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Convolved effect of laser fluence and pulse duration on the property of a nanosecond laser-induced plasma into an argon ambient gas at the atmospheric pressure

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We studied the behavior of the plasma induced by a nanosecond infrared (1064 nm) laser pulse on a metallic target (Al) during its propagation into argon ambient gas at the atmospheric pressure and especially over the delay interval ranging from several hundred nanoseconds to several microseconds. In such interval, the plasma is particularly interesting as a spectroscopic emission source for laser-induced plasma spectroscopy (LIBS). We show a convoluted effect between laser fluence and pulse duration on the structure and the emission property of the plasma. With a relatively high fluence of about 160 J/cm² where a strong plasma shielding effect is observed, a short pulse of about 4 ns duration is shown to be significantly more efficient to excite the optical emission from the ablation vapor than a long pulse of about 25 ns duration. While with a lower fluence of about 65 J/cm², a significantly more efficient excitation is observed with the long pulse. We interpret our observations by considering the post-ablation interaction between the generated plume and the tailing part of the laser pulse. We demonstrate that the ionization of the layer of ambient gas surrounding the ablation vapor plays an important role in plasma shielding. Such ionization is the consequence of laser-supported absorption wave and directly dependent on the laser fluence and the pulse duration. Further observations of the structure of the generated plume in its early stage of expansion support our explanations. © 2013 American Institute of Physics. [<http://dx.doi.org/10.1063/1.4772787>]

I. INTRODUCTION

The performance of laser-induced breakdown spectroscopy (LIBS) is entirely determined by the optical emission property of the induced plasma following the absorption of a laser pulse on the surface of the material to be analyzed. It is not surprising that such property is sensitively dependent on the characteristics of the laser pulse. But what are the parameters for a laser pulse which are critical for LIBS? Laser wavelength and energy are clearly the most commonly considered ones in the literature.^{1–7} Experimentally, they are also the most easily determined and monitored. Pulse duration or more generally temporal profile of a pulse is often not monitored in a LIBS experiment, because of the need of additional equipment, fast photodiode and oscilloscope for example for a nanosecond pulse. However, results from numerical simulation clearly show the dependence of the property of the induced plasma on the pulse duration, even if duration variation occurs within a limited range from several to tens of nanoseconds.⁸ Up to now, experimental investigations on the effect of the pulse duration on laser ablation and on the property of the produced plasma have been mainly focused on the comparison among different regimes of nanosecond, picosecond, and femtosecond ablation.^{9–13} Refined study of the effects of temporal profile and pulse duration of a ns pulse is still needed to understand their implications in

the determination of the optical emission property of the generated plasma.¹⁴ Moreover entangled effect is expected between the above mentioned parameters characterizing a laser pulse. The convoluted effect of laser fluence and pulse duration has been studied for laser ablation process.¹⁵ For LIBS, not only the ablation but also the propagation of the ablation vapor into the ambient gas is crucial for the property of the resulting plasma and sensitively depends on the laser parameters. Dependence of the propagation behavior of the plasma on the laser pulse parameters is mainly due to post-ablation interaction of the generated plume¹⁶ with the remaining part of the laser pulse which continues to arrive after the initiation of the plasma. Plasma shielding represents one of the most significant effects of post-ablation interaction and has been observed affecting ablation efficiency as well as LIBS operation.^{17–19} However, the detailed mechanisms involved in plasma shielding and its dependence on laser parameters such as fluence and pulse duration remain still open issues.

In this paper, we report the results of our study on the convoluted effect of laser fluence and pulse duration on the properties of a plasma induced by a ns pulse. The delay interval between several hundred nanoseconds and several microseconds has been especially investigated because it corresponds to the typical operation range of LIBS. A simple configuration of ablation with an infrared (IR at 1064 nm) nanosecond pulse of a metallic target (Al) in an argon ambient gas of atmospheric pressure was used in order to stress the effect on the behavior of the plasma during its propagation into the ambient gas. The

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expansion process of the plasma was studied in two fluence regimes, a high fluence regime of 160 J/cm^2 and a low fluence regime of 65 J/cm^2 . In each of these regimes, the behavior of the plasma was observed as a function of laser pulse duration. Two types of IR laser pulses of different pulse durations of several ns and several tens of ns were used in the experiment. Time- and space-resolved emission spectroscopy²⁰ was employed to accede to the detailed diagnostics of the resulting plasmas. Significantly different influences of the pulse duration were observed in the two fluence regimes of ablation and over delay interval between several hundred nanoseconds and several microseconds. We explain such difference by considering post-ablation interaction between the generated plume and the laser pulse. Such interaction leads to laser-supported absorption waves (LSAWs)²¹ in the early stage of the propagation of the plume when the laser pulse is still present. We show in particular that the ionized layer of ambient gas generated in a LSAW plays an important role in the plasma shielding. Further observations using differential spectroscopic imaging²² in the early stage of the pulse expansion support our explanation. In the following, we will first present the experimental setup and the measurement protocols. The observed plasma behaviors will then be presented in the high fluence regime for the two types of nanosecond pulses with different durations. The results in the low fluence regime will be thereafter discussed. The effects of laser pulse duration in the two fluence regimes will be compared.

II. EXPERIMENTAL SETUP AND MEASUREMENT PROTOCOLS

A. Description of the experimental setup

The experimental setup is shown in Fig. 1. Two Nd:YAG lasers were used in the experiment. One of them was provided by Quantel laser company (Brilliant) and the other by CILAS laser company, respectively named laser “B” and laser “C” in the following. Both of them operated with a repetition rate of 10 Hz and at the fundamnet of Nd:YAG at 1064 nm. The nominal pulse duration of the laser B was 4 ns (FWHM). The pulse duration of the laser C was significantly longer of several tens of ns and dependent on

the output energy. In our experiment, ablations were performed with two pulse energies with a higher energy of 50 mJ per pulse and a lower energy of 20 mJ delivered on the surface of the target. The pulse duration of the laser C with these two output energies was, respectively, 25 and 45 ns (FWHM). Since the beam diameters were different for the two lasers, a telescope including two lenses, L1 (divergent) and L2 (convergent), was used to magnify the beam diameter of the laser C to fit that of the laser B of 6 mm. The paths of the two lasers were then superimposed by using two high reflection mirrors M1 and M2. The M2 was removable, which allowed the passage of the beam of the laser B. The both laser beams passed through an ensemble of a half-wave plate (HWP) and a Glan prism (GP). Such ensemble allowed a fine adjustment of the laser energy delivered to the target. A mechanical beam shutter (BSH) controlled the delivery of the laser pulse onto the target. A beam splitter (BS) sampled a pulse by sending 4% of it to a photodiode (PD). A synchronization signal was thus generated and used to trigger the detection system. The laser pulses were focused onto the target by a lens (L3) with a focal length of 50 mm. In order to avoid direct breakdown in the ambient gas, the focus point of the beams was located under the surface with a shift of about 1.5 mm. The resulted craters had thus a diameter larger than the waists of the laser beam. The crater diameter was measured using microscopy to be about $200 \mu\text{m}$ for the both lasers. The measured crater size was used in our experiment to estimate the diameter of the laser spot on the surface of the target. The microscopic images of the craters were also used to ensure that the laser beams were perfectly perpendicular to the surface of the target. The fluence delivered to the target (theoretically reachable if not absorbed) was thus estimated at 160 J/cm^2 at 50 mJ and 65 J/cm^2 at 20 mJ. A computer (PC) was used to ensure the synchronization of the different events in the experiment and to control the measurement procedure.

Aluminum targets used in the experiment were of two different qualities. A certified aluminum alloy (Al 89.5%, Si 8.39%, Fe 0.999%, and some traces) was used for measurements of “spectroscopy” type, whereas a pure aluminum (99.99%) was used for those of “image” type. The presence of the traces in the target for the “spectroscopy” type measurement was necessary for the implementation of multi-Saha-Boltzmann plot for the determination of the temperature of the plasma, which will be discussed in detail later in Sec. III A 2. The surface of the target was polished and cleaned prior to ablation. During a series of measurement, the target was translated using a motorized X-Y stage in order to provide a fresh surface for each burst of laser shot. The distance between the focusing lens (L3) and the target surface was kept constant during the measurement by using a monitoring system which consisted of a combination of a laser pointer with its beam in oblique incidence onto the target surface and a video camera installed above the mirror M3 (not shown in Fig. 1). A pair of tubes installed above the target and surrounding the laser ablation zone was used to bring a stream of argon gas of a fixed flow of 8 l/min , which ensured the plasma to expand into pure argon ambient at one atmosphere pressure.

FIG. 1. Experimental setup. M1-M3: Mirrors, L1-L6: lenses, HWP: half wave plate, GP: Glan prism, BSH: Beam shutter, BS: beam splitter, T: Target, F: filter, PD: photodiode, PC: computer.

The produced plasma was imaged by using a two-lens system in a 4-f configuration (L4 and L5 or L6 in Fig. 1). Two types of measurement were realized in our experiment. In the “spectroscopy” type measurement, the combination of the two lenses L4 and L5 of focal length of, respectively, 7.5 cm and 5 cm formed a reduced image of the plasma (inset (a) in Fig. 1). An optical fiber of 50 μm core diameter was positioned in the image plane in order to capture a part of emission from the plasma with a space resolution. In this paper, axial profiles will be presented, which was obtained by translating the fiber step by step along the laser incident axis (z axis) at the middle transversal position of the image of the plasma. Such detection system allowed a space resolution of 75 μm for the local detection of the plasma emission. The output of the fiber was connected to the entrance of an echelle spectrometer, which was in turn connected to an intensified charge-coupled device (ICCD) camera (Mechelle and iStar from Andor Technology). The ensemble provided a spectral range from 200 nm to 800 nm, a resolution power ($\lambda/\Delta\lambda$) of 5000 and a time resolution of about 5 ns. In the “image” type measurement, the combination of the two lenses L4 and L6 (20 cm focal length) formed a magnified image of the plasma (inset (b) in Fig. 1). The resulted image was directly recorded by the ICCD camera. In our experiment, monochromatic images were recorded corresponding to emissions from the species to be studied in the plasma. Such spectroscopic imaging was realized with the help of narrowband filters (F in inset (b) Fig. 1).

B. Experimental protocols

Due to the strong continuum emission in the early stage of the plasma expansion, the “spectroscopy” type measurement could be performed only after a certain delay of several hundred ns in our condition of ablation. Three detection windows were used to record the evolution of the plasma over the delay interval between several hundred ns and several μs . Table I provides the definition of these windows for the two ablation regimes with different fluences. In order to directly compare the emission intensities measured at different delays, the signal was divided by the gate width. This leads to a representative value of the signal for the concerned delay interval independently on the chosen gate width. The gain applied to the intensifier of the ICCD camera was chosen to be the same for all the measurements realized for a

TABLE I. Detection delay and gate width used in “spectroscopy” type measurements for the two ablation regimes with different fluences. The simplified notations will be used to designate the detection windows in this paper.

Ablation regime				
Energy (mJ)	Fluence (J/cm^2)	Delay (ns)	Gate width (ns)	Notation
50	160	500	200	D500
		1000	300	D1000
		2000	400	D2000
		350	150	D350
		800	200	D800
20	65	1500	300	D1500

given ablation regime (60/255 for ablation with 50 mJ pulse, 70/255 for ablation with 20 mJ pulse). Moreover to obtain a good signal-to-noise ratio of the spectra, each of them was accumulated over 200 laser shots. They were distributed over 20 craters with each of them ablated by 10 laser pulses. Therefore in our experiment, a burst of 10 pulses was sent to the target before the last was translated to a new position with a fresh surface in the ablation zone. The displacement between two neighbor craters was 900 μm , much larger than the crater size. Once an emission spectrum was recorded, the intensities of a certain number of emission lines were measured to extract the intensity profiles of the species in the plasma. The intensity of an emission line was measured by fitting the line profile and by calculating the surface under the profile with background subtracted.

The choice of the representative lines for different species in the plasma was guided by the consideration of a negligible self-absorption for these lines. An elementary precaution is to avoid if possible, the ground state as the lower level of the chosen transition. The selected lines are listed in Table II. The ensemble of these lines allowed a complete representation of the distributions of the element evaporated from the target as well as that contributed by the ambient gas in their neutral and ionized states. We can see that the three of them representing, respectively, aluminum ion, argon ion, and atom have a lower level with high energy. We can thus reasonably consider that they suffer less from self-absorption. This is particularly important in our experiment, because the Ar I line at 696.5 nm line was used for the determination of the electron density as we will present in Secs. III A 2 and IV A 2. The self-absorption associated with this line has been estimated as negligible in a similar condition as in Ref. 20. For aluminum atom, we could not avoid the strong resonant line at 309.3 nm, because the high degree of ionization of the aluminum vapor in the condition of our experiment prevents weak emission lines from Al atom from detection with a good enough signal-to-noise ratio, especially at short delay.

In the “image” type measurement, monochromatic images were taken for neutral and ionized aluminum as well as neutral and ionized argon at a short delay of 100 ns after the arrival of the ablation laser pulse on the target. This delay was chosen to be enough closed to the end of the laser pulses. This is necessary for the obtained results to be representative of the state of the plasma in its early stage of expansion. It was at the same time enough away from the laser pulse to avoid a too strong continuum. For each species, an emission line was chosen according to the same criterion discussed above. Since images were taken with a short delay

TABLE II. Selected lines representing the 4 species of interest in the plume with their wavelength, and energy of the upper and the lower levels. Data are extracted from the NIST database.²³

Element	Species	Transition wavelength (nm)	Upper level energy (eV)	Lower level energy (eV)
Al	Ion	281.6	11.8	7.4
	Neutral	309.3	4.0	0.0
Ar	Ion	480.6	19.2	16.6
	Neutral	696.5	13.3	11.5

when a continuum emission was still emitted from the plasma, to image the plasma with the emission line from a given species, a couple of filters was used in a configuration of differential spectroscopic imaging.²² One of these filter was on resonance with the emission line and the other one was off resonance and provided the measurement of the continuum emission in the vicinity of the emission line. The emission image of the species under study resulted from the subtraction between the two images taken successively with the two above discussed filters. Before the subtraction, the intensities of the images corresponding to the two filters were first corrected by the transmission curves of the filters. Table III shows the lines chosen for the monochromatic images of aluminum and argon together with the corresponding filters and their characteristics. For each couple of filters, the gain of the ICCD camera was chosen to fit the intensities of the resulted images. Different gains were thus chosen for different species. So the emission intensities of different species cannot be compared. However the structure of the plasma is clearly revealed with the images corresponding to each species normalized to their own maximum. The normalized images can then be superimposed in a same picture to present the structure of the plasma with emission images of different species in the plume. Similar to the “spectroscopy” type measurement, an image was the result of the accumulation over 100 laser shots distributed over 10 craters with 10 shots by crater.

III. EFFECT OF PULSE DURATION ON THE PROPERTY OF THE PLASMA IN HIGH FLUENCE ABLATION REGIME

In this section, we study the effect of the pulse duration on the behavior of the plasma induced by a pulse with an energy of 50 mJ which represents, with the 200 μm spot focused on the target surface, a fluence of about 160 J/cm². Our previous works show that such fluence is already high enough, especially for an IR pulse, to induce a strong post-ablation interaction leading to a strong plasma shielding.⁴ The behaviors of the plasmas induced by the short pulse and the long pulse will first be compared in detail over the delay interval between 500 ns and 2 μs . The observations clearly show different behaviors between the plasmas produced by these two types of pulse. In order to interpret the observed behaviors, the structure of the plasmas at a shorter delay of 100 ns will be presented in the second part of the section. The behavior of the plasma at longer delay will be thus interpreted with the help of their structure

TABLE III. Emission lines chosen to represent different species in the plume and the narrowband filters used in the “image” type measurement to get spectroscopic images of these species.

Species	Emission line (nm)	Resonant filter		Continuum filter	
		λ_{center} (nm)	Bandwidth (nm)	λ_{center} (nm)	Bandwidth (nm)
Aluminum ion	358.7	360	10	380	10
Aluminum atom	396.1	400		380	
Argon ion	487.9	488		530	
Argon atom	750.1	750		720	

and behavior at shorter delay, since such a structure is a direct consequence of the post-ablation interaction between the induced plume and the laser pulse which occurs at the very early stage of the plasma expansion.

A. Behavior of the plume in the delay interval from 500 ns to 2 μs

1. Axial profiles of emission intensity

Consider now the axial profiles of emission intensities of different species in the plasma. Such profiles are obtained with “spectroscopy” type experimental setup and measurement protocol described in Sec. II. The results are presented in Fig. 2. Look at first the profiles of neutral aluminum (Fig. 2(a)). We specify that the axial position in the figure represents the distance in the plasma with respect to the target surface. The origin of the axis corresponds to the surface. Significant difference can be observed for the plasmas induced by the two types of pulses of different durations. Laser B with 4 ns duration produces an intense emitting aluminum vapor with a large axial extension. As a function of the delay, such profile continues to propagate away from the target. Laser C with 25 ns duration on the other hand, produces a much weaker emitting aluminum vapor. In addition, the emission zone remains static in the region close to the target. The emitting zones of aluminum ions corresponding to the two types of pulse exhibit similar behaviors as the neutrals (Fig. 2(b)). Significant difference is again observed for the emission intensity profiles of neutral argon (Fig. 3(c)) between the plasmas induced by the two types of pulses. What we can see is that with laser B, the emission zone of argon overlaps quite well that of aluminum. Such distributions show the mixing between aluminum vapor and argon gas that we have reported in our previous paper.⁴ For laser C however, an emission zone of argon much more extended along the axial direction than that of aluminum can be observed. And we remark also the emission intensity from argon is stronger with laser C than with laser B at a given delay. The profiles of emission intensity from argon ions exhibit the same trend of a stronger and more extended emission zone observed from the plume induced by the longer pulse.

By taking into account the energies of the upper levels of the detected transitions of neutral aluminum and neutral argon (Table II), the emission intensity profiles presented in Fig. 2 can be used to indicate the distributions of different types of hot gases in an ablation plume. Different structures are, therefore, suggested for the plumes induced by the two different types of laser pulse over the investigated delay interval. With the short duration pulse, the plume corresponds to a mixture of hot gases of aluminum vapor and argon over the total extension of the plume (0 mm to ~ 1.8 mm), such mixture reproduces well the observation reported in our previous paper.²⁰ The aluminum vapor is evaporated from the target. While the hot argon gas is contributed by the layer of the ambient gas initially surrounding the vapor and ionized by absorption of laser energy during the process of laser-supported detonation (LSD) wave as described in Refs. 4 and 21.

With the long duration pulse, axial extension of the hot aluminum vapor is significantly reduced and confirmed in a

FIG. 2. Axial emission intensity profiles of the plasmas induced by the two types of pulses: laser B of pulse duration of 4 ns and laser C with pulse duration of 25 ns. Different species in the plasma are (a) neutral aluminum evaporated from the target, (b) corresponding ions of aluminum, (c) neutral argon contributed by the ambient gas, and (d) corresponding ions of argon.

zone close to the target surface (0 mm to ~ 0.8 mm). The emission from this zone is also much less intense than with the short duration pulse. Above the zone of mixture between aluminum vapor and hot argon gas, there is a thick layer (from 0.8 mm to ~ 2 mm) of almost pure hot argon gas. Such structure is again the consequence of a LSD wave which, in the early stage of the plasma expansion, induces the ionization of argon gas. The thick layer of hot argon gas indicates here a stronger absorption of laser energy by ionized argon for the long duration pulse. In this case, more laser energy is deposited in the layer of ionized argon gas. Correspondently, less laser energy goes to the target and to the aluminum vapor leading to a reduce emission from the aluminum vapor and its confinement within a significantly smaller volume. The effect of pulse duration in high fluence regime where the LSD wave is initiated corresponds therefore to a control of plasma shielding by absorption of laser energy in the layer of excited and ionized ambient gas. A longer pulse is more shielded because the thickness of hot and ionized ambient gas increases with the pulse duration. While a short pulse penetrates better through the excited and ionized layer of ambient gas and deposits more efficiently its energy to the ablation vapor.

2. Axial profiles of electron density and temperature

Measured profiles of the electron density and the temperature of the studied plasmas are shown in Fig. 3. These

parameters are deduced from emission spectra by using the standard plasma diagnostics methods. The electronic density is determined using the Stark broadening of the Ar I 696.5 nm line. The procedure and the used spectroscopic data are presented in our previous work.²⁰ The determination of the temperature needs the plasma to be in local thermodynamic equilibrium (LTE).²⁴ Our previous work shows that in the delay interval considered for the “spectroscopy” type measurement, LTE state represents a reasonably good approximation of the studied plasma.^{20,25} Multi-Saha-Boltzmann plots²⁶ is thus used to deduce the temperature. We estimate the relative standard deviations of the electron density and the temperature to be 15% and 10%, respectively.

We can see in Fig. 3(a) that the axial extension of the electron density corresponds to the zone of mixing between aluminum vapor and hot argon gas for the plume induced by the short laser pulse, from 0 mm to about ~ 1.4 mm for the delay D500. The same extension can be found for the temperature (Fig. 3(b)). Over the extension of the plasma, the profiles show a plateau of slow variation in the middle of the distribution. Both parameters decrease near the target surface and near the propagation front of the plume, because of the thermal conduction in the interfaces with the target and with the cold ambient gas. Look at now the profiles of the plasma induced by the long pulse. The axial profiles of the electron density and the temperature are much more extended than the profile of emission intensity of aluminum. The extension

FIG. 3. Axial profiles of the electron density (a) and the electron temperature (b) of the plasmas induced by the two types of laser pulse, laser B with pulse duration of 4 ns and laser C with pulse duration of 25 ns, at different detection delays.

of the plasma corresponds rather to that of the hot argon gas from the target surface to about 1.8 mm. Electronic population observed in the zone (from 0.8 to 1.8 mm) out of the extension of the aluminum vapor is therefore contributed by the ionization of hot argon gas. This means that in the case of ablation with the long pulse, the produced plume corresponds mainly to an argon plasma consisting of a thick excited argon layer enveloping a reduced mixing zone between aluminum and argon close to the target surface. The above observations confirm that when the LSD wave is initiated at enough high laser fluence, a longer duration pulse deposits more energy into the layer of hot gas, which leads to a deeper propagation of the LSD wave into the ambient gas. A long duration pulse is thus more strongly shielded than a short duration pulse and presents a smaller efficient for ablation and for excitation of the ablated material.

The above result is quite different from what one can learn from the literature about a two-component plasma with a mixture of a metallic vapor and a mono atomic gas like argon. In such plasma, the most of electrons would be contributed by the metallic vapor because of its lower ionization potential. However due to the specific internal structure of the plume that we show in this paper, the situation can be different. In fact when LSD wave is initiated, the layer of shocked gas situated around the axial front of the plume is ionized by laser radiation. This layer in turn efficiently

absorbs the tailing part of the laser pulse, which may have two simultaneous consequences: (i) a high ionization degree of the shocked gas, and (ii) a total shielding of laser pulse preventing the metallic vapor from its further interaction with laser radiation and its further ionization. In this situation, electrons in the plasma can be dominantly contributed by the gas in the regions where its ionization is significant.

Compare now the values of the temperature and the electron density of the plasmas induced by the two types of pulses. For a given delay, we find a higher temperature for the plasma induced by the long pulse (Fig. 3(b)). But at the same time, a lower electron density is found for it (Fig. 3(a)). Such comparison shows on the one hand, the efficiency of a long pulse to heat the ablation plume. On the other hand, it confirms the fact that a long pulse produces a plasma dominated by hot argon gas, while a short pulse generates a mixture between aluminum vapor and hot argon gas. The higher ionization potential of argon leads to a lower electron density for an Ar-dominant plasma than for an Al-Ar mixture plasma at the same temperature.

B. Behavior of the plume in the early stage of expansion

In order to correlate the structure of the plume observed in the delay interval from 500 ns to 2 μ s to its early stage of expansion, where the structure of the plume is immediately resulted from the post-ablation interaction with the laser pulse, spectroscopic images were taken for different species in the plasma by using the “image” type setup and the corresponding measurement protocol presented in Sec. II. Fig. 4 shows composite spectroscopic images of the plasma recorded at a delay of 100 ns after the arrival of the laser pulse on the target surface for ablations with the two types of laser pulses. To get such a composite image, monochromatic images are first taken for each species. The obtained monochromatic images are thus normalized, respectively, to their own maximum before being superposed on a same picture as shown in Fig. 4. We remark that Fig. 4 shows more exactly

FIG. 4. Composite spectroscopic images of the emission intensities corresponding to the lines chosen to represent the 4 different species in the plasma (Table III) taken at a delay of 100 ns with the short pulse (a) and the long pulse (b). The false colors used for representing the different species in the plasma are the red for aluminum ion, the green for neutral aluminum, the gray for argon ion, and the blue for neutral argon. The monochromatic images corresponding to different species are respectively normalized to their own maximum before being superposed on a same picture. The pictures represent real dimensions of 1.5 mm $\times$ 1.5 mm. And the bottom line corresponds to the target surface.

distributions of emission intensities of the lines chosen to representing the 4 studied species in plasma (Table III). Emission intensity is directly related to the population in the upper level of a transition (emitters related to the transition). It can be related to the number density of the corresponding species for a plasma in local thermodynamic equilibrium (LTE) by using the Boltzmann distribution law.^{27,28} In this paper however for simplicity we keep the raw intensities, because they are suitable for a qualitative representation of the species in the plasma. This procedure allows a visualization of the structure of the plume with relative positions of different emitters in the plume. False colors are used to represent the different emitters with the gray for aluminum ion, the red for neutral aluminum, the green for argon ion, and the blue for neutral argon. The pictures shown in Fig. 4 represent real dimensions of 1.5 mm $\times$ 1.5 mm. And the bottom line in the pictures corresponds to the target surface.

We can see in Fig. 4 that 100 ns after the arrival of the ablation pulse on the target surface, the both plumes present a structure dominated by a population of argon ions. Such population envelops a zone where a population of aluminum ions is observed. A population of neutral aluminum is observed for the plume induced by the short pulse. While for the plume induced by the long pulse, neutral aluminum is absent. Neutral argon is observed enveloping the main core of the plume, especially around the lower part of the plume for the plasma induced by the short pulse and in the top of the plume for the plasma induced by the long pulse. The observation of the extended zone of ionized argon confirms our

statement made in Subsection III A concerning the effect of plasma shielding by the layer of ionized argon. The elongated form observed for the population of argon ions demonstrates furthermore the origin of their generation due the LSD wave. The elongated form is here the result of the accelerated propagation of the LSD wave into the ambient gas.

IV. EFFECT OF PULSE DURATION ON THE PROPERTY OF THE PLASMA IN LOW FLUENCE ABLATION REGIME

In this section, we study the effect of the pulse duration in a low fluence ablation regime. In our experiment, a pulse energy of 20 mJ was used with a corresponding fluence of about 65 J/cm². This pulse energy was chosen because it was significantly reduced compared to the pulse energy used in the high fluence ablation regime. And at the same time, it still induced an enough strong plasma emission for an easy detection. As in the precedent section, we first present the results obtained with “spectroscopy” type of measurement in a delay interval between 350 ns and 1.5 μ s. The result of “image” type measurement at 100 ns will be then presented.

A. Behavior of the plume in the delay interval between 350 ns and 1.5 μ s

1. Axial profiles of emission intensity

Look at first the axial profiles of emission intensity of aluminum shown in Figs. 5(a) and 5(b). In contrast with the

FIG. 5. Axial emission intensity profiles of the plasmas induced by the two types of pulses: laser B of pulse duration of 4 ns and laser C with pulse duration of 45 ns. Different species in the plasma are (a) neutral aluminum evaporated from the target, (b) corresponding ions of aluminum, (c) neutral argon contributed by the ambient gas, and (d) corresponding ions of argon.

high fluence regime (Figs. 2(a) and 2(b)), the long duration pulse provided by laser C induces here a stronger emission from aluminum. The extensions of the emission intensity profiles are however quite similar for the plasmas induced by the two types of pulses. Concerning the ambient gas (Figs. 5(c) and 5(d)), the difference between the intensity profiles of the plasmas induced by the two types of pulses is much reduced here as compared to the case of high fluence ablation regime (Figs. 2(c) and 2(d)). We can observe a slightly higher intensity from neutral argon for the long pulse, while a slightly higher intensity from argon ion is observed for the short pulse. Overall the profiles exhibit similar axial extensions for the different species in the plasma. These results show for the both plasmas induced by the two types of pulses, an axial zone of co-existence between aluminum and argon. The notable difference between the plasmas induced by the two types of pulses is a significantly more intense emission from aluminum vapor with the long pulse. This behavior is just the opposite of that observed in the high fluence ablation regime.

The profiles shown in Fig. 5 indicate therefore a similar structure for the both plasmas induced by the short and the long pulses. And this structure appears quite different from that observed in the high fluence regime. First, the global extension of the plume (0 mm to ~ 1.1 mm) is significantly small than that observed in the high fluence regime (0 mm to ~ 2 mm), even though similar maximal emission intensities are recorded in the both cases. Such reduced extension of the plume corresponds to a layer of excited and ionized argon significantly thinner than in the case of the high fluence regime. This suggests a thinner ionized argon layer during the post-ablation interaction between the plume and the laser pulse. Plasma shielding by the layer of ionized argon may thus be incomplete. The laser pulse can in this case transmit through this layer and continue to deposit its energy into the aluminum vapor to further heat it. This configuration of interaction is more suitably described by laser-supported combustion (LSC) wave.²⁰ The effect of the pulse duration in a regime of low fluence where LSC is initiated is therefore to optimize the penetration of the pulse through the ionized ambient gas and to control the absorption of laser energy by the ablation vapor. In this configuration, a long pulse exhibits a higher efficiency to excite the emission from the vapor. Overall plasma shielding effect is less pronounced in the LSC regime than in the LSD regime, because less laser energy is absorbed by the layer of ionized and excited hot argon gas. Higher coupling efficiencies to the target for ablation and to the ablation vapor for heating can be therefore reached.

2. Axial profiles of electron density and temperature

Axial profiles of the electron density and the temperature are shown in Fig. 6. We can see a small extension of the plasma which fits the extension of the emission intensity profiles of aluminum and argon shown in Fig. 5 for the both plasmas induced by the short and the long pulses. We can remark also for a given delay, the plasma induced by the long pulse presents a higher temperature, which corresponds

FIG. 6. Axial profiles of electron density (a) and electron temperature (b) of the plasmas induced by the two types of laser pulses, laser B of pulse duration of 4 ns and laser C with pulse duration of 45 ns at different detection delays.

well to the fact that this pulse induces stronger emissions from aluminum and from argon. The difference with respect to the high fluence regime is that here a higher electron density is also found for the plasma induced by the long pulse. This confirms the fact that in the low fluence regime, an Al-Ar co-existing plasma is produced with the both short and long pulses.

B. Behavior of the plume in the early stage of expansion

In order to confirm the LSC wave propagation in the low fluence regime, we recorded spectroscopic images at a delay of 100 ns for the plasmas induced by the short and long pulses. The result is shown in Fig. 7 with the same presentation as in Fig. 4. Comparing the images shown in Figs. 4 and 7, we can first remark that the plumes induced in the low fluence regime presents a shape which is less elongated than the plumes induced in the high fluence regime. This means that the acceleration of the axial propagation of the plume is less strong. An isotropic expansion is actually one of the specificities of the LSC wave with respect to the LSD wave which presents an elongated form of the plume as shown in Fig. 4. Look at now the internal structure of the plumes in the low fluence regime (Fig. 7). We can see that for the both plasmas, a large emission zone of ionized and neutral aluminum is observed. Such zone occupies the main core of the

FIG. 7. Composite spectroscopic images of the emission intensities corresponding to the lines chosen to represent the 4 different species in the plasma (Table III) taken at a delay of 100 ns with the short pulse (a) and the long pulse (b). The false colors used for representing the different species in the plasma are the red for aluminum ion, the green for neutral aluminum, the gray for argon ion, and the blue for neutral argon. The monochromatic images corresponding to different species are, respectively, normalized to their own maximum before being superposed on a same picture. The pictures represent real dimensions of $1.5 \text{ mm} \times 1.5 \text{ mm}$. And the bottom line corresponds to the target surface.

plume. This observation corresponds to the transmission of the laser pulse through the ionized and excited layer of argon and the efficient absorption of laser energy by the aluminum vapor. In addition, the emission zone of neutral aluminum is larger for the plume induced by the long pulse, which corresponds well to the stronger aluminum emission observed for this pulse at longer delays. Another remarkable feature is also that the emission zone of argon ion is observed here with a significantly reduced extension compared to the high fluence ablation regime. This confirms a reduced shielding of the laser pulse by the ionized and excited layer of argon during the post-ablation interaction. Finally, an envelope of excited neutral argon is observed similar to the case of high fluence regime.

Before going to the conclusion of this work and comparing the results presented in Secs. III and IV, we remark that the morphology of the plasma, including the profiles of the electron density and the temperature, is rather similar for plasmas induced by laser pulses with same fluence but different pulse durations, thus different irradiances. Our results therefore show that in the condition of our experiment, laser fluence is a more relevant parameter to characterize the post-ablation interaction between the ablation plume and the laser radiation, and as a consequence, the expansion of the plasma into the ambient gas.

V. CONCLUSION

We have studied convoluted effect of ablation laser fluence and pulse duration on the behavior of the induced plasma during its propagation into ambient gas. A simple configuration of ablation with an infrared pulse of an aluminum target under argon gas of one atmosphere pressure was used in our study. We have chosen two fluences, 160 J/cm^2 and 65 J/cm^2 , and studied the effect of pulse duration in each of these ablation fluence regimes. The behaviors of the plasmas induced by two types of pulses with different durations of 4 ns (short pulse) and between 25 and 45 ns (long pulse) have been especially studied and compared within the interval of delay between several hundreds and several μs . This

interval is particularly interesting for LIBS operation. In order to explain the behaviors of the plasma in this delay interval, the morphology and the internal structure of the plasma in a shorter delay of 100 ns have been observed.

The ensemble of the results obtained in this study show that in the high fluence ablation regime, the post-ablation interaction corresponds well to the LSD wave. Such laser-supported propagation is characterized by an ionization of a substantial layer of ambient gas due to the absorption of laser energy in this layer. Plasma shielding in this case corresponds therefore to the screening effect due to the layer of ionized ambient gas. The absorption of laser energy accelerates the propagation of this layer into the rest part of the ambient gas still unaffected by the plasma, which leads to an elongated shape of the plume. When LSD is initiated, the effect of pulse duration is to control laser energy absorption in the ionized layer of ambient gas. In our experiment, the long pulse is severely shielded by this layer, which results in a plasma dominated by hot ambient gas. As a consequence, the heating of the ablation vapor is quite inefficient with this pulse. The short pulse is observed less shielded by the layer of ionized ambient gas. The remaining part of the pulse penetrates inside the plume and deposits its energy to the ablation vapor. A plasma is observed in this case as a mixture between the ablation vapor and the hot gas from the ambient. The excitation of the ablation vapor is much more efficient with the short pulse than with the long pulse.

In the low fluence ablation regime, the post-ablation interaction corresponds rather to the LSC wave. Such laser-supported propagation is characterized by a moderate ionization of the ambient gas. A significant part of laser pulse penetrates thus inside the plume and deposits energy into the ablation vapor. Plasma shielding in this case corresponds thus principally to the absorption by the ablation vapor. The driving force of the plasma expansion comes in this case from the center of the plume and it acts as a piston pushing the surrounding ambient gas away. Such mechanism of propagation leads to a rather spherical shape of the plume as we observed in our experiment. When LSC wave is initiated, the effect of pulse duration is to optimize the penetration through the layer of ionized ambient gas and to control the absorption in the ablation vapor. Our results show a better absorption by the ablation vapor for the long pulse, which leads to a more efficient excitation of the emission from the vapor. Such behavior is just the opposite with respect to what happens in the high fluence regime.

Finally, the qualitative explanations provided in this paper for understanding our experimental results need to be confirmed and completed by detailed numerical simulation which represents the only way to take into account all the implications of the convoluted effect of the laser fluence and the pulse duration in a process as complex as the laser ablation and the propagation of the produced plasma into the ambient gas.

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