*Romanian Reports in Physics, Vol. 60, No. 3, P. 789–796, 2008*

*Dedicated to Prof. Ioan-Iovitz Popescu's 75th Anniversary*

# EXPERIMENTAL INVESTIGATION OF THE NANOSECOND LASER ABLATION RATE OF ALUMINUM

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(Received January 30, 2008)

*Abstract.* The dependence of the ablation rate of aluminum of the laser spot diameter and laser fluency is analysed, in the visible and infrared nanosecond-pulses irradiation regimes. Experimental results indicate that, for a constant flux of energy and large laser spots, the ablation rate decays linearly with increasing laser spots diameter reaching maximum values of 5.5 μm/pulse for the visible radiation respectively, 0.35 μm/pulse in the case of infrared laser pulses. This indicates that the infrared radiation is much less efficiently coupled to the metallic sample than the visible radiation. By decreasing the diameter of visible laser-spot below a threshold value (≈ 0.6 mm) a nonlinear increase of the ablation rate are observed. The decrease of the spot diameter leads to an increase of laser fluence. For the values of the fluence in the low and medium regimes, increasing the fluence of the infrared and visible laser waves gives rise to an increase of the ablation rate inversely proportional with the square-root of the fluence. Further increase of the visible laser fluence above a threshold value (≈ 60 J/cm$^2$) leads to a marked quadratic increase of the ablation rate. The transition from the asymptotic to the quadratic increase of the ablation rate with increasing laser fluence, and from the linear to the nonlinear increase with decreasing laser–spot diameter, are related to the transition from a normal vaporization regime to a phase explosion regime.

*Key words:* ablation rate, nanosecond laser pulses, metal.

## 1. INTRODUCTION

Material removal from a sample surface under the action of short and high intensity laser pulses is called pulsed laser ablation (PLA). The efficiency of this process is described by the ablation rate, which gives the maximum layer thickness ablated during irradiation with a laser pulse. Understanding and controlling the ablation rate is essential for micro-patterning in electronics, optoelectronics and micromechanics [1–7].

The ablation rate is strongly influenced by the processing laser beam characteristics (laser spot diameter, laser fluence, laser wavelength, number of pulses and pulse duration), and by the optical and thermal properties of the processed material surface (optical absorption, surface reflectivity, thermal conductance, mass density) [1, 2, 5, 6, 8–12, 15–17]. Experimental investigation of the dependence of the ablation rate on the laser spot diameter in nanopulsed regime irradiation indicates that, in the case of semiconductors and dielectrics, there is a remarkable dependence of the ablation rate on the spot diameter [2, 9]. For spot diameters that are small relative to the plasma hydrodynamic length, there is a marked linear decrease of the ablation rate with increasing the spot diameter, whereas for spot diameters in the order of the plasma hydrodynamic length, increasing spots diameter leaves the ablation rate almost unchanged.

The laser fluence ($F$) and wavelength ($\lambda$) strongly influence the ablation rate of semiconductors and dielectrics, as well as optical and thermal proprieties of the material [2, 8, 10, 13, 14]. For values of the laser fluences higher than a wavelength-dependent threshold value, a non-linearly increasing of the ablation rate takes place for increasing laser fluences. The dependence of the ablation rate and of the threshold laser fluence on the laser wavelength arises from three interrelated phenomena: the decrease of the intrinsic absorption of semiconductors and dielectrics, the increase of the surface reflectivity and the enhancement of the absorptivity of the produced plasma with increasing the laser wavelength. The interplay of these three phenomena drives to a larger ablation rate and smaller threshold fluence for the visible than for infrared laser pulses.

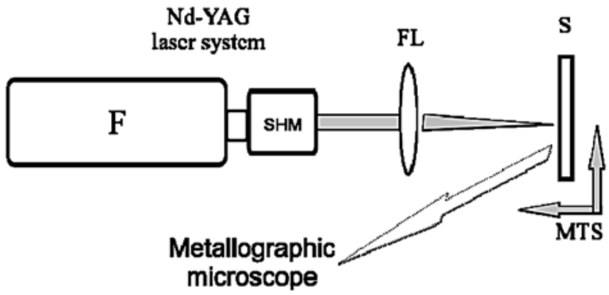
Here, we analyze experimentally the dependence of the ablation rate of metals on the laser spot diameter and on the laser fluence in the nanopulsed infrared and visible regimes. The results indicate that at constant energy flux and large spots, the ablation rate increase linearly with decreasing laser spots diameter. A significantly efficient ablation is obtained when using visible pulse than infrared pulses and for values of the laser spot smaller than a threshold value a marked nonlinear increase of the ablation rate are observed.

The decrease of the spot-diameter leads to an increase of the incident laser fluence and, for values of the laser fluence in the low and medium regime, gives an asymptotic increase of the ablation rate inversely proportional with the square root of the laser fluence. Increasing the laser fluence above a threshold value leads to a marked quadratic increase of the ablation rate with fluence, and a significant number of bright sparks consisting in particulates and melted material are observed within the plasma plume. The threshold fluence and the threshold laser-spot diameter are related to the transition from normal vaporization to phase explosion during pulsed laser ablation of aluminium.

## 2. EXPERIMENTS

The experimental setup is depicted in Fig. 1. The laser pulse from a Q-switched Nd-YAG laser, provided with a second harmonic module (SHM), is

Fig. 1 – Experimental setup: F – laser head generating the fundamental wave, SHM – second harmonic generator module, FL – focusing lens, MTS – mechanical translation stage.




focused on the aluminum target. The laser-pulse is characterized by a duration of 4.5 ns, a repetition rate of 10 Hz and energy values of 360 mJ/pulse for the fundamental ($\lambda$ = 1064 nm) and 180 mJ/pulse for the second harmonic ($\lambda$ = 532 nm), respectively. The aluminum sample, 1 mm tick, is fixed on a three-dimensional mechanical stage which enables the translation of the sample toward the laser system in the axial direction of the laser beam with micrometric resolution. We choose to displace the sample toward laser system to avoid the effect of air breakdown and the loss of the laser energy spending for heating the plasma plume ignited in front of the sample. Initially, the sample is placed in the lens focus with the laser beam at normal incidence and then is translated in steps of 0.5 mm and 0.2 mm in the case of the infrared and the visible radiation, respectively. Thus, the diameter of crater varies in the range of 1–3 mm in the case of infrared pulses and of 0.4–1.6 mm in the case of visible pulses.

The number of the incident laser pulses on the sample surface is chosen so that the ablation rate is maintained approximately constant during multi-pulses irradiation [11]. To obtain a crater sufficiently deep to allow measuring of the ablation rate with a relative error of maximum 5%, we used 200 infrared laser pulses, and 10 visible wavelength pulses, respectively. For a certain position of the sample relative to the focusing lens, and for each irradiation regime, we drilled 10 craters having an approximately cylindrical shape and characterized by a depth and a diameter. The diameter and the depth of the crater were measured using a metallographic microscope with micrometric resolution.

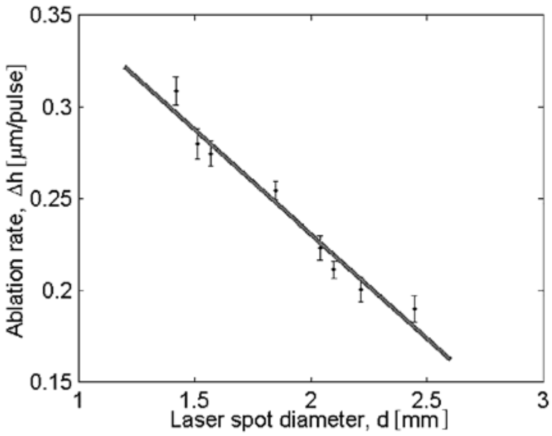
The ablation rate, the laser spot area, and the laser fluence were derived from the diameter and the depth of the crater, as follows. The mean ablation rate was calculated by dividing the crater depth by the number of laser pulses used for drilling the crater, and averaging over the number of craters with similar characteristics. The mean crater diameter was determined by measuring the diameter along two perpendicular directions and averaging over the number of similar craters. Because the crater diameter is at least one order of magnitude bigger than the thermal and optical lengths of the sample, the laser spot diameter could be approximated with the crater diameter. The ratio between the energy of the laser pulse and the laser spot area, which was taken as the area of the mean-diameter crater, gives the mean laser fluence of the laser pulses.

## 3. RESULTS AND DISCUSSION

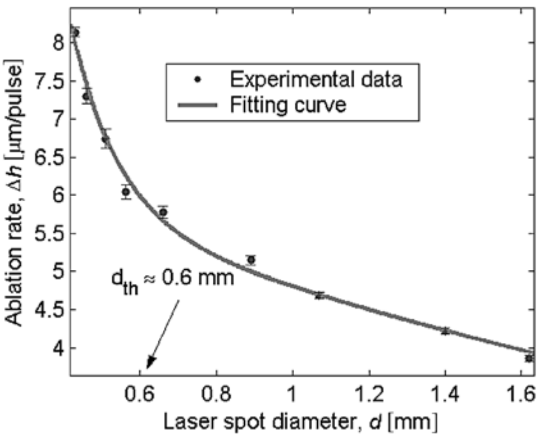
In Fig. 2 is depicted the dependence of the ablation rate of aluminum, $\Delta h$, on the laser spot diameter, $d$, for the infrared and visible laser beams. For a constant infrared laser flux of 72 MJ/s, the decrease of the laser-spot diameter from 3 to 1mm leads to a linear increase of the ablation rate from 0.15 μm/pulse to 3.35 μm/pulse (Fig. 2a). In a similar manner, at a constant visible laser flux of 36 MJ/s, decreasing the laser spot diameter from 1.6 to 0.7 mm gives an approximatey linear increase of the ablation rate from 3.5 μm/pulse to 5.5 μm/pulse (Fig. 2b). The fitting curves for the linear dependence of the ablation rate on the laser spot diameter are described by the equations

$$\Delta h = 0.45 - 0.11 \times 10^{-3}\, d \quad [\mu\text{m}] \tag{1}$$

for the IR pulses, and




a)




b)

Fig. 2 – Ablation rate of aluminum as a function of the laser spor diameter. The laser wavelength is 1064 nm in paneel (a), and 532 nm in panel (b).

$$\Delta h = 6.6 - 1.7 \times 10^{-3}\, d \quad [\mu\text{m}] \tag{2}$$

for the visible pulses.

Equations (1) and (2) indicate that the decaying of the ablation rate $\Delta h$ with increasing spot diameters $d$ is one order of magnitude faster in the case of the visible than in the case of the infrared laser pulses (compare the coefficients $0.11 \times 10^{-3}$ and $1.7 \times 10^{-3}$ in equations (1) and (2), respectively). The much faster decay of the ablation rate when using visible wavelength laser pulses indicates that the visible radiation is more appropriate for producing localized, well defined, microstructures by laser ablation.

A normal vaporization of the irradiated sample and the small value of the hydrodynamic length of the laser-produced plasma [2, 8],

$$l_h = v_p \tau \tag{3}$$

relative to the laser spot diameter results, both in the infrared and visible irradiation regimes, from the linear increase of the ablation rate with decreasing of the laser spots diameter in the case of the large spot domains. In equation (3), $\upsilon_p = \sqrt{\gamma k_B T_p / M}$ is the expansion velocity of the plasma plume considered as an ideal gas with adiabatic coefficient $\gamma = 5/3$, the temperature $T_p$ and the atomic mass $M$. For an usual plasma temperature of $5{\cdot}10^4$ K [8, 12], the plasma hydrodynamic length is $\approx 0.04$ mm, which is one order of magnitude smaller than the laser spot. Thus, the plasma expansion is one-dimensional and the attenuation of the laser beam within the columnar plasma plume is proportional to the plume length regardless of the value of the laser spot diameter.

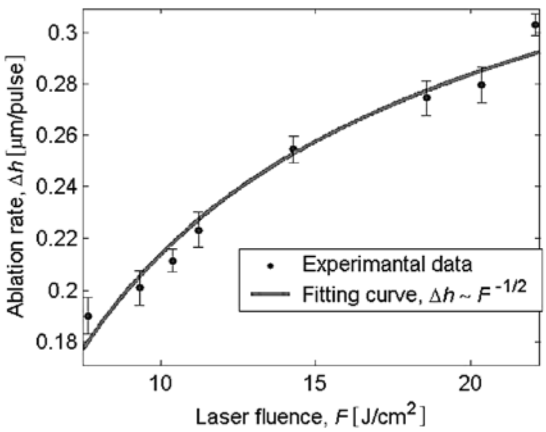
When the laser spot diameters decrease below a threshold value $d_{th}$ ($\approx 0.6$ mm for the visible pulses) a marked non-linear increase of the ablation rate from 5.5 μm/pulse at $d = 0.6$ mm, to 8.0 μm/pulse at $d = 0.4$ mm is observed (Fig. 2b). The transition from the linear to the non-linear increase of the ablation rate with decreasing spot diameters in the visible irradiation regime arise from the transition to the normal vaporization regime (below the threshold diameter) to a phase explosion regime (above the threshold diameter) of the metallic sample. This explosion regime is characterized by an enhanced mass removal rate due to the ejection of particulates and melt-droplets from the irradiated sample.

The higher efficiency of ablation in the case of the visible radiation than in the case of the infrared radiation is explained by the competing effects of two phenomena. First, the decrease of the optical absorptivity of aluminum upon increasing the visible wavelength leads to strong localized heating and an enhanced evaporation of the sample. Second, the dependence of the plasma coefficient $\alpha_{IB}$ on the wavelength $\lambda$,

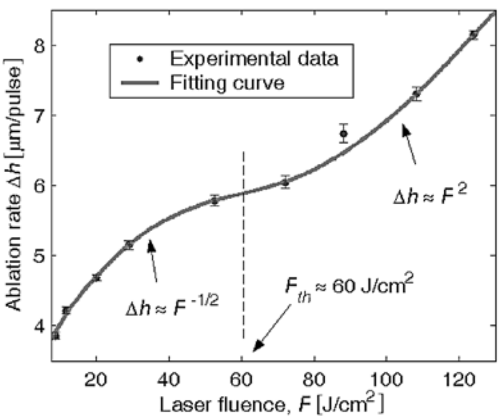
$$\alpha_{IB} \approx \lambda^3 \tag{4}$$

implies that the attenuation of the incoming laser beam into the ignited plasma-plume via the inverse-Bremsstrahlung effect is significantly stronger in the case of longer wavelengths [2, 5, 12]. As a consequence, a large part of the laser-pulse energy is spent for heating the plume, and not for ablating the metallic sample.

The increase of the laser spot diameter leads to a decay of the laser fluence at the target surface. The dependence of the ablation rate of aluminum on the fluence $F$ of the laser pulses in the infrared (Fig. 3a) and visible (Fig. 3b) radiation was assessed by maintaining a constant flux of 72 MJ/s in the case of the infrared laser, and 36 MJ/s for the visible wavelength laser. The results indicate that by increasing the fluence in the low to medium regimes (*i.e.*, up to $\approx 50$ J/cm$^2$ for the visible, and $\approx 30$ J/cm$^2$ for the infrared laser radiation), the ablation rate increases inversely proportional with the square-root of the laser fluence.




a)




b)

Fig. 3 – Ablation rate of aluminum as a function of the laser fluence of a 4.5 ns laser pulse. The laser wavelength is 1064 nm in panel (a), and 532 nm in panel (b).

In the case of the infrared pulses, the fitting curve is described by the equation

$$\Delta h = -0.8/\sqrt{F[\mathrm{J/cm^2}]} + 0.5 \ [\mu\mathrm{m}], \tag{5}$$

whereas for the for the IR pulses the fitting curve is

$$\Delta h = -9.8/\sqrt{F[\mathrm{J/cm^2}]} + 7.1 \ [\mu\mathrm{m}]. \tag{6}$$

Above a threshold value of the laser fluence $F_{th}$, which for the visible pulses is ≈ 60 J/cm$^2$, a marked quadratic increase of the ablation rate with the fluence take place ( Fig. 3b), described by the equation:

$$\Delta h = 1.9\times10^{-4}\ (F[\mathrm{J/cm^2}])^2 + 5.2 \ [\mu\mathrm{m}]. \tag{7}$$

The quadratic increase of the ablation is accompanied by a significant amount of bright sparks that emerge into the plume that appear owing to the transition from a normal vaporization of the irradiated sample, characterized by a small mass-removal rate, to phase-explosion, characterized by ejection of small particulates and melt droplets, and an enhanced mass-removal rate.

## 4. CONCLUSIONS

The results discussed here on the impact of the laser spot diameter and laser fluence on the ablation rate of aluminum demonstrate that, for the both visible and infrared nanosecond-pulse irradiation regimes, at a constant flux of energy at the sample surface and large spot-diameters, the ablation rate increases linearly with decreasing the diameter of the laser spot. The maximum ablation rate in the linear regime is ≈ 5.5 μm/pulse for the visible beam focused to a 0.6 mm spot-diameter, and ≈ 0.35 μm/pulse for an infrared laser beam focused to a 1 mm spot diameter. This indicates that the coupling to the metallic sample is more efficient in the case of the visible radiation than in the case of the infrared radiation. The linear decrease of the ablation rate with increasing laser spot diameters denotes a normal evaporation at the irradiated surface, and an uniform absorption of the processing laser beam within the laser-produced plasma plume whose hydrodynamic length has a value similar to the laser spot diameter.

Decreasing the spot diameter results in an increase of the laser fluence of the incident laser beam and, consequently, to an increase of the ablation rate that is inversely proportional with the square-root of the laser fluence. In the particular case of the visible wavelength laser, decreasing the spot diameter below a threshold value of ≈ 0.6 mm and, hence, increasing the fluence above a threshold fluence of ≈ 60 J/cm$^2$, greatly increases the ablation rate. The significant increase of the ablation rate, accompanied by the appearance of a significant number of bright sparks within the plasma plume, indicates the transition from the normal vaporization regime to a phase-explosion regime.

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