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CO_2 AND Nd:YAG LASER RADIATION INDUCED DAMAGE IN ALUMINIUM

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The change in the electrical properties of pure aluminium (Al 99.999%) after exposure to CO_2 (energy = 2.5 J/pulse, wavelength = 10.6 μm, pulse duration = 200 nsec) and Nd:YAG (energy = 10 mJ/pulse, wavelength = 1.06 μm and pulse duration = 12 nsec) laser radiation is investigated. The samples were exposed to laser radiations for different numbers of pulses. The change in electrical characteristics of Al is studied under different ambient conditions, after irradiating the samples in air, vacuum and hydrogen at different pressures. After exposure, the electrical conductivity of Al is measured by the four probe method. The electrical conductivity decreases with increasing number of pulses. The damage in air and in hydrogen is more pronounced than in vacuum which can be attributed to collisionnal sputtering of Al by plasma ions of air molecules and hydrogen, respectively. The change in the conductivity in hydrogen is pressure-dependent. Some theoretical considerations are also made, e.g. the phonon speed in Al during the photon interaction, minimal melting and evaporation energy per volume, damage threshold energy, penetration depth, the mass of heated volume and average temperature rise at the Al surface during laser irradiation.

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Keywords: TEA CO_2 laser radiation, Nd:YAG laser radiation, damage, electrical conductivity, collisionnal sputtering

1. *Introduction*

The interaction of intense pulsed UV, visible, and infrared laser radiation with solid materials is of considerable current interest in pulsed-laser deposition, laser ablation, and laser sputtering. If laser radiation is focused and absorbed by the surface of a solid material, a variety of processes may occur including excitation, ionization, sputtering, surface heating, melting, vaporization and resolidification, leading to changes, for example, in electrical properties.

Many authors [1 − 3] have studied effects of different parameters of laser radiation like wavelength, pulse duration, different number of pulses and power densities for the study of ablation rate, radiation damage, structural changes, and surface hardness of metals and other materials.

Rafique et al. [4] have measured the electrical conductivity of CdSe after irradiation with neutrons using the four-probe method. The damaging process, observation of fracture and numerical simulations of Al index stressing by intense CO_2 laser radiation was observed by Haitoe et al. [5]

Necheare et al. [6] discussed the change of the resistance of thin Al films irradiated by ruby laser ($\lambda = 694$ nm) radiation. The increase in the bulk resistance of the film with decreasing thickness is evidently due to an increase in the density of defects.

Kapenieks et al. [7] have studied the electrical resistance per unit length of laser-direct writings on PLTZ (Lanthanum doped PZT) as a function of laser power (337 − 356 nm cw Kr^{+1} laser). With the increasing laser intensity, the degree of reduction increases and electrical properties of laser-treated regions change from semiconducting to metallic.

In the present work, the results are presented for the irradiation of Al in air, vacuum and hydrogen at different pressures for various pulse numbers of TEA CO_2 and Nd:YAG laser radiation.

2. *Experimental set up for the exposure of Al with CO_2 and Nd:YAG laser radiation*

2.1. *Lasers*

Transversely excited atmospheric (TEA) CO_2 and Nd:YAG laser radiation induced plasma effects on polycrystalline pure aluminium (99.999 %) is investigated by measuring electrical conductivity. A schematic of the experimental set up for the exposure of the samples with CO_2 and Nd:YAG laser radiation is shown in Fig. 1.

The 10.6 μm, 200 ns TEA CO_2 laser radiation with pulse energy 2 J and power 10 MW, and the 1.06 μm, 12 ns Nd:YAG laser radiation with pulse energy 10 mJ and power 1.1 MW are used to irradiate annealed aluminium samples (1.5 × 1.5 cm^2). The energy of the laser is measured with the help of power-energy meter Scientech model 362 (Boulders Co., USA). The experiments are performed under vacuum at 10^{-3} mbar, in hydrogen at 130 and 1000 mbar and in air at 1000 mbar. The samples are exposed for different numbers of pulses.

CO_2 laser is electrically pumped tandem laser. It is operated by charging and discharging of two capacitors having capacitance 0.075 μF and working voltage

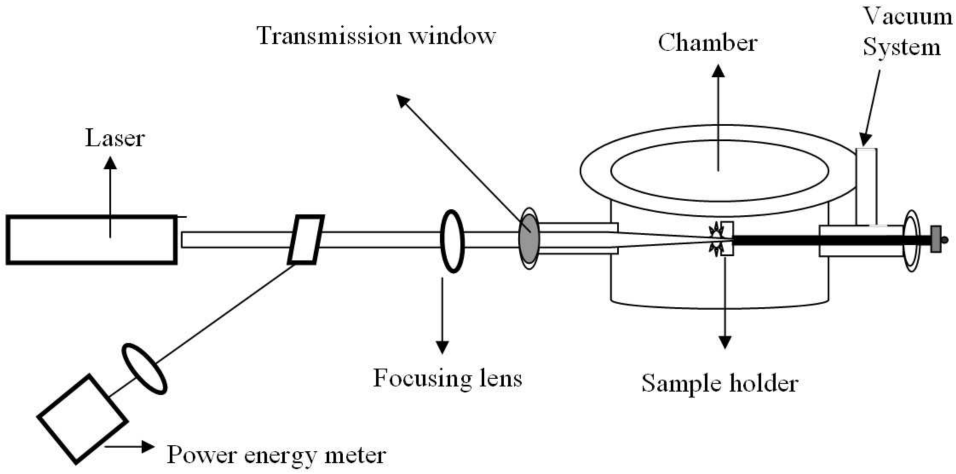

Fig. 1. Scheme of the experimental set up for the exposure of Al *sample to laser radiation.*

50 kV. A HV 28 kV DC (40 kV, 50 mA DC power supply Had Land UK) is used to charge the capacitor. The mixture of $CO_2 : N_2 :$ He (11 : 6 : 83) is used as lasing medium. Its flow rate is 3 l/m in the laser cavity. The spark gap along the triggering circuit and trigger transformer of 20 kV AC is used to synchronize numerous high voltage events within several nanoseconds. The gas used in the spark gap is nitrogen, at the flow rate of 1.5 – 2 l/m.

Nd:YAG laser used in the experiment is a flash-lamp-pumped solid-state laser. It is passively Q-switched with natural cooling system.

CO_2 laser is focused by planoconvex germanium lens and Nd:YAG laser is focused by IR planoconvex lens of focal length 15 cm for each lens. A very bright and luminescent plasma can be observed by naked eye at focal point. The samples are placed exactly at the focus point inside the chamber and are irradiated for 50, 100, 150 and 200 pulses under different ambient conditions.

2.2. Electrical conductivity measurement

2.2.1. **Designing and fabrication of four-probe apparatus**

For the measurement of electrical conductivity of the irradiated samples by laser radiation, the four-probe apparatus was designed and fabricated as shown in Fig. 2. Perspex was used for insulation and Cu probes were used. In order to avoid the thermal emf, the four probes were properly insulated by sleeves and connected by wiring copper-to-copper. The fly-nuts arrangement and two springs provide uniform upward and downward movement of copper rods.

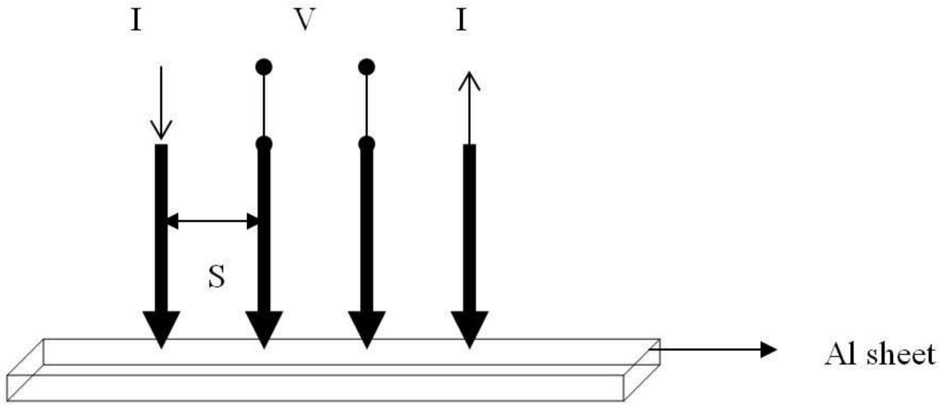

Fig. 2. Four-point-probe method for the measurement of electrical conductivity of Al. *I and V denote the current and voltage, respectively, and S is the spacing between the equidistant probes. It is 4* mm *in the present case.*

2.2.2. **Experimentation**

The in-line arrangement of the probes was used. The experimental arrangement used for electrical conductivity measurement is shown in Fig. 2. A Keithley 2400 sourcemeter with resolution of 1 pA was used as a current source to apply the current across the two end probes. A Keithley 2182 nanovoltmeter with resolution of 1 nV was used to measure the corresponding nanovolt signal across the two central probes.

The electrical conductivity, σ, is measured by the using following formula [8]

$$\sigma = \frac{I}{2\pi VS}, \tag{1}$$

where I = applied current (mA), V = corresponding voltage (nV), S = spacing between the probes = 4 mm (the same for all probes).

3. Results

A comparison of the decrease in the conductivity of aluminium exposed to CO_2 and Nd:YAG laser radiation for increasing numbers of pulses in air and in vacuum is given in Figs. 3 and 4. The resistivity of aluminium increases considerably after increasing the number of pulses of laser irradiation.

The conductivity changes of Al irradiated with Nd:YAG laser radiation are smaller than for CO_2 laser radiation, both in air and vacuum, increasingly with larger numbers of pulses.

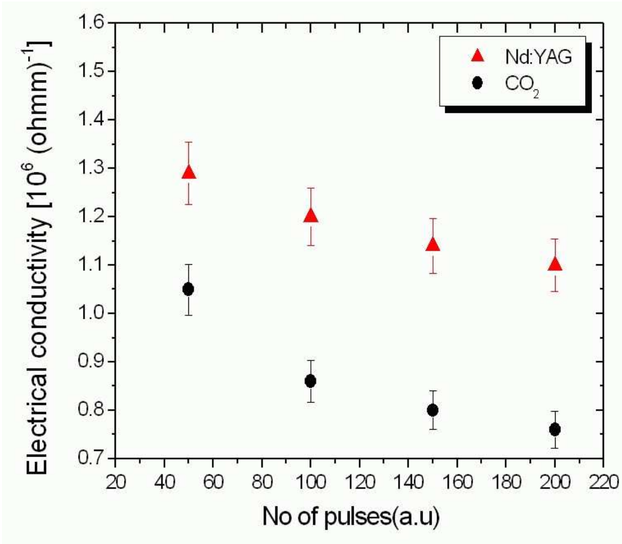

Fig. 3. Change in conductivity of Al *irradiated with* CO_2 *laser radiation (wavelength 10.6* μm, *pulse duration 200* ns, *pulse energy 2* J *and power 10* MW*) and* Nd:YAG *laser radiation (wavelength 1.06* μm, *pulse duration 12* ns, *pulse energy 10* mJ *and power 1.1* MW*) in air at 1000* mbar.

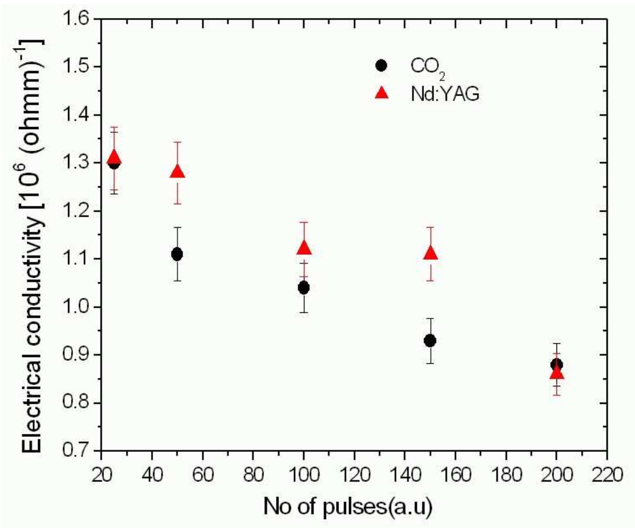

Fig. 4. Change in conductivity of Al *irradiated with* CO_2 *laser radiation (wavelength 10.6* μm, *pulse duration 200* ns, *pulse energy 2* J *and power 10* MW*) and* Nd:YAG *laser radiation (wavelength 1.06* μm, *pulse duration 12* ns, *pulse energy 10* mJ *and power 1.1* MW*) in vacuum* $\sim 10^{-3}$ mbar.

Figures 5 and 6 represent the decrease in the conductivity of Al after irradiation in hydrogen at 130 and 1000 mbar, respectively.

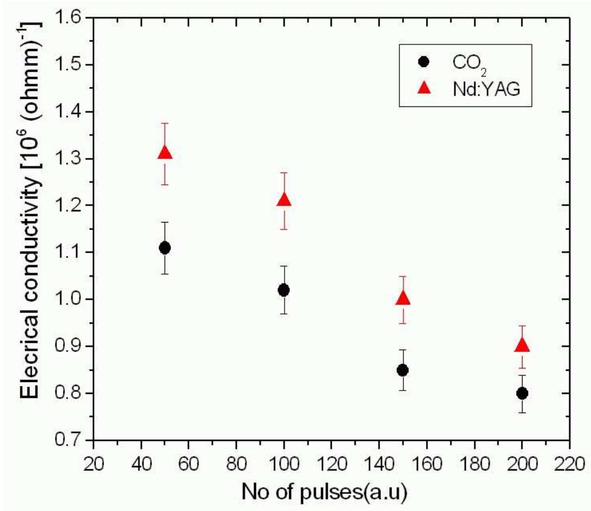

Fig. 5. Change in conductivity of Al *irradiated with with* CO_2 *laser radiation (wavelength 10.6* μm, *pulse duration 200* ns, *pulse energy 2* J *and power 10* MW*) and* Nd:YAG *laser radiation (wavelength 1.06* μm, *pulse duration 12* ns, *pulse energy 10* mJ *and power 1.1* MW*) in hydrogen at 1000* mbar.

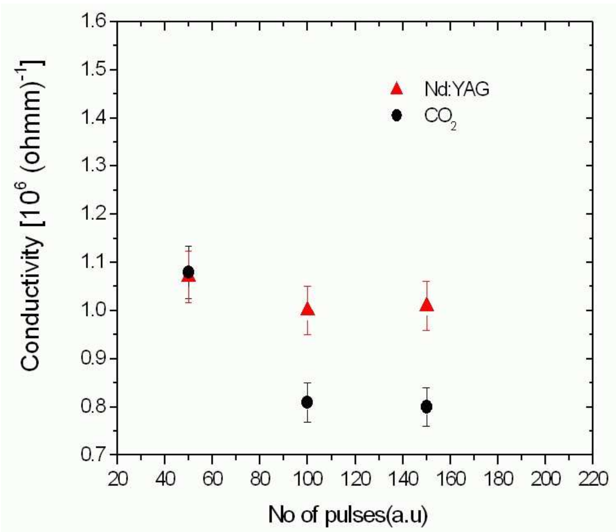

Fig. 6. Change in conductivity of Al *irradiated with with* CO_2 *laser radiation (wavelength 10.6* μm, *pulse duration 200* ns, *pulse energy 2* J *and power 10* MW*) and* Nd:YAG *laser radiation (wavelength 1.06* μm, *pulse duration 12* ns, *pulse energy 10* mJ *and power 1.1* MW*) in hydrogen at 130* mbar.

3.1. Discussion of results

Al sample irradiated both with CO_2 and Nd:YAG laser radiation has shown prominent changes in its electrical properties. Changes in electrical behavior of Al may be due to interstitials, vacancies, clusters, nucleation, voids and spike formation which cause the decrease in electrical conductivity after thermal and collisionnal sputtering caused by irradiation with lasers. Thermal sputtering, in the sense of vaporization from a transiently heated target, may require temperature well above the melting or boiling points [9]. It generally implies a model in which laser light is converted to lattice vibrational energy without bond breaking [10]. The argument of Kelly [11] and Kelly and Rothenberg [12] can be used for describing the process of thermal sputtering. Temperatures for vaporization rate of 1 nm/pulse have been defined for selected solids. For Al it is 3500 K.

Collisional sputtering is a process of indirect sputtering caused by photon-energy transfer to ions produced by laser-induced plasma. If the energy of ions is greater than the displacement threshold energy, then nearby surfaces are ion bombarded and displacement of atoms occurs [13].

For a quantitative characterization of aluminium damage, the data are published by ASTM (American Society for Testing Materials) [14]. According to ASTM, the effective displacement threshold energy, T_{d}, amounts to 16 eV, while the minimum recoil energy to create a stable Frenkal pair, T_{dmin}, is 25 eV, and the activation energies, E_{i}, for thermal migration of interstitials and vacancies amount to $0.1-0.6$ eV for Al. As one CO_2 photon carries the energy of 0.117 eV, and Nd:YAG 1.2 eV, it is a multiphoton process of laser energy deposition for thermal migration of vacancies and interstitials.

With increasing number of pulses, the decrease in electrical conductivity becomes more pronounced, which may be due to the enhanced photothermal and photophysical processes. These enhanced processes may be responsible for the generation of more structural defects and subsequent phase transformation. The photon-phonon interaction can also directly cause the lattice displacements from original position developing with the phonon velocity $v = \chi/l$ [15], which is directly related to the thermal diffusivity χ and the mean free path l. For metals with $l = 10$ nm at 300 K and $\chi = 97.3 \times 10^{-6}$ m/sec for Al, the mean free path reduces to $1-2$ nm at elevated temperature and the calculated phonon speed for Al results in 48.65 m/s at 300 K. With the increasing number of pulses, the surface of Al is heavily irradiated causing oxidation accompanied with possible roughness which causes a decrease in reflectivity and hence in the electrical conductivity. DC conductivity and absorption are related by the following relation [16]

$$\left[\frac{1-R(T)}{1-R(T_0)}\right]_{\mathrm{fee}} = \sqrt{\frac{\sigma_0(T_0)}{\sigma_0(T)}}, \text{ for } \omega \leq 1/\tau \text{ (far IR region)}, \tag{2}$$

$$\left[\frac{1-R(T)}{1-R(T_0)}\right]_{\mathrm{fee}} = \frac{\sigma_0(T_0)}{\sigma_0(T)}, \text{ for } \omega_{\mathrm{p}} \leq \omega \geq 1/\tau \text{ (near IR and visible region)}, \tag{3}$$

where fee represents the free electron absorption, τ the electron relaxation time, ω

the optical frequency, ω_p the plasma frequency and σ_0 the dc conductivity.

The dc conductivity σ_0 is directly proportional to the square of plasma frequency, ω_p^2, and to the electron relaxation time, τ, by the relation [15]

$$\sigma_0 = \omega_p^2 \tau \,, \tag{4}$$

where the electron relaxation time τ arises from the combination of electron-phonon and electron-defect scattering processes. For many metals at 300 K, the electron relaxation time is in the range $10^{-14} \sim 10^{-15}$ sec and is temperature dependent. The Drude model [15] predicts that

$$\tau = 5.3 \times 10^{-16} \, \lambda \, \frac{k^2 - n^2}{2nk} \,, \tag{5}$$

where τ is the electron relaxation time, λ the laser wavelength and k and n are the extinction coefficient and refractive index, respectively. At the initial stage, the electron trapping phenomena are dominant, but with the increasing number of pulses, surface melting and resolidification dominates. The minimal energy per volume, E_m, for melting of Al is 26 J/mm^3 while the minimal energy for evaporation, E_v, for Al is 309 J/mm^3 [17], as derived from the equations

$$E_m = \rho \big[C \big(T_m - T_0 \big) + Q_m \big] \,, \tag{6}$$

$$E_v = \rho \big[C \big(T_v - T_0 \big) + Q_v \big] \,, \tag{7}$$

where E_m is the melting energy/volume, E_v the evaporation energy/volume, ρ the density of the material, C the specific heat capacity, T_m the melting temperature, T_v the evaporation temperature, T_0 the ambient temperature, Q_m the latent heat for melting and Q_v the latent heat for evaporation.

We find a larger change in the results in air than in vacuum both with the CO_2 and the Nd:YAG laser radiation. This difference might be due to the bombardment of Al by laser-induced plasma ions of air molecules (e.g., oxygen and nitrogen ions). The ion flux at 1000 mbar (in air) is much higher than the ion flux at 10^{-3} mbar (in vacuum). The high flux of ions transfers recoil energy to lattice atoms by the primary knock-on atoms (PKA) process. The PKAs displace further Al atoms from their lattice sites producing vacancies, interstitials (Frenkel pairs) and clusters. The resulting defects generated by air molecular ions alter the electronic characteristics of Al more prominently in air than in vacuum.

The observed decrease in electrical conductivity of Al with CO_2 laser radiation is more dominant both in air and vacuum than with Nd:YAG laser radiation, because CO_2 laser radiation is roughly 100 times more energetic than the Nd:YAG laser radiation, causing larger structural defects, e.g., by direct photon-phonon interaction with the generation of more energetic ions. For any metal, the absorption length determines the extension of the plasma along the beam propagation axis. The absorption length in Al for 10.6 μm is 12 nm while for 1.06 μm it is 10 nm

[16]. For a conducting surface, the IR radiation penetrating within the skin depth is absorbed by the free carriers with final conversion to heat in the material. Hence various theories of heat flow are applicable.

For a metallic surface with a constant absorption coefficient, Spark and Low [18] showed that the energy (E_m) required for rising the surface temperature from the ambient to the melting temperature is given by

$$E_\mathrm{m} = T_\mathrm{m} - T_0 \frac{\sqrt{\pi K C t_\mathrm{p}}}{2\alpha_0}\,, \tag{8}$$

where T_m is the melting temperature, T_0 the ambient temperature, K the thermal conductivity, C the specific heat, t_p the pulse duration and α_0 is the absorption coefficient. For CO_2 laser radiation, the single-pulse damage threshold, E_m for Al is calculated to be 0.58 J/cm^2 and for Nd:YAG laser radiation 48 mJ/cm^2. As the damage threshold is proportional to $t_\mathrm{p}^{1/2}$ and inversely proportional to α_0, the damage threshold for CO_2 laser radiation is higher than for Nd:YAG laser radiation. At room temperature, the absorption coefficient which is proportional to the electrical conductivity is higher for Nd:YAG laser radiation than for CO_2 laser radiation due to the smaller wavelength. But at elevated temperature, the absorption coefficient increases, resulting in nearly equal absorption coefficients of CO_2 and Nd:YAG laser radiation at the melting point of Al. Thus the only important parameter for the damage threshold at elevated temperature is the pulse duration. The heat penetration depth $L_\mathrm{p} = 2\sqrt{\chi} t_\mathrm{p}$ [19] in time t_p is numerically equal to 8.82 μm for CO_2 and 2.161 μm for Nd:YAG laser radiation. The mass of heated volume $m = L_\mathrm{p}\rho$ [19], is 23.902 gm/m^3 for CO_2 and 5.85 gm/m^3 for Nd:YAG laser radiation. The average temperature rise at the metallic surface of Al during irradiation is $\sim 10^6$ K both for CO_2 an for Nd:YAG laser radiation given by the relation [15]

$$T(0,t) = T_0 + 2\left(1 - \frac{R}{K}\right) I_0 \sqrt{\frac{\chi t_\mathrm{p}}{\pi}}, \tag{9}$$

where T_0 is the ambient temperature, R the reflectivity, K the thermal conductivity, I_0 the laser irradiance, χ the thermal diffusivity and t_p is the pulse duration.

For the sample in H_2 at pressures of 1000 and 130 mbar, the decrease in the electrical conductivity is larger at the higher pressure. It is due to the more efficient transfer of energy of plasma ions to the material. Al sample exposed to laser radiation in H_2 shows a larger change in its electrical conduction than in vacuum because of the ion implantation of H_2^+ ions. In vacuum the plasma plume undergoes free expansion, reaching finally the thermal equilibrium velocity, while the expansion is different in an ambient gas. This plasma plume consisting of ions, electrons, and other charged species moves like a piston, which accelerates the ambient atoms to supersonic speed, causing a shock wave ahead of the contact surface. The Al samples damaged in H_2 at 1000 mbar show a larger change of electrical conductivity than at 130 mbar, because at higher pressure, the larger density of atoms results in a more pronounced damage. In addition to that, for higher pressures the threshold intensity of laser radiation for gas breakdown is smaller than at lower pressures. The momentum transfer-collision frequency of electrons with neutrals is higher at higher pressures for cascade ionization. Therefore, the degree of ionization is enhanced at higher pressures for the same laser energy and damage is more pronounced.

4. Conclusions

The change in the conductivity of Al surface is observed after irradiation by CO_2 and Nd:YAG lasers in air, vacuum and hydrogen gas. Laser-induced plasma processes are responsible for the subsequent changes in the Al surface. The damage in air and in hydrogen is more pronounced than in vacuum. The change in the conductivity of Al with hydrogen plasma is pressure dependent. It is also observed that the electrical conductivity decreases with the increasing number of pulses. The relations of the dc conductivity to the reflectivity and the electron relaxation time are given. Some theoretical considerations are also taken in to account, e.g. the phonon speed in Al during photon interaction, the minimal melting and evaporation energy per volume, the damage threshold energy, the penetration depth, the mass of heated volume and the average temperature rise at the Al surface during laser irradiation.

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OŠTEĆENJA U ALUMINIJU PROIZVEDENA ZRAČENJEM IZ CO_2 I Nd:YAG LASERA

Proučavamo promjene električnih svojstava čistog aluminija (Al 99.999%) nakon obasjavanja CO_2 (energija = 2.5 J/puls, valna duljina = 10.6 μm, trajanje pulsa = 200 nsec) i Nd:YAG (energija = 10 mJ/puls, valna duljina = 1.06 μm, trajanje pulsa = 12 nsec) laserima. Uzorci su izloženi različitim brojevima pulseva. Proučavali smo promjene električne vodljivosti Al s uzorcima u zraku, vakuumu i u vodiku. Nakon obasjavanja mjerili smo električnu vodljivost metodom četiriju spojišta. Električna se vodljivost smanjuje nakon povećanog broja pulseva. Oštećenja u zraku i vodiku veća su nego u vakuumu, što se pripisuje sudarnom rasprašivanju Al ionima molekula zraka odnosno vodika u plazmi. Promjena vodljivosti uzoraka obasjanih u vodiku ovisna je o tlaku. Razmotrili smo neke teorijske rezultate, npr. fononsku brzinu u Al tijekom obasjavanja, minimalnu energiju taljenja i isparavanja po jedinici volumena, energijski prag oštećenja, dubinu prodiranja, masu zagrijanog volumena i prosječno povećanje temperature površine Al tijekom obasjavanja.