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Absorption of a nanosecond laser pulse by a picosecond laser-induced preformed aluminum plasma

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Abstract

The LIBS (Laser-Induced Breakdown Spectroscopy) method has already demonstrated its reliability and its robustness in many situations for the multi-elemental composition determination of samples. However, certain conditions prevent a totally satisfactory determination. For instance, the method is weakly efficient to measure with accuracy the light elements concentration in metallic matrices. Since the laser pulse used to produce the plasma contributes to its heating, using an additional pulse (double pulse configuration) provides the increase in electron temperature and density without additional ablation. A better signal-to-noise ratio and a lower limit of detection can be reached. The present paper reports the results of different experiments performed to quantify the modifications induced (1) on the electron density by the second laser pulse in a preformed aluminum plasma, and (2) on the second laser pulse itself. The related experiments have been done in the case where the plasma is produced by a picosecond laser pulse and the second laser pulse is of the nanosecond type. The electron density reaches a maximum resulting from the total ionization of the aluminum plasma volume irradiated by the second laser pulse.

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1. Introduction

The Laser-Induced Breakdown Spectroscopy (LIBS) is based on the irradiance of a sample (for instance solid) to be analyzed by a laser pulse above a threshold of $\sim 10^{13} \text{ W m}^{-2}$ leading to the formation of a freely expanding plasma [1].

Two situations can be observed according to the density of free electrons inside the solid. If the material is dielectric, multiphoton ionization (MPI) contributes to the production of the first free electrons. Then, they absorb the laser light by Inverse Bremsstrahlung (IB) [2]. If free electrons exist in the sample, the MPI processes do not participate so much to the ionization of the plasma. Laser photons are absorbed by IB. In the two cases, the electron temperature T_e increases, which leads to the increase in the electron density n_e resulting from the enhancement of the collisional ionization (CI). The bonds between particles inside the sample are broken, the pressure reaches several 10^9 Pa [3] and the matter is strongly ejected from the resulting crater according to a hypersonic expansion regime [4]. The ablated material has a decreasing temperature during its expansion and contributes to a partial ionization of the components of the background gas crossed by the shockwave.

The temperature and density levels are high and allow the emission of light whose spectroscopic analysis is performed for LIBS. The light emission and therefore the collected signal radiance directly depend on the ablated mass [5]. The spectroscopic analysis shows that thermal equilibrium is reached owing to the elastic collisions at high electron density ($n_e > 10^{23} \text{ m}^{-3}$) [6]. This value of n_e coupled with the cooling of the plasma lead to a significant ionic recombination [7]. This impacts the feature

23 of the spectra mainly characterized by a continuum at early time and by ionic then
24 atomic lines at longer time.

25 Due to its easy implementation, most of the LIBS applications are based on the
26 use of a laser single pulse (SP). This explains why so many papers have been dedicated
27 to such a configuration. However, **the limit of detection of some elements is too high**
28 **and should be reduced.** Using a double pulse (DP) configuration can overcome a part
29 of these limitations by increasing the electron temperature and density within the
30 plasma. Despite its difficult implementation, this configuration represents a serious
31 and seducing alternative.

32 DP configurations have been therefore tested in many situations. Most of the
33 experimental setups are of the colinear type. This means that the two pulses belong
34 to the same optical axis, systematically perpendicular to the sample. They use one [8]
35 or two laser sources in ns/ns [9, 10, 11, 12, 13] or fs/ns [14, 15] configurations for
36 the first/second pulse, respectively. They often use infrared radiation for the first
37 pulse [9, 10, 11, 12, 13] and either infrared [9, 10, 11, 12, 15] or visible [13, 14]
38 radiation for the second pulse. For colinear configurations, the use of optical fibres
39 can be very useful [16]. For non colinear configurations, an angle of 90 degrees is
40 considered, using one [14] or two [17] laser sources.

41 **From a more fundamental point of view, detailed studies have been already per-**
42 **formed to put into light the mechanism of interaction of the second pulse with the**
43 **plasma produced by the first pulse. An important part of these studies has been**
44 **focused on plasma experiments under water [18]. But most of them has been per-**
45 **formed in a gaseous environment. A higher temperature level has been put forward**
46 **to explain the change in the emitted light [19]. The morphology modifications have**
47 **been also studied using shadowgraphy [20]. The influence of the second pulse on the**
48 **ablation have been also characterized [21] as well as the one due to the background**

49 gas [22]. An interesting review of these questions can be found in [23]. The DP
50 configuration has been also studied from the modeling point of view [24].

51 The works cited above are focused on the improvement induced by the use of the
52 second pulse in the framework of the LIBS technique. As colinear configurations are
53 mainly implemented, the different delays between the two pulses open the question
54 of the influence on the reported improvement of the additional ablation resulting
55 from the partial transparency of the first laser pulse-induced plasma. Indeed, if the
56 delay is sufficiently long to correspond to a too weak electron density, the second
57 pulse reaches the sample despite its partial absorption by the plasma. Then an over-
58 ablation takes place. The relative contribution of this second step of ablation and of
59 the light absorption by the plasma in the enhancement of the relevant lines is difficult
60 to estimate [25]. This prevents an optimization of the LIBS technique based on an
61 unambiguous understanding of the underlying physics. In our opinion, in parallel
62 with the technical verifications of the advantages of DP configurations in LIBS, a
63 more fundamental study rather dedicated to the interaction between the second laser
64 pulse and the plasma produced by the first laser pulse excluding any influence of the
65 over-ablation has to be performed.

66 The present paper deals with this purpose from the experimental point of view.
67 Since Aluminum is certainly the element mainly studied in laboratory laser-induced
68 plasmas, Al samples have been used. They are irradiated first by a picosecond laser
69 pulse to **reduce as much as possible the** interaction with the plasma in expansion.
70 Then, a second laser pulse of the nanosecond type is used to increase n_e and T_e . The
71 propagation axis of the second pulse is parallel to the sample to avoid any additional
72 ablation when the time delay between the two pulses is increased. In the following,
73 the experimental setup is described. The experiments are then reported. They
74 have been performed to characterize the influence at given energy of the time delay

75 between the two pulses. The energy is kept constant to enable the cross comparison
76 of the maximum electron density obtained after absorption. The dynamics of n_e is
77 deduced from the spectroscopic analysis and correlated to the spectral absorptivity
78 of the second laser pulse by the absorbing plasma.

79 **2. Experimental setup**

80 *2.1. The “PLEIADES” platform*

81 The experiments have been performed on the PLEIADES (Plasmas by LasEr Ir-
82 rADIations and their Experimental Studies) platform of the CORIA laboratory [26].
83 Its modularity allows the study of many kinds of samples irradiated in various ways.
84 As discussed in Introduction, most of the DP experiments are based on two consec-
85 utive coaxial beams shifted in time [27] perpendicular to the sample. Due to the
86 expansion of the plasma, a decrease in density takes place. As a result, the absorp-
87 tivity decreases, which leads to an additional ablation. It is therefore impossible
88 to evaluate unambiguously the influence of the energy absorbed by the plasma. To
89 avoid this difficulty, a perpendicular configuration has been used on PLEIADES.
90 The first pulse corresponds to an optical axis perpendicular to the Al sample that
91 produces the ablation. The second pulse is parallel to the sample. Figure 1 displays
92 the experimental setup where the first optical axis appears in red and the second
93 optical axis appears in green. DURAL (alloy of 95 % Al, 4 % Cu, 0.5 % Mg and
94 0.5 % Mn) is used as sample for the experiments and is placed in air at atmospheric
95 pressure on a central (xyz)-stage holder.

96 The photons belonging to the first pulse are characterized by the wavelength
97 $\lambda_1 = 1064$ nm. They are produced by an EKSPLA PL-2251 source delivering the
98 energy $\bar{E}_1 = 20$ mJ over the pulse duration $\tau_1 = 30$ ps. Its standard deviation σ_{E_1}

99 does not exceed 1 %. The laser beam with the quality factor $M_1^2 \approx 2$ is focused
 100 on the aluminum sample using a plano-convex lens of $f_1 = 125$ mm as focal length.
 101 The beam diameter at $1/e^2$ is $2\omega_1 = 5$ mm before the lens. Its diameter on the
 102 sample is then $2\omega'_1 = 2M_1^2 \frac{\lambda_1 f_1}{\pi\omega_1} \approx 70$ μm . The mean fluence on the sample is then
 103 $F_1 = \frac{\bar{E}_1}{\pi\omega_1'^2} \approx 5$ MJ m^{-2} . The second pulse is produced by a Q-SMART 450 laser
 104 source at the wavelength $\lambda_2 = 532$ nm according to a pulse duration of $\tau_2 = 5$ ns.
 105 The energy is $\bar{E}_2 = 15.2$ mJ with a standard deviation σ_{E_2} lower than 4 %. The
 106 second pulse is focused at the distance $H + 620$ μm from the sample surface on the
 107 first optical axis within the plasma produced by the first pulse using a $f_2 = 150$ mm
 108 focal length plano-convex lens according to figure 2. Before the lens, the second laser
 109 beam has a diameter $2\omega_2 = 6.5$ mm at $1/e^2$ of the central irradiance. With a beam
 110 quality factor $M_2^2 \approx 2$, its diameter within the plasma is $2\omega'_2 = 2M_2^2 \frac{\lambda_2 f_2}{\pi\omega_2} \approx 30$ μm .
 111 This corresponds to a mean fluence of $F_2 = \frac{\bar{E}_2}{\pi\omega_2'^2} \approx 20$ MJ m^{-2} . With this value,
 112 we experimentally verify that the laser irradiance is lower than but very close to
 113 the breakdown threshold in ambient air. In this context, it is therefore possible to
 114 maximize the effect of the absorption on the plasma and to avoid as much as possible
 115 the formation of an air plasma when the aluminum plasma is not produced.

116 After collection by a plano-convex lens leading to a magnification $Ma = 1.6$, a part
 117 of the radiation produced by the plasma is analyzed using a Roper SCT-320 spec-
 118 trometer (focal length $f = 320$ mm, aperture f/4.6, grating with 2400 grooves mm^{-1} ,
 119 entrance slit width of $f_e = 10$ μm) equipped with a PI-MAX 4 camera (HBf emiCCD
 120 sensor with a maximum quantum efficiency around 450 nm and an elementary pixel
 121 size of 13×13 μm^2). Calibrations have been performed prior to the experiments.
 122 Therefore each image obtained on the sensor (1024×1024 pixels) of the camera
 123 can be converted in a three-dimensional (λ, z, L_λ) image where L_λ is the spectral
 124 radiance (cf. figure 2) taking into account the acquisition gate width of 20 ns.

125 In support of the spectroscopic analysis, the absorption of the second pulse is
126 characterized using a high laser power and energy meter (Ophir Nova II) located on
127 the second optical axis, beyond the plasma.

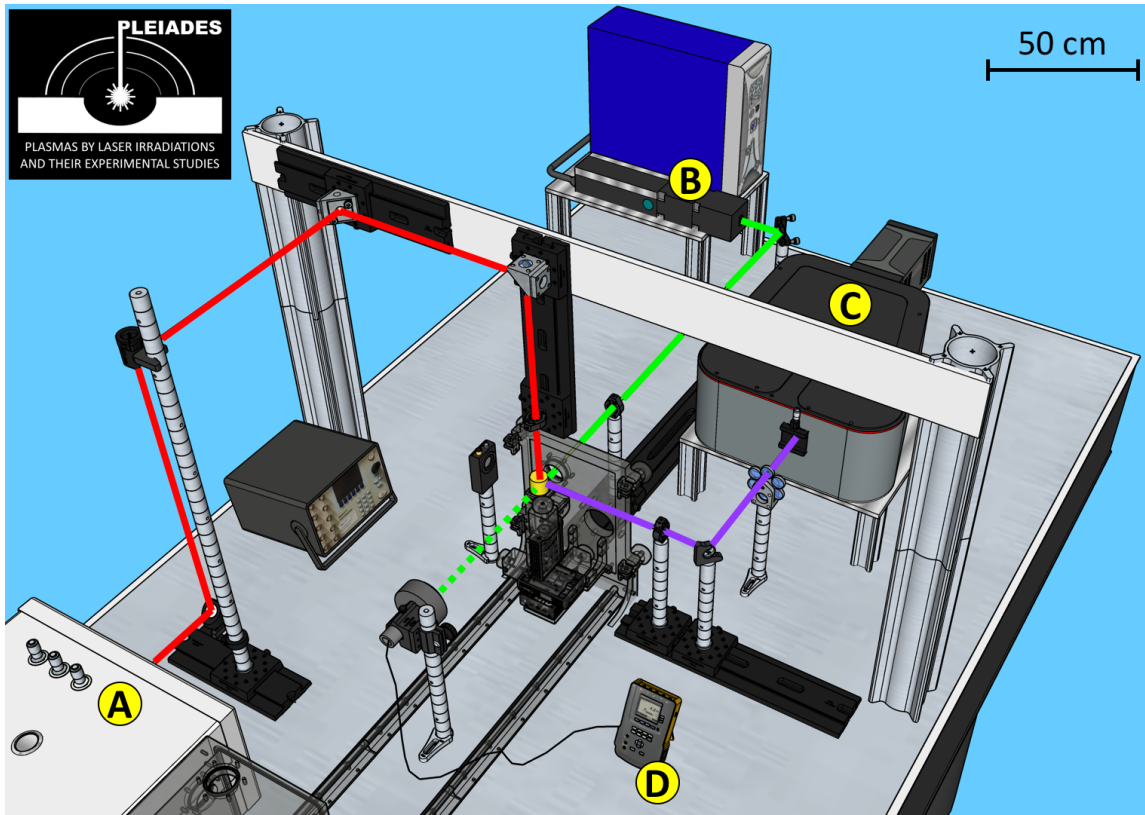


Figure 1: Schematic view of the platform PLEIADES.

The first laser source on the left produces infrared photons along the red axis. After focus, the plasma in yellow is generated on the Al sample placed on the central (xyz)-stage holder. The second laser source in the background produces visible photons along the green axis. They are focused in the plasma whose spectroscopic analysis is performed by the spectrometer on the purple axis.

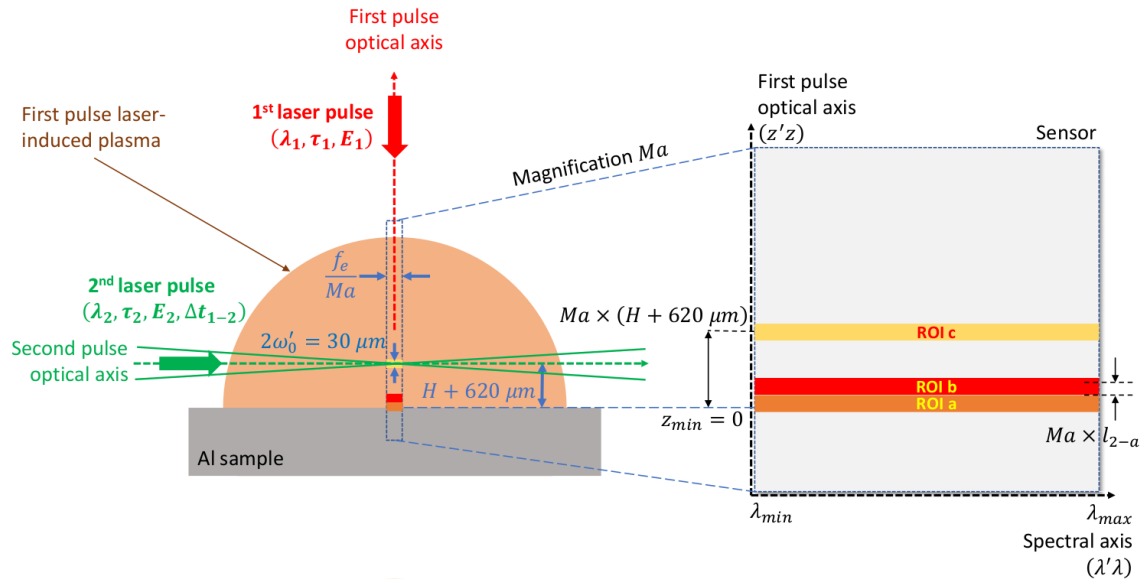


Figure 2: Relative sensor/plasma arrangement in the case where the second optical axis corresponds to the location of ROI c ($H = 1$ mm). When $H = 0$ mm, the distance between the upper border of the ROI a and the second optical axis is l_{2-a} .

128 *2.2. Experimental protocol*

129 In the following, three Regions of Interest (ROIs) of 50 rows of pixels on the
130 sensor of the camera are used. This choice of 50 rows (corresponding to a height of
131 $\delta = 400 \mu\text{m}$ in the plasma) leads the second optical axis to be almost centered on
132 ROI c when $H = 1 \text{ mm}$ (cf. figure 2) and to belong to ROI b when $H = 0 \text{ mm}$.
133 Except when a ROI coincides with the second optical axis, these 50 rows correspond
134 to quasi uniformly emitting plasma regions. They enable to spatially estimate the
135 characteristics of the plasma reheated by the second pulse. These ROIs denoted as
136 ROI a, ROI b and ROI c in the following are located at the distance $z_a = -0.16$,
137 $z_b = +0.24$ and $z_c = +1.36 \text{ mm}$ from the sample surface, respectively. The choice
138 of these values has been guided by the features of the emission after a preliminary
139 study not reported here.

140 The two parameters controlling the absorption of the second pulse by the plasma
141 are

- 142 • the distance $H + 620 \mu\text{m}$ between the surface of the sample and the optical
143 axis of the second pulse,
- 144 • the time delay Δt_{1-2} between the two laser pulses.

145 The distance H is verified by imagery on the spectrometer used at the interferometric
146 order 0 and is modified using the (xyz)-stage with an accuracy of $1 \mu\text{m}$. Since the
147 Rayleigh length of the first laser pulse is higher than H , changing the location of
148 the sample does not significantly modify the irradiance. The time delay Δt_{1-2} is
149 controlled by a pulse generator BNC 577 and is verified using a fast photodiode
150 recording a signal proportional to the plasma radiance.

151 Table 1 summarizes the experimental conditions $(H, \Delta t_{1-2})$ considered for the
152 spatial and temporal shifts between the laser pulses. They have been chosen to cover
153 a wide range of situations. $H = 0 \text{ mm}$ corresponds to the second pulse appearing in

154 the ROI b. $H = 1$ mm is also tested. For each value of H , a time delay Δt_{1-2} of
 155 150, 250 and 350 ns is considered. These conditions enable to estimate the influence
 156 of the initial n_e value on the absorption.

Experiment no j	1	2	3	4	5	6
H (mm)	0	0	0	1	1	1
Δt_{1-2} (ns)	150	250	350	150	250	350

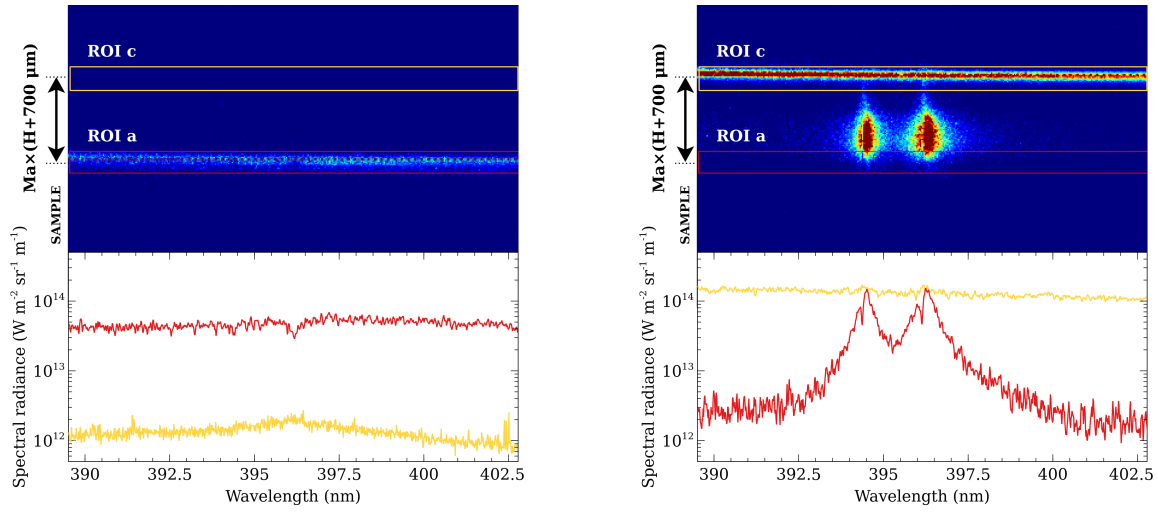
Table 1: Tested experimental conditions.

157 3. Results

158 3.1. Observed spectra

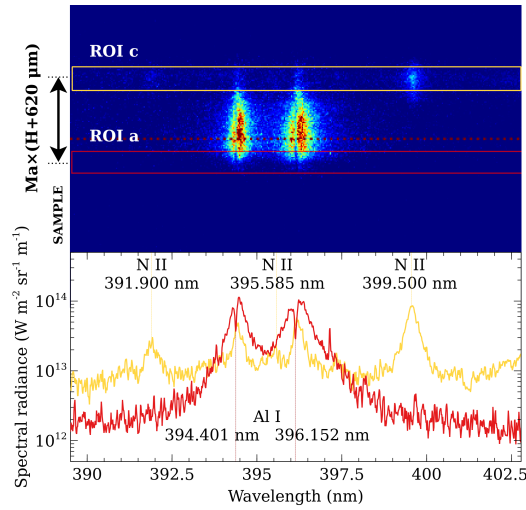
159 Figures 3a, 3b and 3c display typical images obtained on the sensor in the vicinity
 160 of $\lambda = 396$ nm. This wavelength has been chosen since it corresponds to the Al I
 161 resonance doublet. These images have been obtained with an acquisition time t of
 162 40, 380 and 480 ns for $(H, \Delta t_{1-2}) = (1$ mm, 350 ns). The plasma is formed by the
 163 first pulse on the sample at time $t = 0$. The spectra obtained from the ROIs are
 164 also displayed after conversion using the calibration. They correspond to the [390,
 165 402] nm spectral range. The spectra exhibit lines whose dynamics strongly depends
 166 on the distance from the sample.

167 The well-known Al I resonance doublet is observed. **The spectra displayed on**
 168 **figures 3b and 3c show that the concerned lines are not optically thin. Indeed, the**
 169 **upper level of the transition is the same for the two lines while the Einstein coefficient**
 170 **A_{ki} for spontaneous emission is almost in a ratio of 2 (cf. table 2). The intensity**
 171 **of the line at 396.152 nm should be twice the one of the line at 394.401 nm. Two**
 172 **contributions are responsible for this situation. First the plasma column heated**



(a) $t = 40$ ns

(b) $t = 380$ ns



(c) $t = 480$ ns

Figure 3: ROIs and associated spectra for $\lambda \sim 396$ nm at the acquisition time 40, 380 and 480 ns after the production of the plasma by the first pulse at $t = 0$. The plasma is reheated by the second pulse in the conditions $(H, \Delta t_{1-2}) = (1 \text{ mm}, 350 \text{ ns})$ of the experiment no 6. The red frames and spectra correspond to the plasma close to the sample (ROI a). The yellow frames and spectra correspond to the plasma reheated by the second pulse (ROI c).

173 by the second pulse self-absorbs the radiation. Second, the surrounding plasma
174 containing Al atoms on their ground and first excited electronic states absorbs the
175 radiation emitted at the center [28]. The relative estimate of these two contributions
176 is not an easy task since it depends on the density of the lower level of the concerned
177 transitions and on the crossed lengths.

178 Over the spectral range [390, 402] nm, some Al II transitions could be observed
179 when the second pulse is implemented. The related lines correspond to the wave-
180 length 390.068 nm ($E_k = 10.598$ eV, $g_k = 5$, $A_{ki} = 4.8 \times 10^5$ s⁻¹) and a multiplet with
181 a mean wavelength of 399.6 nm ($E_k = 18.164$ eV, $g_k = 21$, $A_{ki} = 7.05 \times 10^6$ s⁻¹). As-
182 suming the local thermodynamic equilibrium and neglecting any absorption process,
183 the radiance ratio $I_{390.068}/I_{399.6}$ between these lines is 5.7 at 15 000 K, 1.3 at 20 000 K
184 and 0.5 at 25 000 K. On the spectrum of the ROI c observed at $t = 480$ ns on figure 3c,
185 the line around 399.5 nm could be ascribed to an Al II transition. In this case, since
186 the previous radiance ratio is close to unity, the Al II transition should be observed
187 at 390.068 nm. But, the spectrum does not exhibit any lines at this wavelength. This
188 means that the line at around 399.5 nm cannot be ascribed to an Al II transition. If
189 the line around 399.5 nm is due to a N II transition, other N II transitions should be
190 observed. Two other N II transitions can indeed be observed on the [390, 402] nm
191 spectral range. The first one corresponds to a wavelength of 391.900 nm and the
192 second one corresponds to a wavelength of 395.585 nm. We can calculate the ratios
193 in case of local thermodynamic equilibrium. The ratio $I_{391.900}/I_{399.500}$ equals 0.08 at
194 15 000 K, 0.12 at 20 000 K and 0.15 at 25 000 K. The ratio $I_{395.585}/I_{399.500}$ is equal
195 to 0.1 for any value of T (the upper level of the transition corresponds to the same
196 excitation energy). These values are in pretty good agreement with the yellow spec-
197 trum of figure 3c. We can conclude that the line around 399.5 nm is due to a N II
198 transition as the other lines observed at 391.9 and 395.6 nm. Similar characteristics

199 have been already observed [29]. Except at 394.401 and 396.152 nm, aluminum does
 200 not radiate and the other lines are due to N II transitions. Table 2 lists the lines
 201 observed.

Line	Transition $i - k$	λ (nm)	A_{ki} (s^{-1})	$g_i - g_k$	E_i (eV) - E_k (eV)	ω_{ki} (nm)	Ref.
Al II	$3s3d\ ^3D_2 - 3s4f\ ^3F_3^o$	358.707	2.09×10^8	5 - 7	11.847 - 15.302	1.60×10^{-2}	[30]
N II	$2s^22p3p\ ^1P - 2s^22p3d\ ^1P_1^o$	391.900	7.56×10^7	3 - 3	20.409 - 23.572	3.54×10^{-2}	[31]
Al I	$3s^23p\ ^2P_{1/2}^o - 3s^24s\ ^2S_{1/2}$	394.401	4.99×10^7	2 - 2	0.000 - 3.143	1.54×10^{-2}	[32]
N II	$2s^22p3s\ ^3P_1^o - 2s^22p3p\ ^1D_2$	395.585	1.21×10^7	3 - 5	18.466 - 21.600	2.77×10^{-2}	[31]
Al I	$3s^23p\ ^2P_{1/2}^o - 3s^24s\ ^2S_{1/2}$	396.152	9.85×10^7	4 - 2	0.014 - 3.143	1.54×10^{-2}	[32]
N II	$2s^22p3s\ ^1P_1^o - 2s^22p3p\ ^1D_2$	399.500	1.22×10^8	3 - 5	18.497 - 21.600	2.80×10^{-2}	[31]
Al II	$3p^2\ ^1D_2 - 3s4p\ ^1P_1^o$	466.306	5.81×10^7	5 - 3	10.598 - 13.256	6.15×10^{-2}	[33]

Table 2: Lines and related transitions used along the present study. The given reference corresponds to the value of the full width at half maximum of the Stark effect-broadened line (at a reference electron density n_e^* of $10^{22}\ m^{-3}$ for the neutrals and of $10^{23}\ m^{-3}$ for the ions).

202 3.2. Direct analysis of the spectra

203 The spectra related to the Al I resonance doublet on figs 3b and 3c exhibit sig-
 204 nificant Stark broadening and shift. The wavelength corresponding to the emission
 205 without Stark shift is obtained by observing the depletion of the spectral radiance re-
 206 sulting from the absorption due to the plasma cold edge. This depletion corresponds
 207 to a ratio of 1/3 with respect to the maximum spectral radiance observed. The N II
 208 transition at 399.500 nm is observed on the ROI c spectrum at $t = 480$ ns only. At
 209 the same time, the resonance doublet lines are also observed at the same location.
 210 This means that (1) the expansion of the aluminum plasma is very rapid, (2) a mix-
 211 ing takes place with the background gas, (3) this mixing leads to the dissociation of
 212 N_2 and (4) the plasma is clearly reheated by the second pulse.

213 Figures 4 illustrates the evolution in time of the maximum spectral radiance from
 214 the ROIs a and c obtained for the lines of the aluminum resonance doublet lines and

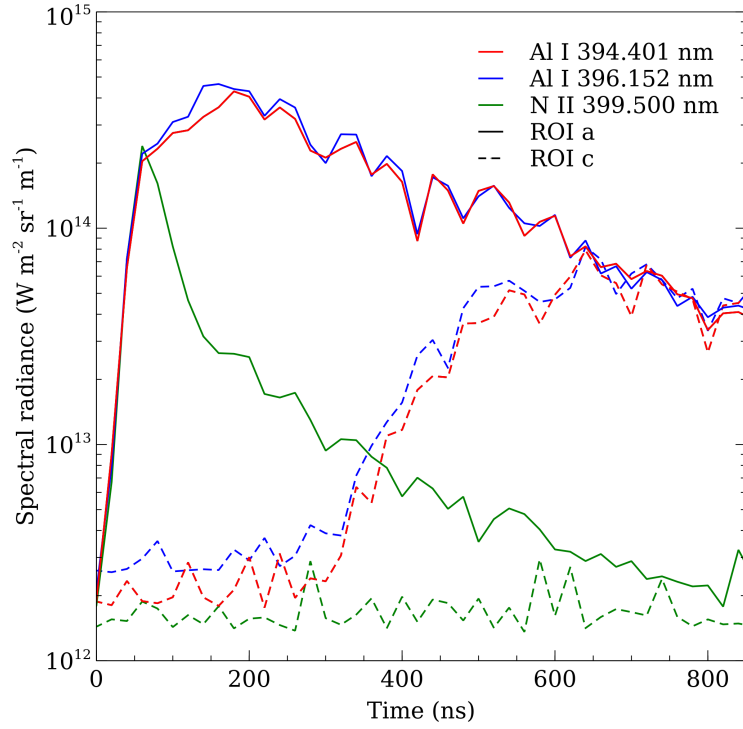
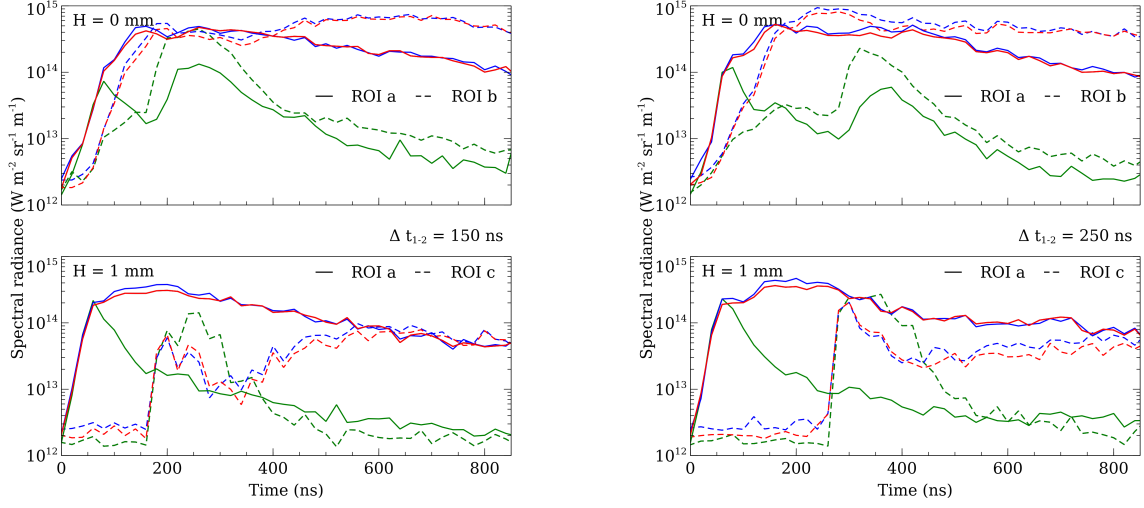


Figure 4: Time evolution of the spectral radiance at the wavelength of the Al I and N II transitions of figure 3 obtained without second pulse close to the sample's surface (ROI a, continuous lines) and ~ 1 mm above (ROI c, dashed lines).

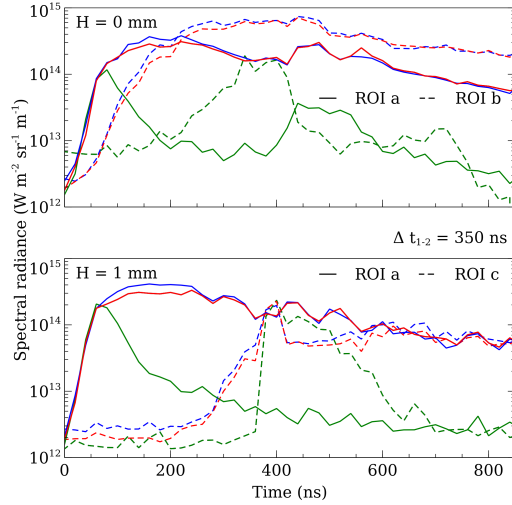
215 for the ionic nitrogen line at 399.500 nm. This evolution has been obtained without
 216 irradiation by the second pulse. On this figure we can observe the sudden increase
 217 in the spectral radiance close to $t = 0$ resulting from the emission of the initial
 218 continuum. Around $t = 60$ ns, the lines start to appear in the spectra and the
 219 spectral radiance globally decreases. At $z_c = 1.36$ mm from the surface where ROI c
 220 is located, a very low value of the spectral radiance is observed, which corresponds
 221 to the noise. From $t' = 320$ ns, the spectral radiance starts to increase. This is due
 222 to the propagation of the plasma from the surface of the sample to this position. An
 223 estimate of the mean plasma speed over t' can therefore be deduced. This value is
 224 $z_c/t' = 4.2$ km s⁻¹. Thus, the expansion is hypersonic.

225 In the case where the second pulse is used, the previous temporal analysis can
 226 be also performed. Figures 5a, 5b and 5c display the related evolutions. Concerning
 227 the figures where $H = 0$ mm, a time delay between the increase in spectral radiance
 228 related to the Al I transitions for the ROIs a and b of around $t'' = 50$ ns can be
 229 logically ascribed to the plasma expansion close to $t = 0$. Beyond the estimate of
 230 the mean speed over t' mentioned previously, the initial speed can be obtained by
 231 the ratio $z_b/t'' = 4.8$ km s⁻¹. This speed is logically higher than the mean one
 232 since the propagation requires a significant momentum transfer to the background
 233 gas which brakes down the plasma expansion. This value of 4.8 km s⁻¹ is in very
 234 good agreement with the value calculated by our model CoRaM-Al implemented in
 235 an Eulerian code [3] developed for similar situations, but with a lower energy $\bar{E}_1 =$
 236 10 mJ. **The calculation is based on the following assumptions. The picosecond laser**
 237 **pulse produces the isochoric heating and ionization of the matter in the crater. Due to**
 238 **the strong increase in pressure, this matter hemispherically expands. Its contact with**
 239 **ambient atmosphere leads to momentum transfer according to a hypersonic regime**
 240 **which produces a strong shockwave freely expanding hemispherically. The mass,**



(a) Experiments 1 and 4.

(b) Experiments 2 and 5.



(c) Experiments 3 and 6.

Figure 5: Time evolution of the spectral radiance at the wavelength of the Al I and N II transitions obtained close to the sample's surface (ROI a, continuous lines) and over the ROI corresponding to the position of the second pulse (dashed lines, ROI b for $H = 0$ mm and ROI c for $H = 1$ mm) for the different values of Δt_{1-2} (colors similar to figure 4).

241 energy and momentum conservation equations are solved in time for these two layers
 242 separated by the contact surface assuming a homothetic speed field. In addition,
 243 the ejected matter is assumed out of thermodynamic equilibrium by considering the
 244 different excited states of aluminum. The input parameters are the energy delivered
 245 by the picosecond laser pulse \bar{E}_1 , the ablated mass M and the pressure p_0 of the
 246 ambient gas. The model has been developed considering N_2 for this gas rather than
 247 air. All results related to the conditions $\bar{E}_1 = 10$ mJ, $M \sim 10^{-10}$ kg and $p_0 = 10^5$ Pa
 248 can be found in [3]. In particular, the evolution in time of the contact surface speed
 249 is given. A value of ~ 4 km s^{-1} is obtained at 50 ns. Performing the ablation with
 250 an energy multiplied by a factor of 2 leads to an ablation rate (depth of the ablated
 251 layer) 50 % higher [26]. As a result, the ablated volume is higher and the energy per
 252 unit mass in the plasma as well as its pressure is almost unchanged. As a result, the
 253 expansion is more or less the same at $\bar{E}_1 = 10$ or 20 mJ. The present experimental
 254 results can then be compared from the expansion point of view with those obtained
 255 by the modeling at lower value of \bar{E}_1 . The agreement is therefore satisfactory.

256 Moreover, comparing ROIs a and b for $H = 0$ mm for the N II transition, we can
 257 see that when the second pulse reaches the plasma, the increase in spectral radiance
 258 can also be observed later on the ROI a. This delay is approximately $\tau_{150} = 30$ ns for
 259 $\Delta t_{1-2} = 150$ ns, $\tau_{250} = 50$ ns for $\Delta t_{1-2} = 250$ ns and $\tau_{350} = 150$ ns for $\Delta t_{1-2} = 350$ ns.
 260 This means that the laser energy absorbed by the plasma at the center of ROI b does
 261 not remain on the optical axis, but diffuses around and in part toward the surface
 262 of the sample. This energy diffusion process takes place simultaneously with the
 263 electron diffusion according to a similar diffusion coefficient [36]. The time of diffusion
 264 increases when the absorbed energy is lower, therefore when the temperature and
 265 electron density reached after absorption are lower. This is an indirect proof of the
 266 better energy absorption within the plasma of the second laser pulse in higher density

267 conditions. Let us note l_{2-a} the distance between the location of the second optical
 268 axis and the upper limit of the ROI a when $H = 0$ mm (cf. figure 2). If we assume
 269 the delay $\tau_{\Delta t_{1-2}}$, the diffusion over l_{2-a} and the diffusion coefficient $D_{\Delta t_{1-2}}$ driven by
 270 the classical diffusion law

$$\tau_{\Delta t_{1-2}} \approx \frac{l_{2-a}^2}{D_{\Delta t_{1-2}}} \quad (1)$$

271 the diffusion coefficient $D_{\Delta t_{1-2}}$ consequently ranges between 1 and 5 m² s⁻¹ approx-
 272 imately, according to the value of Δt_{1-2} . The distance l_{2-a} is 380 μ m. These values
 273 are in satisfactory agreement with those determined either in plasmas at atmospheric
 274 pressure [37] or at low pressure [38] in similar conditions from the temperature point
 275 of view.

276 We can also note that this higher energy induced by the diffusion process leads
 277 to a higher spectral radiance for the Al I transitions for long time. Using the second
 278 pulse to increase the signals without additional ablation is therefore possible. Sys-
 279 tematically, the spectral radiance for the Al I transitions is indeed higher over the
 280 ROI b than over the ROI a for acquisition times $t > 400$ ns. A ratio of a factor
 281 of 4 can be reached at $t = 850$ ns. This effect is also observed for the spectral ra-
 282 diance corresponding to the N II transition which is also globally increased by the
 283 absorption of the second pulse, but with a lower ratio.

284 We have deduced from figure 4 that the characteristic time scale of the plasma
 285 propagation from the sample to the position $z = 1.36$ mm is close to $t' = 320$ ns. As a
 286 result, producing an increase in the spectral radiance of the lines of interest with the
 287 second laser pulse should not be possible if $\Delta t_{1-2} < t'$ for $H = 1$ mm. Nevertheless,
 288 it is possible as illustrated by figures 5a and 5b. This apparent contradiction can
 289 be easily explained by the significant fluctuations of the plasma expansion. These

290 fluctuations are put in evidence by monitoring the spectral transmittivity of the
291 aluminum plasma at each nanosecond pulse. These fluctuations are analyzed in
292 section 3.4. The following comments then refer to the situations corresponding to
293 an expansion fast enough to induce the absorption of the nanosecond pulse by the
294 aluminum plasma for any value of Δt_{1-2} for $H = 1$ mm.

295 The ROI a is obviously too far from the second optical axis to be influenced by
296 the energy absorption and diffusion. The time evolution of the ROI a is indeed the
297 same for figures 5a, 5b and 5c in these conditions. Conversely, the ROI c exhibits
298 strong modifications induced by the second pulse. A continuum is produced at $t =$
299 Δt_{1-2} and just after the absorption (continuum illustrated by figure 3b), the spectral
300 radiance corresponding to the N II transition is higher than the one corresponding
301 to the Al I transitions. Then the order is reversed. It is interesting to note that
302 the spectral radiance of the Al I transitions at $t = 850$ ns becomes the same for the
303 two ROIs. The energy of the second pulse absorbed by the plasma has therefore no
304 influence for long times.

305 *3.3. Electron density dynamics*

306 The estimate of the electron density from the full width at half maximum (FWHM)
307 can be obtained with the lines only when they are optically thin. When they are
308 absorbed as the Al I resonance doublet (see section 3.2), other lines must be used.

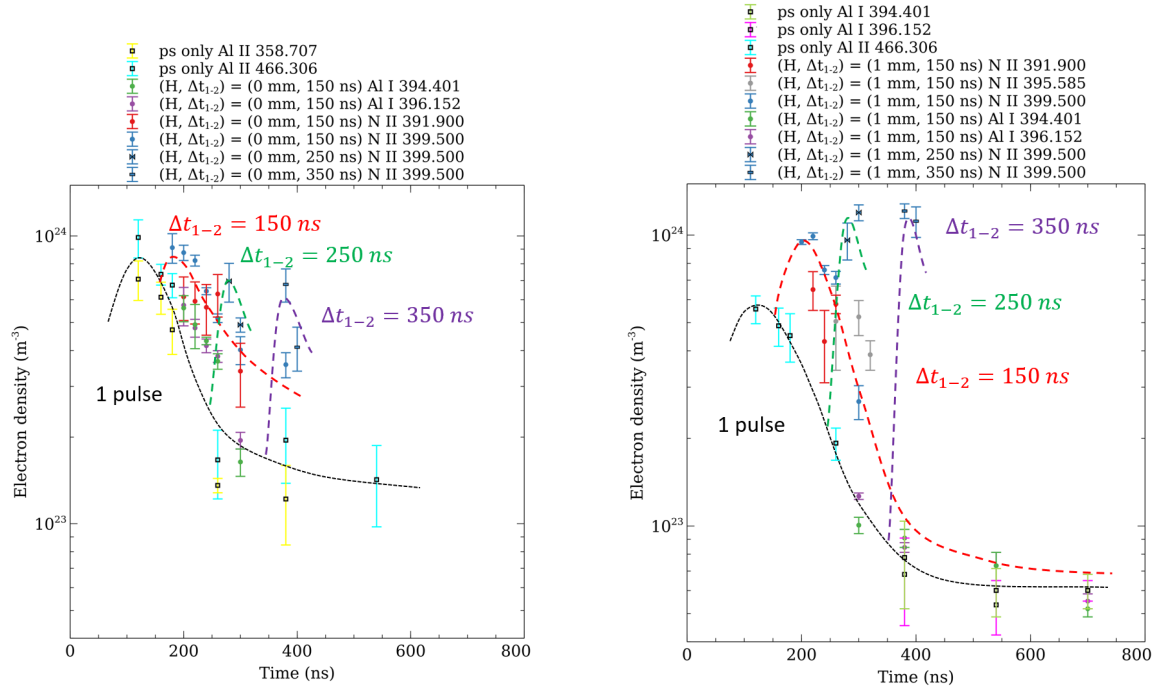
309 Table 2 lists the different transitions and their characteristics exploited for the
310 determination of n_e based on the FWHM. The additional lines with respect to the
311 ones observed on figures 3b and 3c have been chosen because they significantly ra-
312 diate. In table 2, the electron Stark broadening parameter ω_{ki} is listed. The related
313 value is used to determine the electron density n_e from the comparison between the
314 experimental full width at half maximum of the lines and the one classically given

315 by

$$\Delta\lambda_{ki}(n_e, T_e) = 2\omega_{ki}(T_e)\frac{n_e}{n_e^*} + \Delta\lambda_{ki}^{ionic}(n_e, T_e) \quad (2)$$

316 In the previous equation, $\Delta\lambda_{ki}^{ionic}(n_e, T_e)$ is the ionic contribution to the Stark width
317 and n_e^* is an electron density of reference (10^{22} m^{-3} for neutrals lines and 10^{23} m^{-3}
318 for ionic lines) at which $\Delta\lambda_{ki}(n_e^*, T_e) = 2\omega_{ki}(T_e)$ with $\Delta\lambda_{ki}^{ionic}(n_e, T_e)$ negligible. The
319 electron Stark broadening parameter $\omega_{ki}(T_e)$ weakly depends on the electron tem-
320 perature T_e and has been considered here as independent.

321 With these data, we have derived the evolution in time of n_e obtained in the
322 plasma produced by the picosecond laser pulse only. Figures 6a and 6b display this
323 evolution. It is interesting to note that the results satisfactorily match the values
324 predicted by our model CoRaM-Al implemented in an Eulerian code [3]. **Indeed,**
325 **in the conditions given in section 3.2, the calculated electron density reaches its**
326 **maximum value of $2 \times 10^{24} \text{ m}^{-3}$ at time 90 ns and decreases to $5 \times 10^{23} \text{ m}^{-3}$ at**
327 **time 500 ns.** In addition, they are in very good agreement with the values reported
328 recently by Cao *et al.* [39] in very similar conditions from the fluence point of view.
329 In fact their values are weakly higher than ours since the fluence in their experiment
330 is a bit higher. We have also measured this evolution when the second pulse is
331 absorbed. This has been performed in the case $\Delta t_{1-2} = 150 \text{ ns}$ (experiments no 1
332 and 4) for which the density in the plasma is more significant than in the other
333 conditions when the absorption takes place. This evolution is displayed on figures 6a
334 and 6b for $H = 0 \text{ mm}$ and $H = 1 \text{ mm}$, respectively. The uncertainty displayed on
335 these figures results from the uncertainty in the determination of $\Delta\lambda_{ki}$ on the treated
336 spectra. These evolutions have been also obtained for $\Delta t_{1-2} = 250$ and 350 ns . But,
337 for the sake of clarity, only the maximum electron density just after the absorption



(a) $H = 0$ mm. Experiments no 1 ($\Delta t_{1-2} = 150$ ns), 2 ($\Delta t_{1-2} = 250$ ns) and 3 ($\Delta t_{1-2} = 350$ ns).

(b) $H = 1$ mm. Experiments no 4 ($\Delta t_{1-2} = 150$ ns), 5 ($\Delta t_{1-2} = 250$ ns) and 6 ($\Delta t_{1-2} = 350$ ns).

Figure 6: Time evolution of the electron density n_e (trends in colored dashed lines) and comparison with the evolution obtained without the second pulse (trends in thin dashed black lines).

338 has been indicated on figures 6a and 6b. We can observe that the absorption of
 339 the second pulse leads to the increase in n_e . The lower the initial density, the
 340 greater the variation. But this increase leads to almost the same maximum value
 341 $n_{e,max} \approx 10^{24} \text{ m}^{-3}$.

342 The increase in electron density results from MPI and from the collisional ion-
 343 ization enhanced by the increase in electron temperature by IB. This increase is
 344 obtained from the neutrals, ions and electrons already present in the plasma volume
 345 irradiated by the second laser pulse (cf. figure 2). As a result, the obtainable maxi-
 346 mum electron density is a function of the temperature reached after the second pulse
 347 absorption period and of the reservoir of particles likely to be ionized. At the end
 348 of this ionization phase, the maximum $\text{Al}^{2+}/\text{Al}^+$ density ratio is given by the Saha
 349 equilibrium equation [40]

$$\frac{[\text{Al}^{2+}]}{[\text{Al}^+]} = \frac{1}{n_e} \frac{2 Z_{\text{Al}^{2+}}}{Z_{\text{Al}^+}} \left(\frac{2\pi m_e k_B T}{h^2} \right)^{3/2} e^{-\frac{E_i^{\text{Al}^+} - \Delta E}{k_B T}} \quad (3)$$

350 where Z_{Al^+} and $Z_{\text{Al}^{2+}}$ are the partition function of Al^+ and Al^{2+} , m_e the electron
 351 mass, k_B the Boltzmann constant, h the Planck constant, $E_i^{\text{Al}^+}$ the ionization poten-
 352 tial of Al^+ and ΔE the Al^+ ionization potential lowering. Using the NIST database,
 353 the previous ratio has been calculated for $n_e = 10^{23} \text{ m}^{-3}$ and 10^{24} m^{-3} as a function
 354 of T . The results are plotted on figure 7. The influence of the ionization potential
 355 lowering is weak because n_e is rather weak despite the values of T . Mainly, the
 356 values increase when T increases owing to the influence of the exponential term in
 357 equation (3). The curve corresponding to the case $n_e = 10^{23} \text{ m}^{-3}$ is found under
 358 the curve corresponding to the case $n_e = 10^{24} \text{ m}^{-3}$ because of the influence of the
 359 partition functions. The ionization potential of Al^+ is significant with a value of
 360 18.829 eV. This prevents this ratio to be higher than 10^{-2} at 100 000 K.

361 In a configuration similar to ours but with the use of optical fibers preventing
 362 any spatial characterization, Sobral *et al.* [41] have measured a weak increase in the
 363 temperature of around 1 500 K with $\bar{E}_2 = 100$ mJ, $f_2 = 150$ mm and $\lambda_2 = 1064$ nm
 364 when $(H, \Delta t_{1-2}) = (1$ mm, 500 ns). The temperature of the plasma where its
 365 radiation is maximum is then increased from around 15 500 K to 17 000 K. Since
 366 the (ionic) IB efficiency evolves as λ_2^3 [42], the effect on the plasma temperature of
 367 the laser absorption is necessary more significant than in our case. We can conclude
 368 that the temperature is too weak as demonstrated by figure 7 to induce a noticeable
 369 production of Al^{2+} ions. Thus the main ions are Al^+ . Once all the Al atoms present in
 370 the volume irradiated by the second laser pulse are ionized, it is therefore impossible
 371 to further produce ions: the electron density n_e then reaches a saturation state.

372 From this point of view, it is valuable to further analyze the results of Sobral *et al.*
 373 *al.* They have exploited the broadening of the H_α line to derive the n_e values. Such a
 374 determination systematically leads to a weak overestimation of the derived electron
 375 density [43]. They have observed an increase in n_e from $9 \times 10^{23} \text{ m}^{-3}$ to $1.4 \times 10^{24} \text{ m}^{-3}$
 376 due to the second laser pulse. Considering a reduction of n_e required by the use of
 377 the H_α line, the values are therefore in very good agreement with ours. Even if \bar{E}_2
 378 is much higher than in our conditions, the increase in n_e and T_e are therefore close
 379 to our own results. This saturation effect is also observed.

380 All these results have important consequences. Indeed, we can conclude that the
 381 density of Al^{2+} remains negligible with respect to the one of Al^+ for a wide range
 382 of the second laser pulse energy. Since nitrogen is in much lower density within the
 383 plasma because its presence results from scattering elementary processes across the
 384 plasma boundary, its influence on the ionization state can be assumed as negligible.
 385 Anyway, this conclusion can be also deduced about nitrogen since the N^+ ionization
 386 potential is 29.601 eV. As a result, the absorption of the second pulse completes the

387 ionization of the plasma by forming only single charged ions. Due to the available
 388 neutrals and single charged ions already present in the plasma volume irradiated
 389 by the second pulse, the final ionization degree is then close to unity and cannot
 390 correspond to an electron density higher than $n_{e,max} \approx 10^{24} \text{ m}^{-3}$ in the present case.

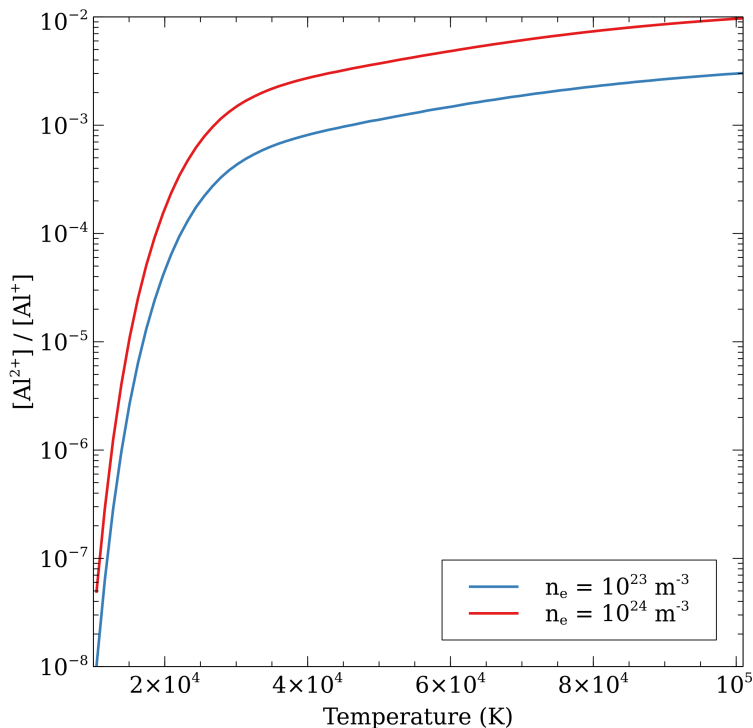


Figure 7: $[Al^{2+}]/[Al^+]$ density ratio as a function of temperature T at equilibrium for $n_e = 10^{23} \text{ m}^{-3}$ (in blue) and for $n_e = 10^{24} \text{ m}^{-3}$ (in red).

391 3.4. Spectral absorptivity measurements

392 In order to further characterize the plasma absorption, the energy of the second
 393 laser pulse is measured after the plasma crossing in the experimental conditions sum-
 394 marized in table 1. The results are plotted on figure 8. Between two experiments j
 395 each corresponding to $N^j \sim 2350$ pulses, the first laser pulse is not operated and the

396 aluminum plasma is therefore not produced. Then the second laser pulse propagates
 397 in ambient air without a significant breakdown formation. The measurement pro-
 398 vided by the energy meter then leads to verify that the second laser source delivers
 399 pulses with almost steady mean energy.

400 The energy delivered by the second laser source is fluctuating (cf. section 2.1).
 401 The plasma absorption corresponds to nonlinear phenomena. As a result, the trans-
 402 mitted energy is fluctuating differently. Thus three mean values have been deter-
 403 mined. The mean transmitted energy is denoted as $\bar{E}_{2,t}^j$. This value is indicated by
 404 a red segment on figure 8. Since the scattering of the laser light by the plasma is
 405 negligible in terms of energy [44], the mean spectral absorptivity $\bar{\alpha}_j = 1 - \frac{\bar{E}_{2,t}^j}{\bar{E}_2^j}$ is
 406 deduced where \bar{E}_2^j is the mean value between before and after the experiment no j .
 407 Sometimes, other mean values rise because a great number of pulses correspond to
 408 a transmitted energy departing significantly from $\bar{E}_{2,t}^j$. Two additional mean values
 409 are then defined. The second mean value $\bar{E}_{2,t,max}^j$ is calculated from the pulses whose
 410 energy transmission is higher than $\bar{E}_{2,t}^j$. This corresponds to a minimum mean spec-
 411 tral absorptivity $\bar{\alpha}_{j,min} = 1 - \frac{\bar{E}_{2,t,max}^j}{\bar{E}_2^j}$ (blue segments in figure 8). The third mean
 412 value $\bar{E}_{2,t,min}^j$ is calculated from the pulses whose energy transmission is lower than
 413 $\bar{E}_{2,t}^j$. This corresponds to a maximum mean spectral absorptivity $\bar{\alpha}_{j,max} = 1 - \frac{\bar{E}_{2,t,min}^j}{\bar{E}_2^j}$
 414 (green segments in figure 8).

415 We can observe that the pulse energy is significantly absorbed by the plasma.
 416 But the second pulse is never totally absorbed. Indeed, the lowest mean energy
 417 measured for the emerging pulse is 7.33 mJ (experiment no 1) for an incident value
 418 of $\bar{E}_2 = 15.49$ mJ. This corresponds to a mean spectral absorptivity of $\bar{\alpha}_1 = 0.53$.
 419 Since the density of the plasma decreases with time, the spectral absorptivity is lower
 420 when Δt_{1-2} is increased. Indeed, we see that for experiments 2 and 3, the mean value
 421 of the transmitted energy is higher, therefore the spectral absorptivity is lower. The

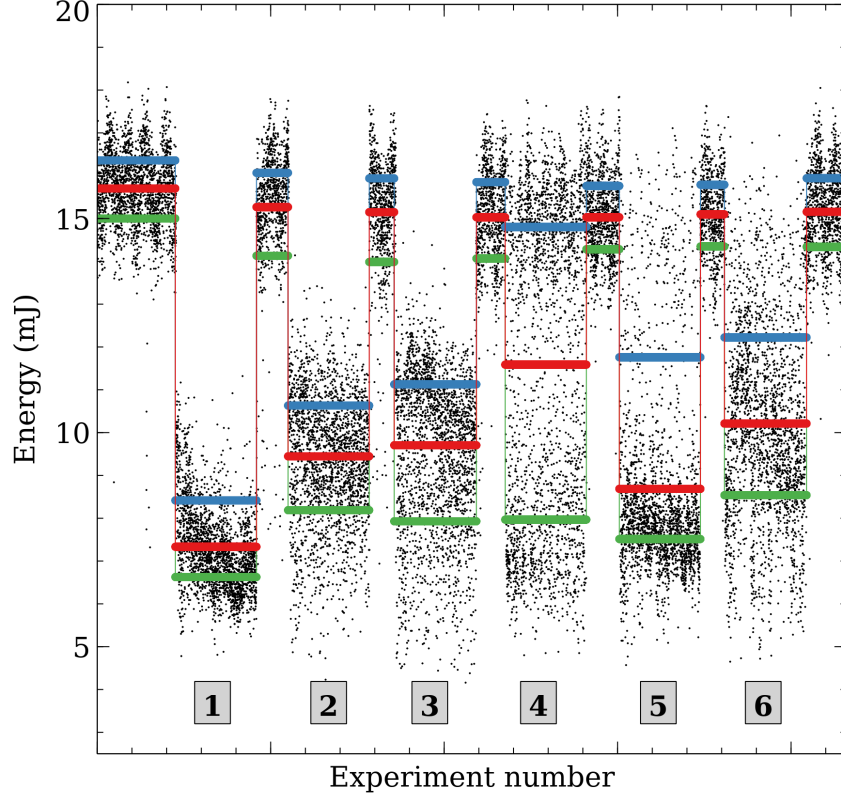


Figure 8: Energy of the second laser pulse emerging from the aluminum plasma region. Each black point corresponds to a laser pulse. The nominal energy is $\bar{E}_2 = 15.2$ mJ (cf. section 2.1) and is satisfactorily recovered between experiments. The transmitted energy is measured for each experimental condition listed in table 1. The mean values indicated by large segments have been obtained over ~ 2350 pulses. The blue segments correspond to mean values excluding pulses whose transmitted energy is lower than the red average and the green segments correspond to the complementary case.

422 mean spectral absorptivities are listed in table 3.

Experiment no j	1	2	3	4	5	6
\bar{E}_2^j (mJ)	15.49	15.21	15.09	15.03	15.06	15.13
$\bar{E}_{2,max}^j$ (mJ)	16.21	16.00	15.89	15.81	15.78	15.87
$\bar{E}_{2,min}^j$ (mJ)	14.57	14.06	14.03	14.17	14.32	14.34
$\bar{E}_{2,t}^j$ (mJ)	7.33	9.45	9.71	11.59	8.69	10.21
$\bar{E}_{2,t,max}^j$ (mJ)	8.42	10.63	11.13	14.80	11.76	12.22
$\bar{E}_{2,t,min}^j$ (mJ)	6.63	8.19	7.93	7.97	7.51	8.54
N^j	2340	2340	2370	2340	2340	2370
N_{min}^j	920	1210	1320	1240	650	1080
N_{max}^j	1420	1130	1050	1100	1690	1290
$\bar{\alpha}_j$	0.53	0.38	0.36	0.23	0.42	0.33
$\bar{\alpha}_{j,min}$	0.46	0.30	0.26	0.02	0.22	0.19
$\bar{\alpha}_{j,max}$	0.57	0.46	0.48	0.47	0.50	0.44

Table 3: Mean energy incident to the plasma (\bar{E}_2^j , $\bar{E}_{2,min}^j$, $\bar{E}_{2,max}^j$), mean energy transmitted by the plasma ($\bar{E}_{2,t}^j$, $\bar{E}_{2,t,min}^j$, $\bar{E}_{2,t,max}^j$), number of pulses (N^j , N_{min}^j , N_{max}^j) and resulting spectral absorptivity ($\bar{\alpha}_j$, $\bar{\alpha}_{j,min}$, $\bar{\alpha}_{j,max}$) for the experiments of figure 8 performed in the conditions of table 1.

423 Experiments 4, 5 and 6 have been performed at a higher distance from the sample
424 surface. Since an aluminum plasma produced by a picosecond laser pulse seems to
425 be characterized by gradients as weak as in the case of a nanosecond laser-induced
426 plasma [45], we should observe more or less the same experimental results as those
427 observed for experiments 1, 2 and 3. Obviously, figure 8 shows that it is not the
428 case except between experiments 3 and 6 despite the fluctuations. Indeed, we see in
429 table 3 that the spectral absorptivity of the latter experiments is close to each other.

430 In addition, the fluctuations of experiments 4, 5 and 6 are much more significant
 431 than for experiments 1, 2 and 3. Besides the mean transmitted energy $\bar{E}_{2,t,max}^j$ for
 432 experiment 4 is almost equal to the mean transmitted energy \bar{E}_2^j without plasma, and
 433 the mean transmitted energy $\bar{E}_{2,t,min}^j$ for experiment 4 is close to the mean energy
 434 $\bar{E}_{2,t}^j$ for experiment 1. These features are the direct result of the hypersonic expansion
 435 of the plasma put in evidence in section 3.2. Indeed, the laser pulse mainly interacts
 436 with the ambient atmosphere before the arrival of the plasma produced on the sample
 437 surface. In section 3.2, we have shown that the characteristic plasma expansion time
 438 over $z = 1.36$ mm is $t' = 320$ ns. This time is higher than $\Delta t_{1-2} = 150$ ns. The
 439 plasma should not have reached the position of the second laser pulse. Since the
 440 second laser fluence F_2 is lower than but very close to the air breakdown threshold,
 441 an air plasma is rarely produced. The transmitted energy $\bar{E}_{2,t,max}^4 = 14.80$ mJ is
 442 therefore close to the one observed without aluminum plasma $\bar{E}_2^4 = 15.03$ mJ. Over
 443 the $N^4 = 2340$ pulses corresponding to experiment 4, $N_{min}^4 = 1240$ pulses (i.e.
 444 53 %) correspond to this situation and lead to $\bar{E}_{2,t,max}^4 = 14.80$ mJ. Nevertheless,
 445 the second laser pulse often interacts with the aluminum plasma because of the
 446 fluctuations. This is the case for the other $N_{max}^4 = 1100$ pulses (47 % of the total
 447 number of pulses) leading to a significant absorption. The resulting mean spectral
 448 absorptivity $\bar{\alpha}_{4,max} = 0.47$ is then close to the value $\bar{\alpha}_1 = 0.53$. The existence of
 449 these two complementary regimes is particularly obvious on figure 8. Indeed, two
 450 clouds of points are clearly observed around the mean values $\bar{E}_{2,t,min}^4$ and $\bar{E}_{2,t,max}^4$.

451 In the case of experiment 5, Δt_{1-2} is again lower than t' . Therefore the previous
 452 behaviour is still observed but in a reduced way. $N_{max}^5 = 1690$ pulses lead to the mean
 453 spectral absorptivity $\bar{\alpha}_{5,max} = 0.50$ and $N_{min}^5 = 650$ pulses lead to $\bar{\alpha}_{5,min} = 0.22$.
 454 For experiment 6, $\Delta t_{1-2} > t'$ and $N_{max}^6 = 1290$ pulses lead to the mean spectral
 455 absorptivity $\bar{\alpha}_{6,max} = 0.44$ and $N_{min}^6 = 1080$ pulses lead to $\bar{\alpha}_{6,min} = 0.19$.

456 In section 3.3, we have mentioned that the absorption of the second laser pulse by
457 the aluminum plasma clearly illustrates a typical behaviour of saturation. Indeed,
458 in any case the electron density reached by the plasma after absorption does not
459 exceed around 10^{24} m^{-3} . This saturation-like situation is confirmed by the spectral
460 absorptivity listed in table 3. Indeed, the reported values are never close to unity.
461 The highest value for $\bar{\alpha}_j$ is 0.53. This means that the energy absorption is limited
462 despite the high value energy available in the second laser pulse. Around 50 % of
463 this energy crosses the plasma and does not interact with its particles.

464 **4. Conclusion**

465 A thorough study of the absorption of a nanosecond laser pulse at 532 nm by
466 a picosecond laser-induced preformed aluminum plasma has been performed using
467 a specific experimental device. The experiments have been carried out in air at at-
468 mospheric pressure. The absorption has been studied according to two parameters:
469 the time delay between the plasma production and the time of absorption, and the
470 location of the absorption with respect to the aluminum sample. The values of the
471 electron density have been derived from the study of the Stark broadening of reliable
472 ionic and atomic lines. We have observed that the absorption leads to a similar
473 electron density of around 10^{24} m^{-3} for any starting value of the electron density.
474 Absorptivity measurements have been also performed to quantitatively estimate the
475 relationship between the increase in the electron density and the energy actually
476 absorbed by the plasma. These measurements have put in evidence the strong fluc-
477 tuations of this absorption due to those of the plasma propagation. The energy
478 absorbed is at best of around 50 % of the incident energy. These results accredit the
479 idea of a saturation of the absorption process due to the saturation of the production

480 of ions from atoms and ions already present in the plasma volume irradiated by the
481 second pulse.

482 To verify this interpretation, similar measurements will be shortly performed at
483 wavelengths 355 and 1064 nm in exactly the same configuration. Besides, these ex-
484 periments will enable to verify that the energy is predominantly absorbed by inverse
485 Bremsstrahlung. These measurements will be analyzed in the light of the modelling
486 of the whole absorption process.

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501 **Declaration of interest**

502 None

503 **Credit author statement**

504 Aurélien Favre: investigation, writing

505 Vincent Morel: supervision

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507 Gilles Godard: resources

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509 Christian Grisolia: resources

510

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GRAPHICAL ABSTRACT

Figure 1: Schematic view of the platform PLEIADES.

The first laser source on the left produces infrared photons along the red axis. After focus, the plasma in yellow is generated on the Al sample placed on the central (xyz)-stage holder. The second laser source in the background produces visible photons along the green axis. They are focused in the plasma whose spectroscopic analysis is performed by the spectrometer on the purple axis.

Figure 2: Relative sensor/plasma arrangement in the case where the second optical axis corresponds to the location of ROI c ($H = 1$ mm). When $H = 0$ mm, the distance between the upper border of the ROI a and the second optical axis is l_{2-a} .

Figure 3: ROIs and associated spectra for $\lambda \sim 396$ nm at the acquisition time 40, 380 and 480 ns after the production of the plasma by the first pulse at $t = 0$. The plasma is reheated by the second pulse in the conditions $(H; \Delta t_{1-2}) = (1$ mm, 350 ns) of the experiment no 6. The red frames and spectra correspond to the plasma close to the sample (ROI a). The yellow frames and spectra correspond to the plasma reheated by the second pulse (ROI c).

Figure 4: Time evolution of the spectral radiance at the wavelength of the Al I and N II transitions of figure 3 obtained without second pulse close to the sample's surface (ROI a, continuous lines) and ~ 1 mm above (ROI c, dashed lines).

Figure 5: Time evolution of the spectral radiance at the wavelength of the Al I and N II transitions obtained close to the sample's surface (ROI a, continuous lines) and over the ROI corresponding to the position of the second pulse (dashed lines, ROI b for $H = 0$ mm and ROI c for $H = 1$ mm) for the different values of Δt_{1-2} (colors similar to figure 4).

Figure 6: Time evolution of the electron density n_e (trends in colored dashed lines) and comparison with the evolution obtained without the second pulse (trends in thin dashed black lines).

Figure 7: $[Al^{2+}]/[Al^+]$ density ratio as a function of temperature T at equilibrium for $n_e = 10^{23} \text{ m}^{-3}$ (in blue) and for $n_e = 10^{24} \text{ m}^{-3}$ (in red).

Figure 8: Energy of the second laser pulse emerging from the aluminum plasma region. Each black point corresponds to a laser pulse. The nominal energy is $E_2 = 15.2$ mJ (cf. section 2.1) and is satisfactorily recovered between experiments. The transmitted energy is measured for each experimental condition listed in table 1. The mean values indicated by large segments have been obtained over ~ 2350 pulses. The blue segments correspond to mean values excluding pulses whose transmitted energy is lower than the red average and the green segments correspond to the complementary case.