W cm}^{-2}\), and from this irradiance level upwards, a linear trend is clearly noticeable. This separate response could be related to a different physical mechanism taking place at higher irradiance levels. To pursue the matter further, a plot of the broadband acoustic energy content of the signal versus irradiance is plotted in Fig. 4. Again, two zones with a positive linear trend with markedly different slopes are observed, the steeper one corresponding to the higher irradiance regime. Such slope changes have been previously described in the literature for ablation rates [14–16] with piezoelectric transducers being used by Srinivasan and Braren [17]. These changes in the ablation rates lead to different plasma expansion dynamics. It is worth noting that the turning point for the appearance of the 7.62 kHz peak (and consequently for the steeper linear growth of acoustic energy with irradiance) lies close to \(5 \times 10^9 \text{ W cm}^{-2}\) while the theoretical models of plasma supported waves propose the transition from the LSD regime to the LSR regime at irradiances above \(\sim 10^9 \text{ W cm}^{-2}\). The change in slope to higher acoustic energy levels measured is in good agreement with the larger amount of ablated mass and higher propagation velocity involved in LSR waves.

As a result of the former, larger plasma volumes should be expected at high irradiance levels. In order to gain complementary information regarding the plume expansion dynamics two-dimensional side-view images of the plasmas were obtained. Fig. 5 shows the results at irradiance levels of \(2.6 \times 10^9\) and \(12.2 \times 10^9 \text{ W cm}^{-2}\). The acquisition was done using the delays (referred to the laser Q-switch synchrony pulse) shown in the figure. The gray intensity scale was chosen individually for each delay to ensure optimum contrast, but it was kept constant for any given couple of irradiances plotted for comparison purposes. The images corresponding to the lower irradiance show a plasma shape resembling a hyperboloid of revolution—bow shock—which is characteristic of waves propagating at supersonic velocities. This kind of behavior corresponds to a steadily growing spherical plume drifting along the target normal axis. The material ablated by the laser occupies a conical annular region and the opening angle is determined by the ratio of expansion to translation velocities [18]. The expansion of these plasmas persists up to 180 ns after the trigger event, and then starts to decay. Increasing the irradiance by about one order of magnitude in our experiments resulted in two effects: firstly, the plasmas obtained were invariably larger in size, and their growth was longer, lasting up to 340 ns after the Q-switch trigger signal. This behavior is in good agreement with a transition from an LSD regime to an LSR regime. Sec-

![Fig. 5. Temporal evolution of an aluminum laser-induced plasma at 2.6 GW cm\(^{-2}\) (left) and 12.2 GW cm\(^{-2}\) (right).](image-url)
ondly, the increase in irradiance dramatically modifies the shape of the plasma, rather resembling that of a light bulb, with a secondary plasma clearly detached from the sample surface at high irradiances. Given that the natural frequency of resonance of any source is directly dependant on its flexural rigidity and inversely dependant on its density, mass and dimensions [19], the shift to lower frequencies observed in the acoustic spectrum with the increasing laser irradiance is in good agreement with the increase in size of the plasma. In fact, plasmas under the LSR regime are not only larger than under the LSD regime but denser, what is in good agreement with the acoustic frequency shift.

Additionally, Fig. 5 reveals an evolution in the plasma shape. Plasma emission begins on the target surface soon after the laser photons reach the surface, and as time evolves, it separates into two components. The first component keeps close to the target surface, and the second component expands very rapidly with time in a highly forward-directed pattern [20]. The first component results from the phase explosion-induced by the laser–sample interaction, whereas the latter appears as a result of the strong laser–plasma interaction, where the existing species present —mostly ions—are fuelled by inverse bremsstrahlung. When the LSR regime is verified, the interaction between laser and plasma starts ahead of the shock wave front. In this scenario, the plasma core moves away from the target under higher irradiance levels. This transition would explain both the appearance of a larger plasma, and the different, more spherical shape, as the ratio of translational to expansion velocities has been altered by means of the different energy and mass absorption rates. In Fig. 6, the dimensional and morphological evolution of the plasma under increasing irradiances at a fixed delay of 260 ns is plotted, so that the aforementioned physical growth and transfiguration can be appreciated.

4. Conclusions

Acoustic spectra corresponding to laser-induced plasmas from aluminum, copper and silicon samples obtained at irradiances ranging from $1.1 \times 10^9$ to $12.2 \times 10^9$ W cm$^{-2}$ have been studied. The appearance at higher irradiance levels of an spectral feature absent at low irradiance levels is reported and a tentative explanation for the underlying mechanism has been proposed based on theoretical models. The acquisition of plasma images was used as a means of validating the feasibility of the proposed mechanism. Acoustic spectra of laser-induced plasmas have shown a high sensitivity for detection of the actual plasma expansion dynamics that follows the arrival of the laser pulse.

Acknowledgments

The present work has been funded by the Spanish Ministerio de Ciencia y Tecnología (project BQU2001-1854).

References


