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The dependence of the ablation rate of metals on nanosecond laser fluence and wavelength

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The dependence of ablation rate of aluminium, titanium and copper on the nanosecond laser fluence at 532 nm and, respectively, 1064nm wavelengths is investigated in atmospheric air. The wavelength is varied by exchanging the fundamental and second harmonic modules of a Q-switched Nd-YAG laser system, while the fluence of the pulses is varied by changing the diameter of the irradiated area at the target surface. The results indicate an approximately logarithmic increase of the ablation rate with fluence for both wavelengths, and an approximately double ablation rate in the case of visible pulses as compare to infrared pulses. By extrapolating the ablation rate vs. fluence fitting curve toward zero, we estimate the ablation threshold fluence, F_{th}. The ablation threshold fluence is strongly dependent on the wavelength and target material. Thus, F_{th} in the case of infrared pulses is twice as large as F_{th} corresponding to the visible pulses, while F_{th} is lower for the aluminium and titanium as compared to the copper. A different behavior occurs in the case of copper, for which we obtained a two times smaller F_{th} when using infrared pulses comparative with the visible pulses.

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1. Introduction

The material removal efficiency under the action of short and high intensity laser pulses is described by the ablation rate, Δh, which gives the maximum of the layer thickness ablated during a laser pulse. Understanding and controlling the ablation rate of metals is an essential key parameter in micropatterning and pulsed laser deposition (PLD), optoelectronics and micromechanics [1-8].

The ablation rate is strongly influenced by the characteristics of the laser beam (e.g., pulse duration, number of pulses, energy, fluence, wavelength) [1, 2, 5, 6, 8-18], processed material (e.g., mass density, surface reflectivity, optical absorptivity, thermal conductance) [1-6,19,20], and by ambient conditions [1, 20, 21]. Previous experiments on ablation indicate that the ablation rate increases logarithmically with fluence in the case of single metals, semiconductors and dielectrics [1, 2, 9, 10]. They also demonstrate that the wavelength of the laser has a strongly influence on the ablation rate: the shorter wavelength, the higher ablation. This behavior is mainly attributed to the superposition of three inter-related phenomena: the decrease of the intrinsic absorption of metals, semiconductors and dielectrics, the increase of the surface reflectivity, and the increment of the absorptivity of the produced plasma with increasing wavelength.

Here, we investigate experimentally the dependence of the ablation rate of different metals (aluminium, copper and titanium) on fluence of the nanosecond laser in atmospheric air at two wavelengths: 1064 nm and 532 nm. The wavelength is varied by exchanging the fundamental and second harmonic modules of a Q-switched Nd-YAG laser system. The variation of the fluence of the pulses on the metallic surface was made by changing the diameter of the irradiated area on the sample surface. By extrapolating the ablation rate vs. fluence fitting curves toward zero, we estimate the ablation threshold fluence, F_{th}. The experiments demonstrate additionally the influence of the thermal and optical properties of metals on the efficiency of the ablation.

2. Experiments

The experiments were carried out in atmospheric air with a Q-switched Nd-YAG laser system that works in the TEM00 mode and generates fundamental pulses at 1064 nm wavelength. By sending them through a second harmonic generator module we doubled the frequency (532 nm wavelength). The laser pulses are characterized by duration of 4.5 ns, a repetition rate of 10 Hz, and energies of 360 mJ/pulse and 180 mJ/pulse for the fundamental and second harmonic pulses, respectively. The metallic targets (aluminium, copper and titanium plates thicker than 1 mm) are placed in the focal plane of a convergent lens (f/10, f=10 cm). In order to vary the fluence of the laser beam, we varied the diameter of irradiated area by moving the targets away from focal plane. In the focal plane of focusing lens, for the fundamental pulses, the diameter of irradiated area is ≈ 1.1 mm. In the case of second harmonic pulses, the diameter of the irradiated area is ≈ 0.2 mm for aluminium and copper and ≈ 0.3 mm for the titanium. To avoid the effect of air breakdown that would lead to a loss of energy in heating plasma plume ignited in front of the targets, the samples were translated with micrometer resolution axially toward the incoming laser beam by using a mechanical stage on which the target were clamped. The samples are translated axially with increments of 2 mm in both wavelengths regimes. Because the diameter of the crater is higher than its depth (the aspect ratio is < 1), the ablation can be considered as one-dimensional and, consequently, the diameter of the irradiated area could be approximated by the diameter of the crater that is drilled into the metallic targets [20, 22]. The fluence was determined by dividing the energy of the pulse by the irradiated area [2].

The diameter and the depth of the crater were measured using a metallographic microscope with micrometric resolution. The ablation rate was calculated by dividing the crater depth by the number of laser pulses used to drill the crater in each particular position of the targets along their axial path. In order to obtain a crater sufficiently deep for measurement of the ablation rate with a relative error of maximum 5% we irradiated the targets with a number of 20 consecutive pulses. This number ensures an approximately constant ablation rate during multiple pulses drilling [1, 15].

3. Results and discussion

The dependence of ablation rate (*Δh*) of metals on the laser fluence (*F*) for the fundamental (1064nm) and second harmonic (532 nm) laser pulses, in air, is depicted in Fig. 1.

For the case of the second harmonic pulses (Fig. 1 a) an increment of the fluence to a value of ~470 J/cm^2 leads to an approximately logarithmic increase of the ablation rate of aluminium to a maximum of ~9.5 µm/pulse (fig.1a, solid line). In the case of titanium, the increase of the fluence to a value of ~210 J/cm^2 leads to a logarithmic increase of ablation rate to a maximum value of ~7.0 µm/pulse (Fig. 1 a, dash line), while for copper the increase of fluence to ~660 J/cm^2 leads to maximum value of the ablation rate of ~7.0 µm/pulse (Fig. 1 a, dot line).

The fitting curves are described by the equations

$$\Delta h = 1.7\ln[F(\mathrm{J/cm^2})] - 1.0\ (\mu\mathrm{m}) \tag{1}$$

for aluminium,

$$\Delta h = 1.1\ln[F(\mathrm{J/cm^2})] - 0.1\ (\mu\mathrm{m}) \tag{2}$$

for titanium, and

$$\Delta h = 1.5\ln[F(\mathrm{J/cm^2})] - 3.1\ (\mu\mathrm{m}) \tag{3}$$

in the case of the copper.

When fundamental pulses were used (fig 1b), an increase of the fluence to a value of ~25 J/cm^2 leads to an approximately logarithmic increase of the ablation rate of aluminium to a maximum of ~5.5 µm/pulse (fig.1 b, solid line). In a similar manner, a maximum values of ablation rate ~ 5.3 µm/pulse for titanium (fig.1 b, dash line) and ~4.0 µm/pulse for copper (fig.1 b, dot line) are obtained when fluence increases to ~30 J/cm^2 and ~ 25 J/cm^2, respectively. The fitting curves are described by the equations

$$\Delta h = 2.8\ln[F(\mathrm{J/cm^2})] - 3.8\ (\mu\mathrm{m}) \tag{4}$$

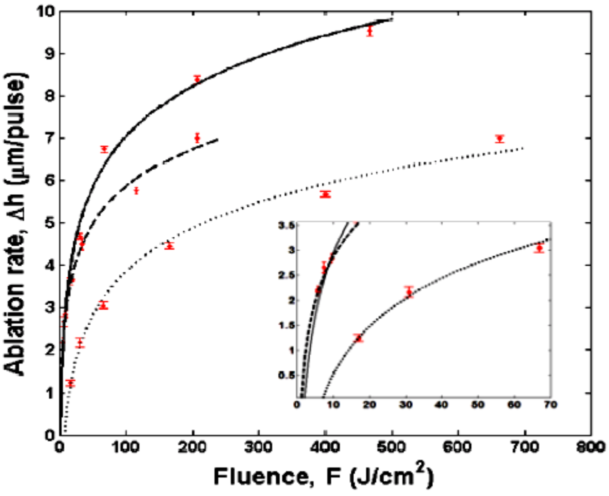
for the aluminium,

$$\Delta h = 2.8\ln[F(\mathrm{J/cm^2})] - 3.9\ (\mu\mathrm{m}) \tag{5}$$

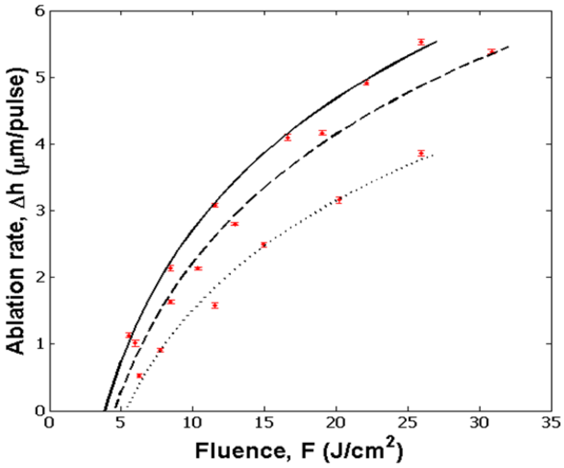
for titanium, and

$$\Delta h = 2.4\ln[F(\mathrm{J/cm^2})] - 4.2\ (\mu\mathrm{m}) \tag{6}$$

in the case of copper.



(a)



(b)

Fig. 1. Ablation rate of metals in air as a function of laser fluence of a 4.5 ns laser pulses for (a) 532nm and (b) 1064nm wavelengths. The solid line represent aluminium, the dash line titanium and the dot line copper. In the inset of (a) is detailed the threshold fluence zone for visible pulses.

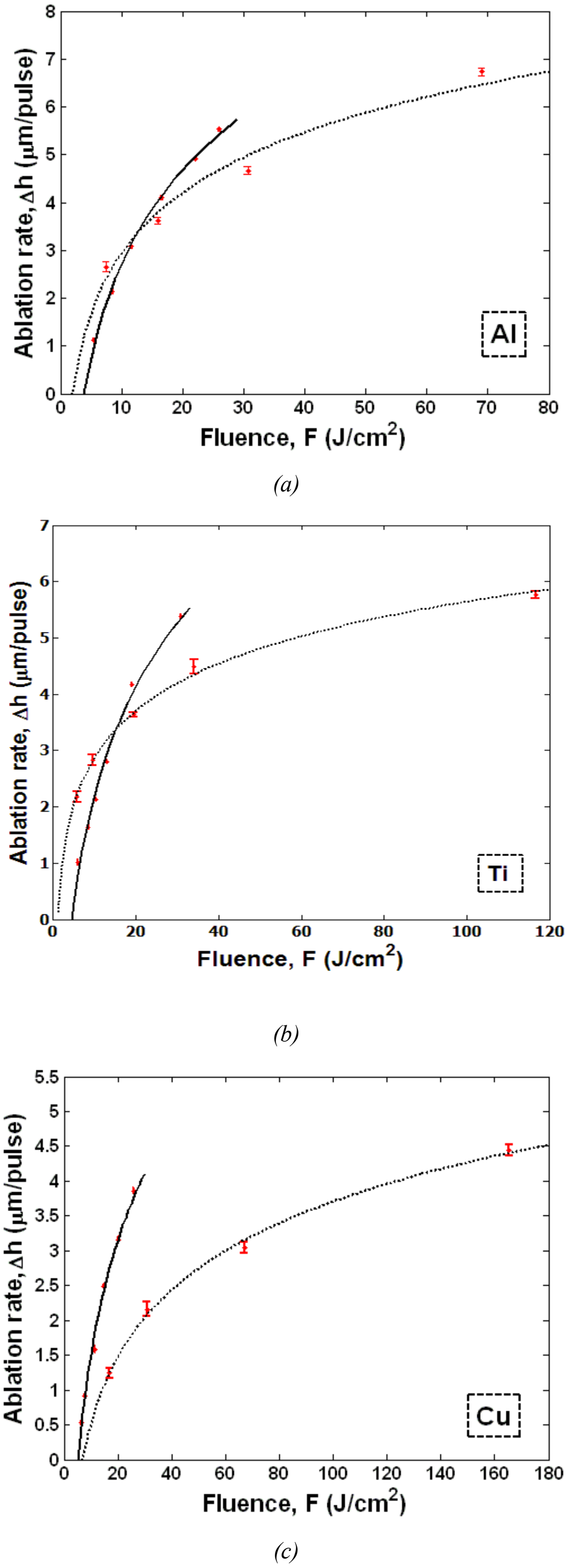
The differences between ablation rates of metals in visible and infrared irradiation regimes originate in the different optical and thermal proprieties at these wavelengths. For metals, absorption and reflection of short light pulses are linear at intensities of the order of 10^{13} W/cm^2 that we used in this experiment [23]. Optical absorption is usually dominated by electrons absorption through inverse-Bremsstrahlung [1, 23]. In the visible and infrared spectral region, for most metals the linear absorption coefficient α is in the range $(5\div15)\times10^5$ cm^{-1}. At the nanosecond scale of the laser pulse, the electrons and the atoms equilibrate leading to a strong heating of the irradiated volume, the localization of the heating being determined by the thermal depth $l_{th} = \sqrt{D\tau_l}$: the smaller is l_{th}, the more localized is the heating. Here, D and τ_l state for the thermal diffusivity and pulse duration, respectively. Table I [1, 26] indicates that the optical depth $l_\alpha=1/\alpha$ and thermal depth l_{th} of copper at 532 nm wavelength are larger than titanium's and aluminium's. This leads to a higher ablation rate of aluminium (~8.5 μm/pulse) and titanium (~7.0 μm/pulse) as compare to copper (~5 μm/pulse) at a fluence of ~200 J/cm^2.

Table 1. Thermal and optical proprieties of selected metals.

Material	Cu	Ti	Al
ρ (g/cm^3)	8.94	4.52	2.70
R	0.43 [0.5] 0.98 [1.06]	0.49 [0.5] 0.55 [1.06]	0.92 [0.5] 0.94 [1.06]
α (cm^{-1})	7.1E5 [0.5] 7.7E5[1.06]	1.1E6 [0.5] 1.3E6[1.06]	1.5E6 [0.5] 1.0E6 [1.06]
l_α (nm)	14 [0.5] 13 [1.06]	9.1 [0.5] 7.7 [1.06]	6.7 [0.5] 10 [1.06]
c_p (J/gK)	0.39	0.52	0.90
$k^{x)}$ (W/cmK)	4.01	0.21	2.37
D (cm^2/s)	1.15	0.09	0.98
l_{th} (μm)	1.43	0.40	1.32

x)For T=300K
The square brackets contain the wavelength in μm.

In a similar manner, when fundamental pulses were used higher value of ablation rates for aluminium and titanium (~5.5 μm/pulse and ~5.3 μm/pulse, respectively) are obtained comparative with the copper (~4.0 μm/pulse). Whereas the maximum ablation rates value for aluminium are obtained at a laser fluence of ~25 J/cm^2, for the titanium this are obtained at a laser fluence of ~30 J/cm^2. Although the thermal penetration depth of the titanium ($l_{th} \approx 0.40\mu\mathrm{m}$) is lower than aluminium ($l_{th} \approx 1.32\mu\mathrm{m}$), the optical absorption coefficient of titanium ($l_\alpha \approx 9.1\mathrm{nm}$), bigger comparative with the aluminium ($l_\alpha \approx 6.7\mathrm{nm}$), prevent the diffusion of the heat in the probe during pulse duration and a higher fluence is necessary to achieve maximum ablation rate.



(a)

(b)

(c)

Fig. 2. Ablation rate of (a) aluminium, (b) titanium and (c) copper, in open air, as a function of laser fluence and wavelength of a 4.5 ns laser pulses. The solid line represent results obtained at 1064 nm and the dot line results obtained at 532 nm.

By extrapolating the ablation rate vs. fluence fitting curves described by relations (1-6) toward zero, we estimate the threshold fluence, F_{th} (Fig.1, (a) and (b)). For 532 nm wavelength the threshold values of fluences are ~2.0 J/cm^2 for aluminium, ~1.0 J/cm^2 for titanium, and ~8.0 J/cm^2 for copper. At 1064 nm wavelength we obtained a threshold laser fluence of ~4.0 J/cm^2 for aluminium, ~4.5 J/cm^2 for titanium and ~5.5 J/cm^2 in the case of copper. The larger ablation threshold fluence of copper can be related to his larger thermal depth as compare to aluminium and titanium as follows. The large thermal depth of copper results in a less localization of the excitation energy and, thereby, in a more difficult process of melting and the evaporation of the copper surface.

The results on the existence of a higher ablation rate for a given fluence in the case of visible pulses as compared infrared pulses (Fig. 2), originate in the superposition of the three phenomena: increase of the optical length and metallic surface reflectivity with wavelength, oxidation of the surface during the interval between the pulses, and the increase of the plasma absorption coefficient with wavelength. The incoming laser light can be absorbed partly or completely by the plasma plume through inverse-Bremsstrahlung which is characterized by the absorption coefficient [1, 28]

$$\alpha_{IB} \sim \lambda^3 \left[1 - \exp\left(-\frac{hc}{\lambda k_B T_e} \right) \right] \text{ (cm}^{-1}\text{)} \qquad (8)$$

where λ the wavelength (in cm), T_e the electron temperature (in K), h Planck's constant, k_B Boltzmann's constant, and c the velocity of light. Since the ratio of the laser energy hc/λ to the plasma temperature $k_B T_e$ enters into the exponential as $hc/\lambda k_B T_e$, the dynamic behavior of the system changes for variations in the wavelength and the plasma energy. The plasma absorption increases drastically with increasing wavelength and, thereby, the laser light at 1064 nm is shielded stronger than light at 532 nm.

4. Conclusions

The dependence of the ablation rate of different metals (aluminium, copper, titanium) on the nanosecond laser fluence was investigated at 1064 and 532 nm wavelengths in atmospheric air. We varied the fluence of the laser pulses by changing the diameter of the irradiated area on the probe surface while the wavelength was changed by exchanging the fundamental and the second harmonic modules of a Q-switched Nd-YAG laser system. The results indicate that the ablation rate has an approximately logarithmic increase with fluence in both wavelength regimes. When visible pulses were used, we obtained a maximum ablation rate of ~9.5 μm/pulse for aluminium, ~7.0 μm/pulse for titanium, and ~7.0 μm/pulse for copper. In the case of using infrared pulses we obtained an approximate two times smaller ablation rate as compare to the visible pulses: ~5.5 μm/pulse for aluminium, ~5.3 μm/pulse for titanium, and ~4.0 μm/pulse for copper. The smallest ablation rate obtained in the case of copper is related to its large values of the thermal and optical lengths as compare to titanium's and aluminium's.

The smaller ablation rates obtained with the infrared pulses indicate a much higher efficiency of the ablation in the case of second harmonic pulses. This is due to the superposition of following effects: the increase of the optical length and optical reflectivity of metallic samples with wavelength, the weaker oxidation of the surface target irradiated at short wavelengths, and the enhanced shielding of the incident beam via inverse-Bremsstrahlung into the ignited plasma at the higher wavelengths. By extrapolating the ablation rate vs. fluence fitting curves toward zero we estimate the ablation threshold fluence F_{th}. In both wavelength regimes we obtained lower F_{th} for the aluminium and titanium (~2.0 J/cm^2 and ~1.0 J/cm^2 respectively at 532 nm, and ~4.0 J/cm^2 and ~4.5 J/cm^2 respectively at 1064 nm) as compared with copper (8.0 J/cm^2 for VIS and 5.5 J/cm^2 for UV pulses), due to their small thermal diffusivities that leads to a localization of the excitation energy into a smaller volume.

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