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Surface modifications of crystalline silicon created by high intensity 1064 nm picosecond Nd:YAG laser pulses

M.S. Trtica^{a,*}, B.M. Gakovic^a, D. Maravic^a, D. Batani^b, T. Desai^b, R. Redaelli^b

^a VINCA Institute of Nuclear Sciences, P.O. Box 522, 11001 Belgrade, Serbia

^b Universita degli Studi di Milano BICOCCA, Dipartimento di Fisica "G. Occhialini", Piazza della Scienza 3, 20126 Milano, Italy

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Abstract

A study of silicon modification induced by a high intensity picosecond Nd:YAG laser, emitting at 1064 nm, is presented. It is shown that laser intensities in the range of 5×10^{10} – 0.7×10^{12} W cm⁻² drastically modified the silicon surface. The main modifications and effects can be considered as the appearance of a crater, hydrodynamic/deposition features, plasma, etc. The highest intensity of $\sim 0.7 \times 10^{12}$ W cm⁻² leads to the burning through a 500 µm thick sample. At these intensities, the surface morphology exhibits the transpiring of the explosive boiling/phase explosion (EB) in the interaction area. The picosecond Nd:YAG laser-silicon interaction was typically accompanied by massive ejection of target material in the surrounding environment. The threshold for the explosive boiling/phase explosion (TEB) was estimated to be in the interval 1.0×10^{10} W cm⁻² < TEB $\leq 3.8 \times 10^{10}$ W cm⁻².

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1. Introduction

Surface modification studies of semi-conductors, including silicon, by various types of energetic beams including laser beams are of great fundamental and technological interest. Owing to its very good thermodynamic, physico-chemical and semi-conducting properties [1] silicon is attractive for numerous applications, e.g., in microelectronics, sensor technologies, biomedicine, etc. Interest for studies of laser beam interaction with silicon has been continuously high. The Nd:YAG [2–6], excimer [7,8], CO₂ (pulsed) [9], Ti:sapphire [10,11] are some of the lasers typically used for these purposes.

Studies of silicone interaction with Nd:YAG laser pulsed in picosecond time domain are insufficient in literature, especially for high laser intensities, in contrast to the nanosecond domain. The present work puts emphasis on morphological effects on silicon induced by a high intensity picosecond laser emitting at 1064 nm, the intensity varying from 5×10^{10} to 0.7×10^{12} W cm⁻².

2. Experimental

A single crystal silicon plate ($15 \text{ mm} \times 10 \text{ mm} \times 0.5 \text{ mm}$) with a (100) orientation was used in the experiment. The face side was polished, and the back was left as it is. The face roughness was evaluated by AFM and it was less than 10 nm. Prior to laser irradiation the sample was prepared using a standard procedure that includes cleaning, rinsing, etc.

Irradiations were performed in air at 1013 mbar, by a beam (near TEM₀₀ mode) focused through a quartz lens of 12.0 cm focal length, the incidence of the beam perpendicular to the sample surface. Spot size was estimated by a direct measurement of the beam trace at the target (after one laser pulse). The diameter measured was between 230 and 330 μ m, depending on the fluence used. The laser was an active–passive mode-locked Nd:YAG system [12]. Pulse duration of about 40 ± 2 ps was obtained by a saturable absorber dye and an acusto-optic standing wave modulator. During the experiments, pulse-to-pulse energy variation was $\leq 5\%$.

Various analytical techniques were used for characterization of the sample. Single crystal silicone orientation was confirmed by an X-ray diffractometer. The surface morphology was monitored by optical microscopy (OM), scanning electron

^{*} Corresponding author. Tel.: +381 11 2453 967; fax: +381 11 2447 207. *E-mail address:* etrtica@vin.bg.ac.yu (M.S. Trtica).

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microscope (SEM) and atomic force microscopy (AFM). The SEM was coupled to an energy dispersive analyzer (EDAX) for determining surface compositions of the targets. Profilometry was used for determination of crater properties.

3. Results and discussion

Laser induced silicon morphological changes/modifications have shown dependence on laser beam characteristics:

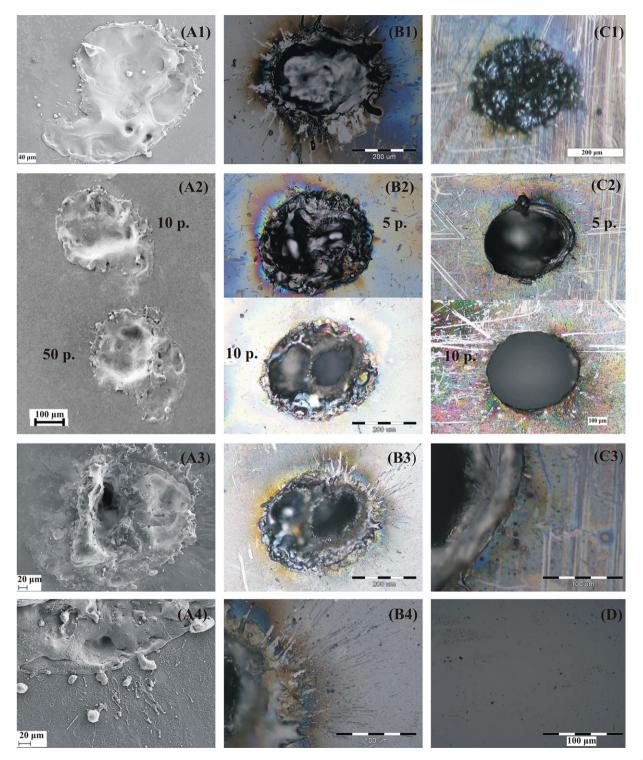


Fig. 1. Picosecond Nd:YAG laser-induced silicon morphology changes recorded by optical and scanning electron microscope. The laser was operating at 1064 nm; TEM₀₀ mode; intensity 5×10^{10} , 1.9×10^{11} and 0.7×10^{12} W cm⁻² (fluence 2, 7.6 and 27.4 J cm⁻²) for (A), (B) and (C), respectively. (A) SEM analysis; (A1), (A2), (A3) silicon after 1, (10, 50) and 100 laser pulses, respectively. (A4) Periphery of the damage area A3; (B) OM analysis; (B1), (B2), (B3) silicon after 1, (5, 10) and 100 laser pulses, respectively. (A4) Periphery of the damage area B3; (C) OM analysis; (C1), (C2) silicon after 1 and (5, 10) laser pulses, respectively. (C3) Periphery of the damage area C2; (D) The view of the silicon surface prior to laser action.

primarily on the energy density, beam intensity, pulse duration, number of accumulated pulses, laser wavelength, etc.

Surface morphology of the impact site at various laser radiation intensity (I) is presented in Fig. 1. The same figure (Fig. 1A-C) also represents the resulting morphology features as a function of the number of accumulated laser pulses, at a given intensity. It should be emphasized that all of the three intensities used, 5×10^{10} , 1.9×10^{11} and 0.7×10^{12} W cm⁻² (i.e., fluences 2, 7.6 and 27.4 J cm^{-2} ; Fig. 1) induced modifications of silicon. The following radiation intensity regimes can be distinctly recognized. (i) With a single pulse interaction, intensity of about $5 \times 10^{10} \text{ W cm}^{-2}$, the silicon morphology can be characterized by appearance of a crater (Fig. 1A1) with a relatively smooth bottom in the central part of the irradiated zone. The depth of the crater was about 2 µm. (ii) Similarly, for a single laser pulse at $I \sim 1.9 \times 10^{11} \text{ W cm}^{-2}$ a flowery structured crater was created, with inhomogeneity in the central part (Fig. 1B1). The crater depth was found to be about 20 µm. (iii) Finally, a single laser pulse of intensity 0.7×10^{12} W cm⁻² caused massive ejection of material which was accumulated at the periphery, Fig. 1C1. The central part of the damage was inhomogeneous.

Detailed evolution of the silicon morphology with the number of laser pulses (*N*) for laser intensity of 5×10^{10} W cm⁻² is presented in Fig. 1A. Increasing *N* at a constant intensity tended to radically modify the silicon surface. The morphology of the craters evolves from hemispherical (Fig. 1A1 and A2, only for 10 pulses) to complexnon-hemispherical shape after 10 pulses. The craters are characterized by a deep hole, approximately centrally located, with a surrounding thermally affected zone (Fig. 1A2, 50 pulses, A3). At the periphery of the irradiated area hydrodynamic effects, especially in the form of resolidified droplets, are visible (Fig. 1A3 and A4). In addition, with $N \ge 100$ pulses, the processes lead to the appearance of cracks at the periphery as shown in Fig. 2.

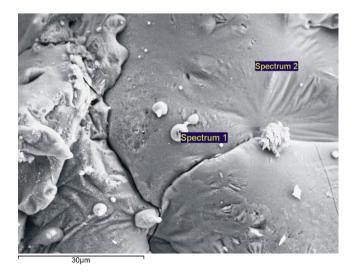


Fig. 2. SEM image of silicon surface at the near periphery, of the spot presented in Fig. 1 (A3) after 100 laser pulses (higher magnification). Spectrum 1 and spectrum 2 mark locations of EDAX analyses. Irradiation parameters: intensity 5×10^{10} W cm⁻²; TEM₀₀ mode; wavelength 1064 nm.

Silicon morphology evolution as a function of N for laser intensities 1.9×10^{11} and 0.7×10^{12} W cm⁻² is presented in Fig. 1B and C, respectively. The beam of $1.9 \times 10^{11} \text{ W cm}^{-2}$ produced a crater of non-hemispherical shapes for interval from 1 to 100 pulses (Fig. 1B). Hydrodynamic/deposition effects (Fig. 1B1-B4) are more expressed than in the case of the lower intensity $(5 \times 10^{10} \text{ W cm}^{-2})$, whereas even with one laser pulse existing droplets were larger than 10 µm in diameter (Fig. 1B1, B4). The processes leading to such morphology are of violent character. The highest laser intensity, $0.7 \times 10^{12} \,\mathrm{W \, cm^{-2}}$ (Fig. 1C), produced drastic morphological changes on silicon surface in the first pulse, and continued in the subsequent pulses. Accumulation of 5-10 laser pulses led to a complete burn-through in the 500 µm thick silicon sample (Fig. 1C2). The rim of the damage spot is relatively sharp in this case, with some sporadic deposition effects at the periphery (Fig. 1C3). Droplets deposited on the target were also relatively large (diameter > 10 μ m).

Target irradiation with laser intensities in the range 5×10^{10} to 0.7×10^{12} W cm⁻² were always accompanied by appearance of plasma in front of the target. The form of the plasma varied from torch with sporadic sparking (at 5×10^{10} and 1.9×10^{11} W cm⁻²) to continuous sparking (at 0.7×10^{12} W cm⁻²). In the latter case the sparks were large and it can be assumed that they carried parts of the target material. The process was of explosive character, with clearly visible material ejected to a distance of several centimetres in front of the target.

Surface oxidation during the irradiation of the silicon sample was quite possible, since the process was carried out in air. To confirm the oxidation process, EDAX compositional analyses of the central part and the periphery of the irradiated area were performed. No evidence of oxidation was found in central part of the irradiated area. Two locations at the periphery were investigated in Fig. 2, the top of a resolidified droplet and its vicinity. The EDAX analysis confirmed the presence of oxide in form of SiO₂ in both cases. Oxygen concentration was 8.48% at the top of the droplet and 15.00% in its vicinity (wt.%).

Generally speaking, the picosecond Nd: YAG laser interaction with silicon can be considered as absorption of photons primarily by electrons. The processes of one-, two- or multiplephoton absorption are quite probable. In addition, removal of material is possible by thermal and non-thermal processes. As the pulse duration in the present case was in the order of 40 ps, thermal effects must be taken into account. The excited electrons, further, give away energy creating phonons through electron-phonon relaxation. The energy is redistributed through lattice vibrations, producing heat that may melt the silicon and locally raise its temperature to produce vaporization [3]. Melting, vaporization and probably ejection of micron-sized droplets are common during this interaction. When laser intensity is above 1×10^9 W cm⁻² [3,5], it is possible to obtain heating of the sample over the boiling point so that a superheated metastable liquid layer can be produced. Material removal from the target, in this specific case, may be explained by homogeneous nucleation within the liquid layer. In that case the molten layer rapidly transforms into a mixture of liquid droplets and vapour, which "explode" from the heated surface

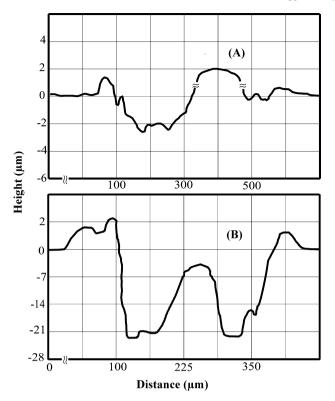


Fig. 3. Cross-sectional view of the craters formed after one laser pulse at intensities $5 \times 10^{10} \text{ W cm}^{-2}$ (A) and $1.9 \times 10^{11} \text{ W cm}^{-2}$ (B).

[3]. These conditions can be reached only in a special case, i.e., with a very short laser pulse and relatively high laser energy.

The morphological features obtained (e.g., special shape of the crater; appearance of large ejected droplets) as well as the accompanying phenomena at laser intensities $>5 \times 10^{10} \text{ W cm}^{-2}$, indicate that explosive boiling, often termed phase explosion, takes place. Although Martynyuk [13] has initially considered the process of explosive boiling/ phase explosion (EB), considerable effort in explaining of the phenomenon was made only recently [14,15]. One of the features that accompanies EB is apparently a special shape of the crater [3,5]. Recent reports by Yoo et al. [3] and Karnakis [5] showed that a Nd:YAG laser intensities $>2.2 \times 10^{10}$ or $1.15 \times 10^{10} \,\mathrm{W \, cm^{-2}}$, respectively, were sufficient to create a crater of this special shape. Karnakis reports transition from a conical to a semi-hyperboloidal shape for intensity $\geq 1.15 \times 10^{10} \,\mathrm{W \, cm^{-2}}$, whereas Yoo et al. report evolution from hemi- to non-hemispherical shape at intensities $\geq 2.2 \times 10^{10} \text{ W cm}^{-2}$. In the latter case the non-hemispherical shape was described as a deep hole near the center of the crater with thermally affected area surrounding the hole. Our experimental results showed that at the intensity of $1.9 \times 10^{11} \text{ W cm}^{-2}$ and even at $N \ge 1$ pulse a crater of nonhemispherical shape was obtained as in Fig. 3(B). This shape, similar to the one in reference [3], features a deep hole(s) near the center of the crater. Assuming that in the present experiment the intensity between 5.0×10^{10} and 1.9×10^{11} W cm⁻² was the minimum intensity that produces an explosive boiling/ phase explosion process on silicon, and taking that only 20% of the laser power is transmitted to the target [3], the threshold for EB can be estimated at $1.0 \times 10^{10} \text{ W cm}^{-2} < \text{TEB} \le 3.8 \times 10^{10} \text{ W cm}^{-2}$. Yoo et al. [3] and Karnakis [5] established the EB threshold at 4.4×10^9 and $1.15 \times 10^{10} \text{ W cm}^{-2}$, respectively, but for nanosecond Nd:YAG laser pulses (1064 nm and 355 nm) on silicon.

4. Conclusion

A study of silicon modification induced by high intensity a picosecond Nd:YAG laser emitting at 1064 nm is presented. It is shown that laser intensities of 5×10^{10} , 1.9×10^{11} and $0.7 \times 10^{12} \,\mathrm{W \, cm^{-2}}$ drastically modified silicon surface. The main modifications and effects can be considered to be appearance of crater, hydrodynamic/deposition features, plasma creation at the target, etc. The crater was typically of non-hemispherical shape, and large resolidified/deposited droplets were recorded at the periphery. The highest laser pulse intensity $0.7 \times 10^{12} \,\mathrm{W \, cm^{-2}}$ used in the present experiment led to the burn-through of the 500 µm thick sample even at the subsequent five pulses. The laser pulse intensities used, specific morphology features obtained, and the violent/explosive character of the processes indicate that explosive boiling/phase explosion phenomena were generated. The threshold for this phenomenon was estimated at $1.0 \times 10^{10} \text{ W cm}^{-2} < \text{TEB}^{-2} < 3.8 \times 10^{10} \text{ W cm}^{-2}$.

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