ISSN 1330–0008
CODEN FIZAE4

TIME–INTEGRATED PHOTOGRAPHIC STUDY OF LASER ABLATION AT LARGE IMPACT ANGLES

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Received 3 April 1998; revised manuscript received 5 July 1998
Accepted 13 July 1998

The shape of the plasma cloud generated by pulsed laser ablation of solid targets when the laser beam impacts onto the target surface at large angles is investigated. Time–integrated photographs of the expanding plasma clouds clearly show that plasma is produced at large impact angles too, and that it always expands perpendicularly to the target surface. The cone angles of the expanding plasma clouds increase with the impact angle, indicating that the kinetic energy of the plasma particles is reduced at large angles. The intensity of the plasma emission drops at angles larger than about 60° to the surface normal, indicating that the amount of plasma produced at such large angles is smaller than during ablation at small angles. The apparent discrepancy in the observed plasma formation threshold and the on–surface energy density is explained by the mechanism of plasma initiation on localized surface defects whose surfaces are placed at favourable angles to the incoming laser radiation.

PACS numbers: 52.50.Jm, 61.80.Ba UDC 533.9

Keywords: plasma production and heating by laser beams

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1. Introduction

In most ablation experiments described in the literature the laser beam impacts perpendicularly onto the target surface. In applications where material ejected from the target surface itself is further used, the laser beam is shifted to a side of the target normal to separate it from the expanding plasma cloud. The plasma cloud can then be freely used for deposition or for some other purpose. Even in this case, the impact angles are kept as close to the normal as possible and the plasma cloud behavior is similar to the case of perpendicular impact of the laser beam onto the target surface. Regardless of the impact angle of the laser beam, the plasma cloud always expands perpendicularly to the mean plane of the target surface. This is verified for angles of up to about 45° from the surface normal [1]. The simple experiments reported in this article show that the plasma is formed right up to the grazing incidence of the laser beam onto the target surface and that the plasma cloud always expands perpendicularly to the target surface.

Initially one would assume that the laser beam striking the target at a large angle to the surface normal would hardly be able to ablate it due to the increased coefficient of reflection at large angles and due to the reduced on–surface energy density. This conclusion stems from the idealized picture of target surface being ideally flat. In reality, however, even highly polished surfaces show many surface irregularities and surface defects at submicron scales. Surfaces produced by the usual machining techniques are highly irregular at such scales. Further, at low energy densities plasma is initiated at local surface defects or irregularities either by favourable laser beam reflections that enhance local electric fields or due to reduced heat flow into the bulk target material that prevents cooling of the surface defect with the end result that it is overheated and vaporized [2,3]. Even when the laser beam impacts at the target surface at a grazing angle to the mean surface plane, some favourably placed surface irregularities can still be found on the target surface, although at a greatly reduced density, so a smaller amount of plasma will still be produced.

Several applications of laser ablation may benefit from this ablation geometry. One of them is determination of chemical composition of target material from spectral emission of the laser–produced plasma. In this application, a small amount of the target material is vaporized and ionized by the laser radiation. The optical emission from the resulting plasma is then used to determine the chemical composition of the target material [4–6]. The method is inherently simple and attractive due to its speed and its capability of analyzing microsamples, but is still far from trouble-free. One of the difficulties associated with it is that the high density of the produced plasma often results in optically thick spectral lines which complicate the interpretation of the observed spectra. The less dense plasma produced by the ablation at large impact angles would in this case be beneficial.

Another application of interest are XUV and X-ray lasers where laser–produced plasmas serve as laser medium. Such short–wavelength lasers are of great interest as they are needed in many important applications ranging from sub–micron lithography [7] to the real–time holography and microscopy of biological specimens [8]. The lasing of the plasma produced by the ablation of a capillary wall and further excited by a second laser pulse was demonstrated recently [9], thus stressing the importance of further studies of laser ablation at large or even grazing angles (as is the case when the capillary wall is ablated from inside).

2. Experimental setup

A nitrogen laser [10,11] was used as a source of laser radiation. The laser beam was focused onto the target surface with a quartz planoconvex lens (160 mm focal length). The on–target beam cross–section and the temporal pulse profile were studied in detail before [12]. The main parameters of the laser beam are summarized in Table 1. A 2 mm thick sintered boron–nitride (HDBN) plate was used as a target. The target was placed inside the vacuum chamber and all experiments were carried out in vacuum at a base pressure of about 0.1 Pa. The plasma cloud was observed side–on with a photographic camera. The low intensity of the plasma emission required use of a high speed film (ILFORD HP–5). The film was developed for maximal contrast, and positives were printed onto very hard photographic paper. Such a technique produces a greatly reduced gray scale with black–white transition corresponding to an isophote. The isophote shape was extracted from the positives with the help of a digitizing table.

TABLE 1. On–target parameters of the laser beam.

wavelength (nm)	337.1
pulse energy (mJ)	7
temporal pulse profile	approx. gaussian
spatial pulse profile	irregular
pulse FWHM (ns)	6
max. energy density (J/cm^2)	0.7
max. power density (MW/cm^2)	90
burn mark (mm$\times$mm)	0.3x1

3. Discussion

The maximal on–target energy density was 0.7 J/cm^2 in the case of perpendicular impact. The plasma formation threshold was found to be about 0.3 J/cm^2 in this case. The expanding plasma cloud generated with the laser beam impacting at various angles to the surface normal are shown in Fig. 1. Note a characteristic large forward directivity of the expanding plasma resulting in a highly elongated isophote shapes in the direction of the surface normal. The forward directivity is a distinct characteristics of the laser produced plasmas, and it increases with the increase of the on–target energy density [13,14]. In case of no preferential direction of expansion, the angular distribution of the evaporated particles is given by the $\cos\vartheta$ function, where ϑ is the angle between the direction of the flight of the particle and the target surface normal. In the case of preferential forward directivity, the particle distribution is given by the $\cos^n\vartheta$ function, with $n > 1$ [14]. By measuring the angles between the tangents to the outer plasma envelope and the surface normal, the exponent n can be judged. The results of this procedure are summarized in Table 2.

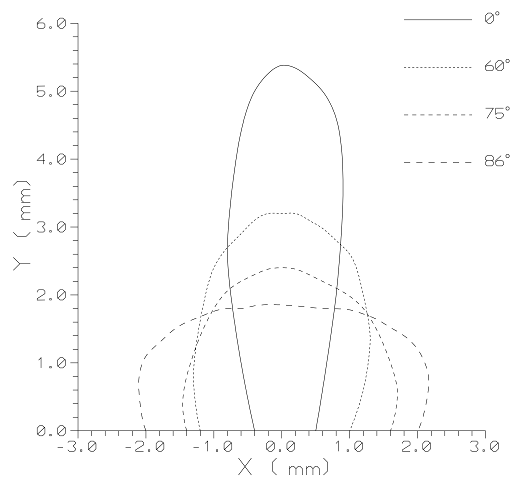

Fig. 1. Plasma cloud shape as a function of the impact angle of the laser beam to the target normal. The target surface corresponds to the Y=0 plane (X-axis on the graph).

TABLE 2. Particle directivities inferred from the photographs of the plasma clouds. The exponent n corresponds to the $\cos^n$ yield directivities of Ref. 14.

impact angle	cone angle	n
0°	22° ± 3°	25±5
60°	34° ± 5°	10±3
75°	70° ± 10°	2±0.5
86°	90° ± 20°	≈ 1
90°	90° ± 20°	≈ 1

Up to the impact angles of about 60°, the appearance of the plasma cloud remains about the same. It is only slightly widened in the direction parallel to the target surface as the focal spot is elongated in this direction due to the oblique impact of the laser beam onto the target surface. At larger impact angles the plasma brightness decreases and the size of the observable cloud becomes smaller. The plasma is still detectable at 86°, although it is very weak and shows no directivity anymore.

In the case when the impact angle was 86° to the target normal, the maximal energy density projected to the surface plane was reduced to 0.05 J/cm^2, far under the plasma formation threshold. Yet, the plasma is clearly observed in the second case, too. This is already explained by the fact that plasma is initiated at surface defects whose surfaces are placed at favourable angles to the incoming laser beam.

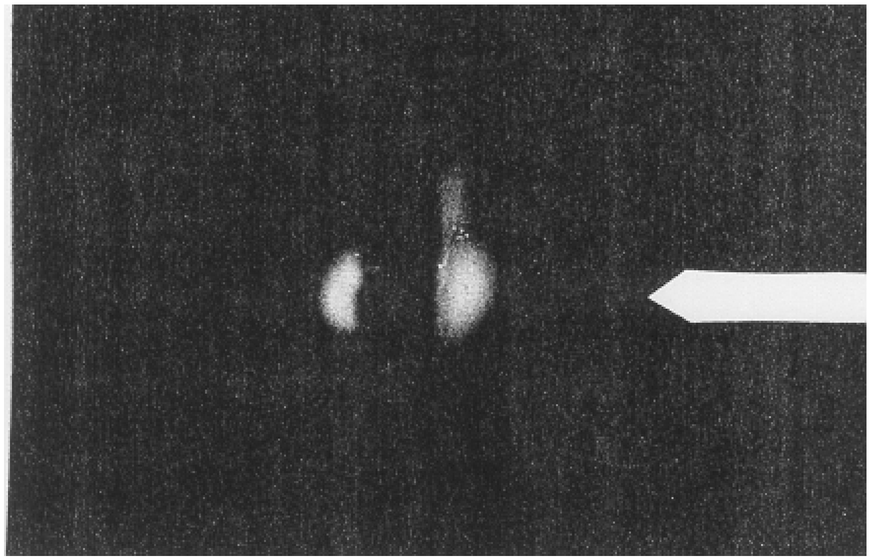

Fig. 2. Two plasma clouds emanate out of the ends of a 0.5 mm *diameter capillary bored through a 3 mm thick copper target. The laser beam direction is indicated by the white arrow.*

Last, but not least, it is interesting to note that plasma is formed even when the axis of the laser beam is parallel to the surface. To prove this, a simple experiment was performed. The laser beam was directed through a small hole bored through the target in such a way that the laser beam axis was parallel to the hole axis. A cooper target was used in this experiment as the intense blue–green emission of Cu I, and the absence of fluorescence of the target material, makes cooper plasmas much easier to detect. Two plasma clouds are observed in such a case (Fig. 2). One expands from the front hole opening towards the laser, and the other from the back hole opening in the opposite direction. Both clouds are relatively small and show no preferable direction of expansion. About an order of magnitude larger number of laser pulses is needed to register the plasma images with the similar density as in the case of small impact angles, which points to much lover densities and temperatures of plasmas generated this way.

4. Conclusion

The analysis of time–integrated photographs of the expanding plasma clouds clearly show that plasma is produced at all impact angles, and that it always expands perpendicularly to the target surface. At constant laser energy density the forward directivity of plasma particles decreases with the increase of the impact angle, indicating that their kinetic energy is reduced. The time–integrated brightness of the plasma cloud rapidly decreases at angles larger than about 60° to the surface normal. From this follows that both the plasma temperature and the amount of plasma produced are smaller at large angles. This is consistent with expected decrease of density of favourably placed surface defects needed to initiate the plasma formation. The apparent discrepancy in the observed plasma formation threshold and the on–surface energy density is consistent with the mechanism of plasma initiation on localized surface defects whose surfaces are oriented at favourable angles to the incoming laser radiation.

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VREMENSKI INTEGRIRANO FOTOGRAFSKO PROUČAVANJE LASERSKE ABLACIJE PRI VELIKIM UPADNIM KUTOVIMA

Iz fotografija oblaka plazme proizvedenih laserskom ablacijom pri različitim upadnim kutovima, određena je ovisnost oblika plazmenog oblaka o upadnom kutu i pokazano je da se ablacija događa i kod vrlo velikih upadnih kutova.