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## Investigation of laser-induced plasma at varying pressure and laser focusing



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#### ABSTRACT

Expansion dynamics of laser-induced plasma is studied for different focal positions of the ablation laser in the pressure range  $10^{-2} - 10^5$  Pa of the ambient air. The experimental results indicate that both the parameters significantly affect the plasma size, shape, intensity, reproducibility, and distance from the target surface. At pressures above 10 Pa, the plasma plume is confined by the ambient gas; the plumes are more compact and travel shorter distances from the target as compared to the analogous plume characteristics at pressures below 10 Pa. The pulse-to-pulse reproducibility of the integral emission intensity of the plasma is also different for different focal positions and pressures. It is found that the focal positions -1 cm and -2 cm below the target surface yield the most reproducible and intense emission signals as measured at the 600 ns delay time with the 100 ns gate. The information obtained can be of importance for pulsel alser deposition, laser welding, and analytical spectroscopy at reduced pressures. In general, a correct choice of the focal position and pressure of an ambient gas is very important for obtaining the strongest plasma emission, good reproducibility, and desired plasma plume shape.

#### 1. Introduction

Laser-induced plasma technique is used in various fields, such as laser ablation inductively coupled plasma mass spectroscopy (LA-ICP-MS) [1], pulse laser deposition (PLD) [2], and laser-induced breakdown spectroscopy (LIBS) [3,4]. The improvement of the signal intensity and plasma reproducibility are the important research topics for laser-induced plasma applications. Due to the complexity of laser ablation mechanisms, such as the plasma-target interaction, laser-plasma shielding, and plasma plume expansion, the whole physical picture of the laser-induced plasma is not yet conclusively understood.

In the nanosecond laser ablation, coupling the laser energy to the target leads to heating, melting, and evaporation of the target material. These processes are in turn affected by the plasma generated above the target surface. The absorption of laser energy depends on the target material and on ambient conditions [5–7]. In the presence of an ambient gas, the plasma expansion dynamics is different from that in vacuum because of the confinement effect. The confinement effect results in denser plasma and increase of frequency of collisions between ablated and ambient gas species [8]. The denser plasma partly shields the laser radiation and thus reduces the ablated target mass.

The initial state of the plasma is crucial for determining its subsequent evolution. First, changing the distance between the lens and target surface results in different laser fluence (J/cm<sup>2</sup>) and greatly affects the plasma formation [9]. For example, this parameter is important for the efficient production of particles by pulsed laser ablation in liquids (PLAL) [10]. Second, the pressure of an ambient gas also greatly affects the plasma formation and expansion dynamics [11]. This paper presents an investigation of the evolution of the laser-induced plasma plume at different pressures and laser focusing positions. We show that the plasma intensity, pulse-to-pulse reproducibility, plume length, and plume shape critically depend on these two parameters. This is the important information for many laser-induced plasma applications.

#### 2. Experimental setup

Fig. 1(a) shows the experimental setup. A Q-switched Nd:YAG laser (Brilliant Eazy, Quantel, France) operated at a fundamental wavelength of 1064 nm, 10 mJ pulse energy, and 5 ns pulse width is used to ablate the target. The laser beam is guided inside a vacuum chamber through a quartz window and focused on a target using a 15 cm focal length plano-convex lens. A mechanical (Ecodry M, Leybold Vakuum, Germany) and turbo molecular (Turbovac 361 Leybold Vakuum, Germany) pumps are used to evacuate the chamber. A hot ion combi vacuum gauge (Ionvac ITR90, Leybold Vakuum, Germany) continuously

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**Fig. 1.** (a) Experimental setup; (b) Laser focal point distances from the target surface. The arrows indicate the direction of motion of the focusing lens; (c) Ablation craters at different focal positions at 10<sup>5</sup> Pa photographed at magnifications 3.2x and 20x of the optical microscope.

measures pressure inside the chamber. In our experiment, the pressure is gradually varied from  $10^{-2}$  Pa to  $10^{5}$  Pa at a one-order-of-magnitude step. A copper foil of a technical grade (thickness 0.1 mm) is used as an ablation target. The foil is polished with a 800 fineness abrasive paper (grain size  $\sim 12.6 \,\mu$ m). The surface roughness is measured with a white light interferometer and is found to be  $\sim 1 \,\mu m$ . This tiny roughness ensures the high reproducibility of the plasma emission signal and independence of this signal of the ablation position on the target surface. The target is placed on a XYZ-stepping motor stage, which is mounted inside the vacuum chamber. The moving stage provides a fresh ablation spot for each laser pulse and eliminates the possible confining effect due to previously ablated craters. The plasma images are captured by an intensified CCD (ICCD) camera (DH734, Andor, UK) using a 15 cm focal length, 2.54 cm diameter objective lens set at a 90° angle with respect to the laser beam. No filter is used for the selection of a specific emission range. The images are processed and displayed by a computer. The magnification of the optical system is 0.65. In order to obtain the exact relationship between the image and physical dimensions of the plasma plume, a ruler is placed at the position of the plasma and projected onto the ICCD. This yields 50 pixels on a camera chip per 1 mm of the physical length. The ICCD is set to a 100 ns gate width and zero gain. A delay generator (DG645, SRS, USA) controls timing of the laser and ICCD.

Fig. 1(b) shows the four different focusing positions with the focal points located at 0, -1, -2, and -3 cm relative to the target surface. It is verified that no air breakdown is created at any focal position for all tested pressures. The focal position is controlled by moving the focusing lens along the direction of the laser beam thus ensuring the same spatial position of the laser-induced plasma with respect to the collection optics. The diameters of the ablated craters are 0.1, 0.3, 0.5 and 0.7 mm at the pressure of  $10^5 \text{ Pa}$  as the focal point moves from 0 cmdown to -3 cm. The corresponding laser fluences are 127.4, 14.2, 5.1, and  $2.6 \text{ J/cm}^2$ . The diameters of the ablated craters corresponding to the four focal positions are shown in Fig. 1(c). They are measured with an optical microscope at 3.2x and 20x magnifications. A conical crater with a measurable depth of about  $2 \,\mu m$  is created only at the 0 cm focal position; at other focal positions and decreasing laser fluence, only shallow craters are formed whose depths are of order of surface roughness, i.e. 1 µm.

#### 3. Results and discussion

Fig. 2 shows the laser-induced plasma images obtained at the 600 ns delay time with the 100 ns gate in the pressure range  $10^{-2}$  -  $10^5$  Pa and

focal positions 0 - (-3) cm. One sees that the focal position significantly affects the plasma shape and intensity. The radiation intensity integrated over the entire plume, has its lowest value for the lowest fluence. This corresponds to the position of the laser focusing lens closest to the target surface and, thus, to the maximal spot size. This is position 3 with the focal point -3 cm below the target surface. At this position, the plasma is unstable, especially at pressures below 100 Pa as compared to other focal positions. The laser fluence here is  $2.6 \text{ J/cm}^2$  that is close to the breakdown threshold [12]. At focal positions -2, -1, and 0 cm the plasma exhibits different behavior.

When the pressure decreases from 10<sup>5</sup> Pa to 10 Pa both the plasma size and distance from the target increase. In the interval 10 Pa - 1 Pa the plume size decreases. At pressures below 1 Pa, both the plasma size and plasma radiation intensity increase again. This can be explained by different mechanisms which drive the plasma expansion. With ambient pressure exceeding 10 Pa, the plasma expansion becomes confined by the residual ambient air. Below 10 Pa, the confinement is negligible, the plasma size becomes larger and the distance between the target and the plasma plume center of mass increases. With further decrease of pressure down to 1 Pa, the dominating factors driving the plume expansion become the laser-target and laser-plasma interactions at the onset of laser ablation. Harilal et al. [13,14] showed that both the plasma density and plasma temperature decrease insignificantly when the ambient pressure decreases from 1 Pa to  $10^{-2}$  Pa. This indicates that less laser energy is absorbed by the plasma and more by the target. Therefore, more target material is ablated leading to the larger plasma size at pressures  $10^{-2}$ –1 Pa. Noteworthy, the plasma generated at positions -1 and -2 cm below the target surface exhibits a tear-like shape. The formation of this shape can be explained as follows. The initial formation of the plasma plume is strongly affected by the absorption of the leading edge of the laser pulse by the target. At this stage, the expanding plume, consisting of target material, does not experience too many collisions with the molecules of the rarefied ambient gas. On the contrary, the tailing part of the laser pulse is absorbed partly by the plasma and partly by the target. The material expelled from the target at this stage expands into the dense environment of the initial plasma plume and thus experiences a large number of collisions. This causes the tailing part of the plume to be confined and to have a much lower expansion speed as compared to the leading part of the plume. Therefore, the plume acquires a tear-like shape.

The images in Fig. 2 show the plasma plume shapes at all focal positions and pressures. For further analysis, the background noise of the ICCD is subtracted from all images. The spatially-integrated plume intensity at the position -2 cm and each pressure is taken as the

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