NANOPULSED ABLATION RATE OF METALS DEPENDENCE ON THE LASER FLUENCE AND WAVELENGTH IN ATMOSPHERIC AIR

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În acest articol este investigată dependența ratei de ablație a aluminiului, titaniului şi a cuprului de fluența nanopulsurilor laser, având lungimile de undă de 1064 nm, recpectiv 532 nm în condiții atmosferice normale. Modificarea lungimi de undă a fost realizată prin utilizarea pulsurilor fundamentalei şi a armonicii a doua a unui laser Nd-YAG Q-switched, iar variația fluenței s-a realizat prin modificarea diametrului spotului laser pe suprafața probei iradiate. Rezultatele obținute indică o creştere logaritmică a ratei de ablație cu fluența laser, în cazul ambelor regimuri de iradiere, şi o eficiență mai mare a ablațieila 532 nm.

The dependence of ablation rate of aluminum, titanium and copper on the nanosecond laser fluence with 532 nm and respectively 1064 nm wavelengths is investigated in atmospheric air. The wavelength is varied by using the fundamental and second harmonic of a Q-switched Nd-YAG laser system and the fluence of the pulses is varied by changing the diameter of the irradiated area at the surface target. The results indicate an approximately logarithmic increase of the ablation rate with the fluence for both irradiation regime and a higher efficiency of the ablation in the case of 532 nm pulses.

Keywords: ablation rate, nanosecond laser pulses, metals

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 [1-5].

The ablation rate is strongly influenced by the characteristics of the laser beam (e.g., pulse duration, number of pulses, energy, fluence, wavelength) [1,2,4,6-16], processed material (e.g., mass density, surface reflectivity, optical absorptivity, thermal conductance) [1-4,14,15], and by ambient conditions [1,15]. The experiments indicate that the ablation rate increases logarithmically with the nanopulses

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fluence in the case of single metals, semiconductors and dielectrics [1,2,6] and the wavelength of the laser has a strongly influence on the ablation rate, e.g., the shorter wavelength, the higher ablation rate. Here, we investigate the dependence of the ablation rate of aluminium, copper and titanium on the wavelength and fluence of nanopulsed laser in atmospheric air, using two irradiation regimes: 1064 nm and 532 nm. We demonstrate that the ablation rate increases approximately logarithmic with the fluence for both irradiation regimes and a higher efficiency of the ablation in the case of 532 nm pulses is obtained.

2. Experiments

The experiments were carried out in air atmosphere with a Q-switched Nd-YAG laser system that works in the TEM00 mode and generates fundamental pulses at wavelength of 1064 nm which we doubled the frequency (532 nm wavelength) by sending them through a second harmonic generator module. 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 laser pulses were focused at normal incidence on a 1-mm thick aluminium and 2.5 mm-thick copper and titanium by a focusing lens system. The targets were first placed in the focal plane of a convergent lens (f/10, f=10 cm) and then, to vary the fluence of the laser beam, we varied the diameter of irradiated area by moving them away from focal plane. In the focal plane we obtained a diameter of irradiated area of ~1.1 mm for the fundamental pulses, and ~0.2 mm in the case of second harmonic pulses. 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 increments of 2 mm axially toward the incoming laser beam with the aid of a mechanical stage on which the sample is fixed. 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 [15,16]. The fluence was determined by dividing the energy of the pulse by the irradiated area [2].

From the depth of the crater drilled in multiple-pulse regime into the metallic targets, at each particular position along the axial path, we gathered the dependence of ablation rate on the laser fluence. Because the ablation rate usually depends on the consecutive laser pulses [1,10], the 20 fundamental and second harmonic laser pulses ensure an approximately constant ablation rate for each successive pulse and, moreover, enable us to obtain a crater sufficiently deep to allow measurement of the ablation rate with a relative error of maximum 5%. Then, the ablation rate was calculated by dividing the crater depth by the number of laser pulses used to drill the crater. The depths and the diameter of craters were measured using a metallographic microscope with micrometric resolution.

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 second harmonic pulses (fig. 1 a) an increase of the fluence to a value of ~470 J/cm^2 leads to an approximately logarithmic increase of the ablation rate of aluminum to a maximum of ~9.5 μm/pulse (fig.1 a, solid line). In a similar manner, the logarithmic increase of the fluence to a value of ~210 J/cm^2 leads to an increase of ablation rate of titanium to a maximum value of ~7.0 μm/pulse (fig.1 a, dash line) and the approximately logarithmic increase of fluence to ~660 J/cm^2 leads to maximum value of the ablation rate of copper to ~7.0 μm/pulse (fig. 1 a, dot line). The fitting curves are described by the equation

$$\Delta h = 1.7\ln[\mathrm{F(J/cm^2)}] - 1.00 \qquad (\mu\mathrm{m}) \qquad (1)$$

for aluminum,

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

for titanium, and

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

in the case of the copper.

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

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

for the aluminum,

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

for titanium, and

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

in the case of copper.

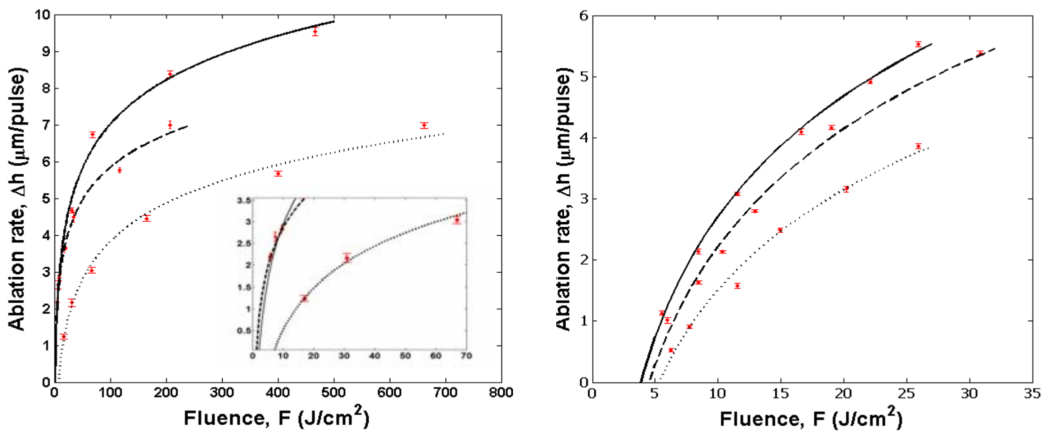

Fig.1 Ablation rate of metals in air as a function of laser fluence of a 4.5 ns laser pulses for (a) 532nm and (b) 1064nm wavelengths. The solid line represents aluminum, the dash line titanium and the dot line copper. In the inset of (a) is detailed the threshold fluence zone for visible pulses.

The differences between ablation rates indicated above originate in the different optical and thermal properties of metals and the wavelength of laser pulses. For metals, absorption and reflection of short light pulses obey the laws of linear metal optics up to intensities of 10^{15} W/cm^2 [17]. Optical absorption is usually dominated by free carrier absorption, i.e. electrons in the conduction band absorb photons through inverse-Bremsstrahlung and gain energy [1,17]. In the visible and infrared spectral region, for most metals the linear absorption coefficient α is in the range (5-15)$\times 10^5$ cm^{-1}, the absorption being responsible for the generation of a highly non-equilibrium state of excited electrons, which relax by electron–electron collisions. The electrons first thermalize among themselves and subsequently lose energy to the lattice. That is strictly true as the absorbed energy density increases. In fact, the higher is the delivered energy the faster is the relaxation process, owing to several factors. First, if a larger energy density is deposited into the material, the temperature of the thermal part of the distribution increases more rapidly. This, in turn, increases the number of available electrons with which the non-thermal electrons can scatter, that is, increases the scattering rate. Moreover, as the thermalization process between the rapidly heating thermal component and the non-thermal part of the distribution goes on, the non-thermal part becomes more and more composed only of the higher energy electrons, which have the shortest energy relaxation time. All these factors lead to a decrease in the thermalization time as the excitation density increases.

Table I

Thermal and optical proprieties of selected metals

Material	***ρ(g/cm³)***	***α* (λ[μm]) (cm⁻¹)**	***l*$_α$ (λ[μm]) *(nm)***	***c*$_p$ *(J/gK)***	***k* (W/cmK)***	***D* *(cm²/s)***	***l*$_{th}$ *(μm)***
Al	2.70	1.5E6 (0.532) 1.0E6 (1.06)	6.7 (0.532) 10 (1.06)	0.90	2.37	0.98	1.32
Cu	8.94	7.1E5 (0.532) 7.7E5 (1.06)	14 (0.532) 13 (1.06)	0.39	4.01	1.15	1.43
Ti	4.52	1.1E6 (0.532) 1.3E6 (1.06)	9.1 (0.532) 7.7 (1.06)	0.52	0.21	0.09	0.4

**For T=300K*

The results obtained with second harmonic pulses indicate a higher value of ablation rates in the case of aluminum in comparison with titanium and copper. This occurs from different values the optical penetration deep $l_α=1/α$ and the heat penetration deep $l_{th}≈2(D\tau_L)^{1/2}$ (see table I [1]), i.e. the energy deposited by the laser light in the material bulk, where τ_L is laser pulse length and D is the thermal diffusivity, $D=k/\rho c_p$. The thermal penetration depth of the aluminum ($l_{th} \approx 1.32\mu m$) and titanium ($l_{th} \approx 0.40\mu m$), lower than copper ($l_{th} \approx 1.43\mu m$), implies a reduced thermal diffusivity and achievement to a higher ablation rates, ~9.5 μm/pulse and ~7.0 μm/pulse for aluminum and titanium, at a lower laser fluences (~470 J/cm^2 and ~210 J/cm^2, respectively) in this metals comparative with maximum ablation rates of copper ~7.0 μm/pulse obtained at a laser fluence of ~660 J/cm^2. Further, because the heat penetration deep of titanium is approximately one order of magnitude lower than heat penetration deep of the copper, the heat diffusion is smaller in the titanium probe and a lower fluences ~210 J/cm^2 is necessary to obtain a maximum value of ablation rate ~7.0 μm/pulse, whereas approximately the same value of ablation rate on the copper sample are obtained at a fluence of ~660 J/cm^2.

In a similar manner, when fundamental pulses are 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 these are obtained at a laser fluence of ~30 J/cm^2. Although the thermal penetration depth of the titanium ($l_{th} \approx 0.40\mu\text{m}$) is lower than aluminium ($l_{th} \approx 1.32\mu\text{m}$), the optical absorption coefficient of titanium ($l_\alpha \approx 9.1\text{nm}$), bigger comparative with the aluminium ($l_\alpha \approx 6.7\text{nm}$), prevents the diffusion of the heat in the probe during pulse duration and a higher fluence is necessary to achieve maximum ablation rate.

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. How the dissipation of heat in a volume is determined by the laser spot diameter and l_{th} [18] the large thermal depth of copper leads to 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 above on the existence of a higher value of ablation rate for a given fluences in the case of visible pulses instead of infrared pulses (Fig. 1) originate in the superposition of the three phenomena: increases of the optical absorptivity upon decreasing wavelengths, oxidation of the surface during the interval between the pulses and the dependence of the absorption coefficient of the plasma α_{IB} on the laser wavelengths. The incoming laser light can be absorbed partly or completely by the plasma plume by inverse-Bremsstrahlung (IB) processes and direct single-photon excitation processes. The absorption coefficient α_{IB} can be described [1,17] by

$$\alpha_{IB} \approx 1.369\times 10^{-23}\cdot \lambda^3 \frac{Z^3 N_i^2}{T_e^{1/2}}\left[1-\exp\left(-\frac{h\nu}{k_B T_e}\right)\right] (\mathrm{cm}^{-1}) \tag{9}$$

where Z is the charge state of the ion, N_i is the ion density, λ the wavelength (in cm), T_e the electron temperature (in K), h Planck's constant, ν the laser frequency and c the velocity of light. For an usual plasma the absorption increases drastically with increasing wavelength, e.g. laser light at 10.6 μm has an absorption coefficient which is several orders of magnitude larger that than of IR light at 1064 nm and that for VIS and UV lasers.

The dependence of F_{th} on the laser wavelength is related of following phenomena. First, the increase in intrinsic absorption of metals α with decreasing wavelengths [1,2,19] leads to a strong heating of the surface sample, because the energy will be localized within a smaller volume. Second, laser induced plasma formation plays an important role, in particular with infrared pulses, because the absorption coefficient of the plasma α_{IB} is much higher in the case of longer wavelengths (see eq. (9)) [1,2,7,8]. As a direct consequence of this, the plasma produced by the leading crest of the infrared pulse becomes more absorbing for the remaining pulse as compared with visible pulse. This prevents the energy of infrared pulse reaching the surface shortly after the onset of the pulse. Third, the higher reflectivity of the metals in IR prevents efficient coupling of the pulse energy to the metals surface [1].

4. Conclusion

The dependence of the nanosecond ablation rate of aluminium, copper and titanium on the laser fluence and wavelength was investigated in atmospheric air. We varied the fluence of laser pulses by changing the diameter of the irradiated area on the probe surface, and the wavelength by using the fundamental and the second harmonic beams of a Q-switched Nd-YAG laser system. The results indicate that the ablation rate has an approximately logarithmic increase with the fluence, for the both irradiation regime, to maximum values of ~9.5 μm/pulse for aluminum and ~7.0 μm/pulse for titanium and copper when visible pulses was used. The smaller ablation rates obtained with the infrared pulses, ~5.5 μm/pulse for aluminum, ~5.3 μm/pulse for titanium and ~4.0 μm/pulse for copper 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 absorptivity of metals with decreasing wavelengths, the weaker oxidation of the surface target irradiated at short wavelengths and the increase of the absorption, in the ignited plasma plume via the inverse-Bremsstrahlung effect, at the higher wavelengths. By extrapolating the fitting curves towards zero ablation rate we estimate the threshold fluence F_{th}. In both irradiation regimes we obtained lower F_{th} for the aluminum and titanium as compared with copper, due to the smaller thermal diffusivity of the heat in these samples that leads to a localization of the excitation energy into a smaller volume. The dependence of F_{th} on the wavelength is related of following effects: increase of the absorption of metals with decreasing wavelengths, the higher reflectivity of the metals in IR that prevents efficient coupling of the pulse energy to the metals surface and the plasma produced by the leading crest of the infrared pulse becomes more absorbing for the remaining pulse as compared with the visible pulse.

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