Formation of Conical Microstructures of Silicon with Picosecond Laser Pulses in Air

Nastaran Mansour[a], Kazem Jamshidi-Ghaleh[b] and David Ashkenasi[c]

[a]*Department of Physics, Shahid Beheshti University, Tehran, Iran*
E-mail address: n-mansour@cc.sbu.ac.ir
[b]*Department of Physics, Azarbijan Tarbiat Moallem University, Tabriz, Iran*
[c]*Department of Applied Laser Technology of the Laser and Medical Technology, GmbH, Berlin, Germany*

We report the development of conical microspikes that grew on silicon surface exposed in air to Nd: YAG picosecond (ps) pulsed laser irradiation. The formation of microstructures occurs at high number of laser pulses (~ 5000) at laser fluence of 1.3 - 1.5 J/cm^2. The microstructures are cones which grew toward the laser beam and surrounded by deep holes. The ps-formed microstructures are roughly 10 µm tall and separated by 5-10 µm in the center of the irradiation spot. Nanocolumns (or Nanodroplets) are formed on top of every cone of the laser-induced microstructures surface, reaching a height of ~1 µm and diameter of 100-250 nm. At higher applied laser fluence levels, no conical spikes formed and smooth surface is obtained due to high rate of thermal ablation. We will discuss the possible mechanism underlying these experimental observations.

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1. Introduction

Recently, considerable interest has arisen in laser-induced surface microstructuring of silicon due to its potential applications in optoelectronic [1-5]. Formation of sharp conical spikes on silicon surface has been observed [6-11] by repetitive nanosecond and femtosecond pulsed laser irradiation. The picosecond formed silicon conical microspikes in SF_6 ambient are reported in [12, 13]. Based on these experimental observations, it is proposed that the growth of conical spikes occurs in two stages: first the development of surface periodic shallow depression and then its evolution into a growing cone. It has been shown that the laser-induced initial depression continuously deepen with the number of laser pulses [8]. Applying more laser pulses to the same irradiated spot, the conical spikes are formed. The conical microstructures grow toward the applied laser beam and are surrounded by deep holes. In addition, in all the experimental results for the formation of silicon conical microstructure, a narrow range for the applied pulsed laser fluence is reported [6 - 13].

In this work, we present the experimental observations of laser-induced surface modification of silicon under the repetitive irradiation of 25 picosecond laser pulses in air at different applied laser fluence levels. We will show that the conical microstructures form on the surface after irradiating the silicon targets by cumulative laser pulses at fluence levels of very close to the melting threshold. However, at higher applied laser fluence levels, no conical spikes formed and smooth surface is obtained for the ablated regions. These experimental results may lead us to distinguish between two regime of irradiation in which conical microspikes formations occur or are suppressed. We will discuss on responsible mechanism for the laser-induced surface modification of silicon for picosecond pulses.

2. Experimental

The n-type (100) silicon wafers irradiated in air with a normally incident Nd: YAG laser beam. The laser generated 25 picosecond pulses at 1064 nm. The silicon wafers were mounted on a three-axis translation stage. The laser pulses were focused with a 25 cm focal length lens. The spatial profile of the laser pulse was Gaussian, with a 150 µm beam waist (FW1/e^{2}M) at the target. The silicon wafers were irradiated with laser fluence range of 1 - 4 J/cm^2 and the laser repetition rate was 1 kHz. The surface morphology of pulsed laser irradiated wafers is determined by Nomarski optical microscopy and scanning electron microscopy (SEM).

3. Results and discussion

Under exposure of silicon surface to 1000 laser pulses (τ_p = 25 ps, λ =1064 nm) in air at applied laser fluence level of about 1.3 J/cm2, a significant change in the surface morphology is observed (Fig.1). The surface morphology includes the formation of periodic surface depressions in the entire area of the irradiated spot. The separation between two consecutive depressions is measured about 3 µm. The laser-induced surface depressions develop into sharp conical spikes after irradiated by 5000 laser pulses [Fig.2]. We must mention that the depressions start to form only when plasma appears in front of the silicon targets. Figure 2 shows the SEM images of the center area of the laser-induced conical microstructures of silicon viewed at two different angles: (a) viewed at an angle 30 degree from the surface normal, (b) viewed perpendicular to the surface. The laser-induced

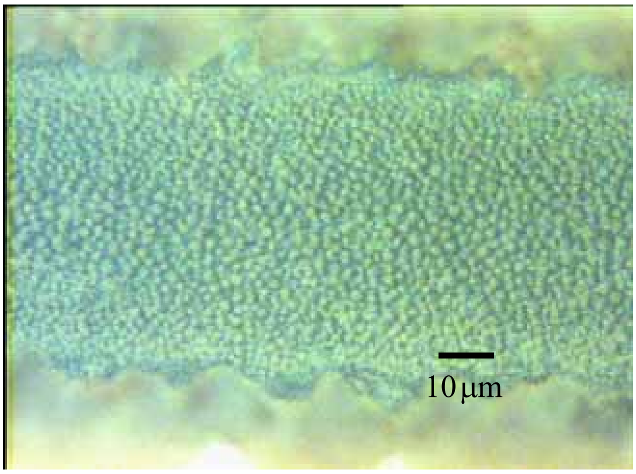
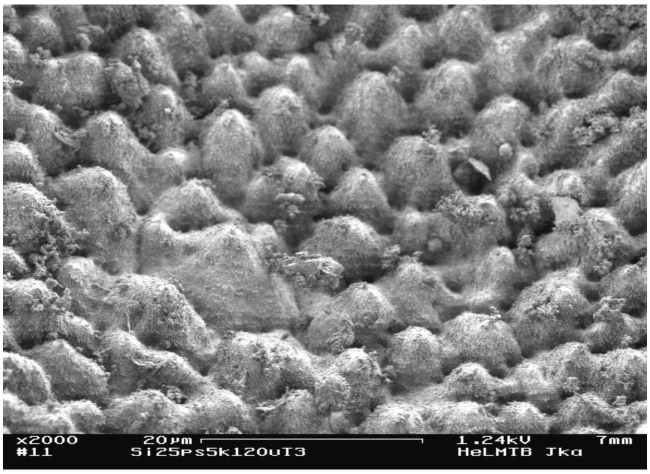
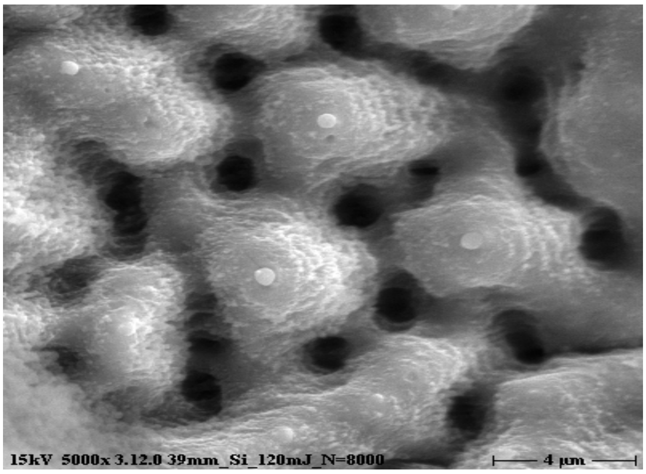


Fig.1. Nomarski micrograph of laser-induced periodic structures formed on silicon surface after 1000 laser pulses of 25 picosecond duration in air at applied laser fluence of 1.3 J/cm^2



(a)



(b)

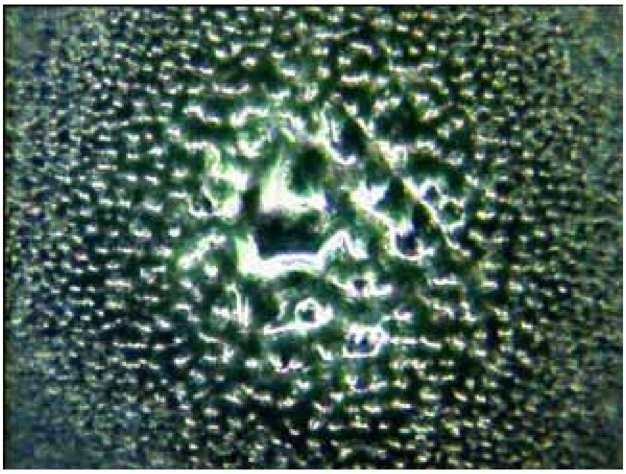
Fig.2. SEM images showing the center area of the laser-induced conical microstructures of silicon viewed at two different angles: (a) viewed at an angle 30 degree from the surface normal, (b) viewed perpendicular to the surface. The silicon microspikes formed after 5000 laser pulses of 25 picosecond duration in air at applied laser fluence of 1.3 J/cm2.

conical spikes are up to 10-13 µm high (Fig.4) and separated by about 5 - 8 µm in the center of the irradiated spot. The microstructures separations are reduced to 3 – 5 µm around the edge of the spot (Fig.2). This shows that the silicon microspikes separation is increased at higher applied laser fluence. As it is clearly shown in Fig. 2b every conical spike is surrounded by deep holes. Nanocolumns (or Nanodroplets) are formed on top of every cone of the laser-induced microstructures surface, reaching a height of ~1 µm and diameter of 100-250 nm (Fig.3). Similar experimental results for formation of the silicon conical microstructures using 35 picosecond cumulative laser irradiation at 0.8 J/cm^2 (8 kJ/m^2) in 500 torr SF_6 are reported in reference [12]. The reported silicon conical structures are up to 15 µm high and separated by 8-10 um using picosecond pulse laser irradiation at 1064 nm. It is also reported that no conical spikes formed in air for the applied laser fluence range of 0.6 - 1.2 J/cm^2 (6 to 12 kJ/m^2). Compare to our results, it seems that the applied fluence range is less than the threshold fluence required for the laser-induced formation of conical microspikes of silicon for picosecond pulses. In addition, the contribution of SF_6 only makes the cones grow longer.

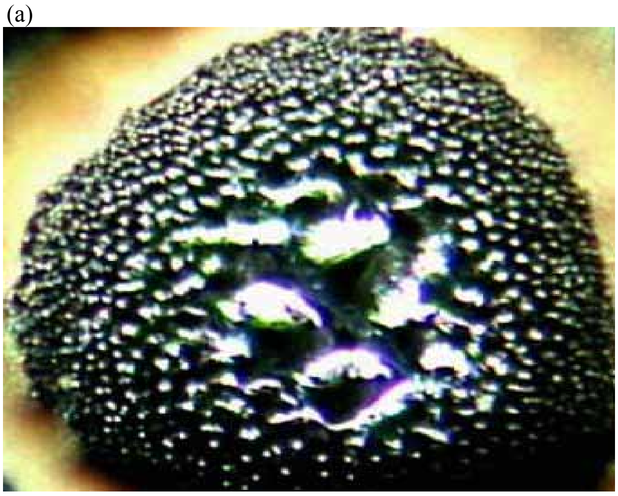
Fig.3. SEM image showing a nanocolumn formed on top of the conical microspike of silicon.

Figures 4(a) shows the morphology of the silicon surface irradiated with 1000 pulses at applied laser fluence level of 2 J/cm^2. The laser-induced surface structures at the center area of the irradiated spot are quite different from the structures formed on the edge of the site. At the center area, the surface structures are not ordered with no uniform structure spacing and are formed at lower level below the silicon surface. The ripple structures of the center area disappearing after irradiating the same spot with 1000 more laser pulses (Fig. 4(b)).

Figures 5(a) and 5(b) show the evolution of laser-induced modification of the silicon by cumulative irradiation at applied laser fluence of 3.3 J/cm^2. After irradiation with up to 1000 pulses, a hole is formed at the center area of the irradiated spot. Notice that the morphology of the ablated surface around the hole looks similar to the surface periodic structures formed at the lower applied fluence of 1.3 J/cm^2. This means that the trace of laser-induced modification of the silicon surface at lower applied laser fluence is similar to the edge trace at the higher applied laser fluence. The inside surface of the hole is roughly smooth after irradiating with more 1000 laser pulses (Fig. 5(b)).



(a)



(b)

Fig.4. Nomarski micrographs showing the evolution of the surface morphology of silicon produced with 25 picosecond duration at the applied laser fluences of 2 J/cm2 in air with the given number of pulses: (a) 1000, (b) 2000.

We have shown that the silicon microspikes form on the surface after irradiating the targets by cumulative 25 picosecond laser pulses at fluence range of 1.3-1.5 J/cm^2. By increasing the applied laser fluence to 2 J/cm^2, no conical spikes formed and smooth surface is obtained. It is to be noted that in our experimental condition, the silicon surface morphology starts to form when plasma appears in front of the samples. However, the plasma oscillation can not be responsible for the silicon microspikes formation. By increasing the applied laser fluence results in increasing the plasma density, and therefore the plasma frequency [14]. Thus, reduction of microspikes separation is expected in the presence of plasma oscillation at higher applied laser fluence. However, our experimental results show increase in the laser-induced microspikes separations at higher applied laser fluence. Therefore plasma oscillation is not responsible mechanism for the ps-formed microspikes of silicon.

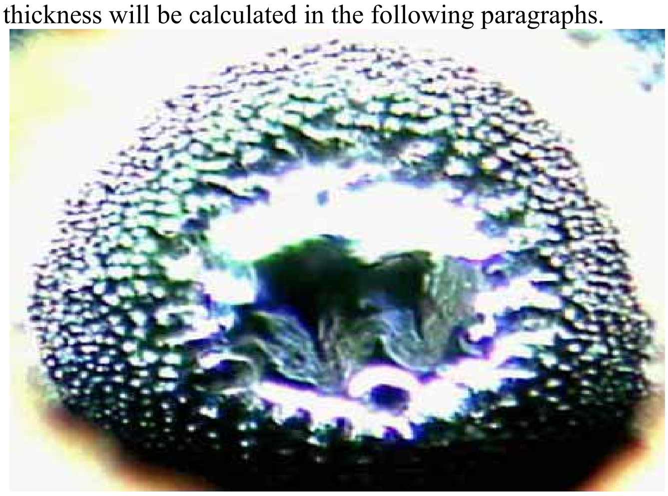
Dolgaev et al [9], demonstrated that the capillary waves can be responsible for the formation of the laser-induced surface microstructures. The spatial period of capillary waves in a shallow liquid layer is given by the following expression:

$$\lambda = \left[\frac{\sigma h}{\rho}\right]^{1/4} (2\pi\tau)^{1/2} \tag{1}$$

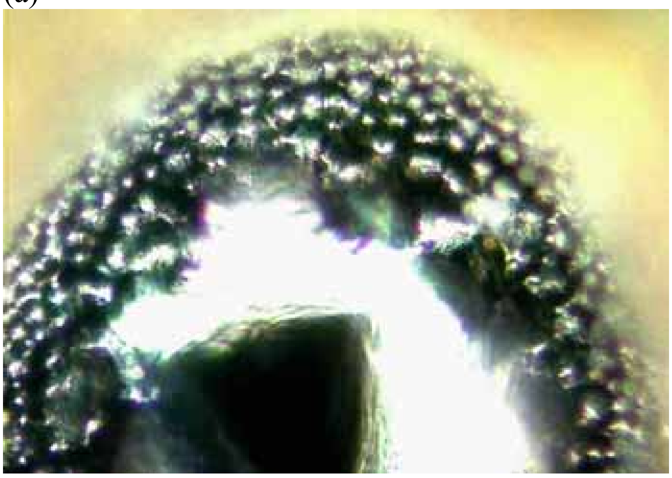
where λ is the wavelength of the capillary wave, τ is the period of the wave, σ, h and ρ are the surface tension coefficient, depth, and mass density of the liquid layer, respectively. Considering the fact that the capillary wave period can not exceed the life time of the liquid layer τ_L and therefore the average microstructures separations d, can be estimated by

$$d = \left[\frac{\sigma h}{\rho}\right]^{1/4} (2\pi\tau_L)^{1/2} \quad . \tag{2}$$

For estimating d, the silicon liquid life time and the melt thickness will be calculated in the following paragraphs.



(a)



(b)

Fig.5. Nomarski micrographs showing the evolution of laser-induced modification of the silicon by cumulative irradiation at applied laser fluence of 3.3 J/cm^2 in air with the given number of pulses: (a) 1000, (b) 2000.

Including the specific heat of melting, an energy density greater than 7000 J/cm^3 is needed to melt silicon at room temperature. The energy density U_s deposited at the surface of silicon is given by

$$U_s = \alpha(1 - R)F_0 \quad (3)$$

where α is the linear absorption coefficient, R is the reflectivity, and F_0 is the applied laser fluence. Using equation (1) and the silicon parameters given in table 1, the energy density deposited at the silicon surface for the applied laser fluence range of 1.3 – 1.5 J/cm^2 is calculated to be 44 – 53 J/cm^3. This means that the deposited energy density at the silicon surface associated with the applied laser fluence range of 1.3 - 1.5 J/cm^2 is not able to melt the surface. On the other hand, the experimental measurements of melting threshold fluence of silicon at 1064 nm for pulse duration of 6 – 225 ps are reported in [15]. The threshold is found to range from 0.6 ± 0.15 J/cm^2 for 6 ps pulses to 2.7 ± 0.3 J/cm^2 for 225 ps pulses. Using their experimental results, the melting threshold of silicon is obtained to be 1.3 - 1.4 J/cm^2 for 25 picosecond pulse duration. Two conclusions can be obtained from the reported experimental measurements of silicon melting threshold. First, compared to our results the silicon microspikes formation occurs at fluence levels close to the melting threshold for picosecond pulses. Then, the optical penetration depth (1/α) is not responsible for transferring energy from the pulse to the silicon lattice. The thermal diffusion length associated with free carriers contributes for transferring energy from the pulse to the lattice of silicon for ps pulses [15]. The diffusion depth δ, is estimated to be 350 nm using the experimental results for melting threshold versus the laser pulse width. Considering δ as the thermal penetration depth, the life time of the silicon liquid layer τ_l, is given by [16]

$$\tau_l = (2D)^{-1}\delta^2 \quad (4)$$

where D is the thermal diffusivity. Using the equation (2), the τ_l is calculated to be 650 ps.

We calculate h (the thickness of melt) from the expression given in [17] for molten layer thickness of silicon by

$$h = \frac{F_0(1 - R)}{c_v T_m + L_m} \quad (5)$$

where F_0 is the applied laser fluence, R is the reflectivity, T_m is the melting temperature, c_v is the specific heat capacity, and L_m is the specific heat of melting.

Using equations (2) and (5), h is calculated to be 1.2 μm and the silicon parameters given in Table 1, the microstructures separation is estimated to be 0.3 μm which is significantly lower than the observed silicon microstructures spacing of 3-8 μm. This analysis shows that the capillary waves on the molten silicon surface are not the responsible mechanism for the ps-formed microspikes of silicon.

Table 1. Silicon parameter used in calculation

Parameter	Value	Ref.
Absorption coefficient α, cm^{-1}	50	[18]
Reflectivity R	0.3	[18]
Liquid mass density ρ, g/cm^3	2.52	[18]
Specific heat of melting L_m, J/cm^3	4130	[18]
Specific heat capacity c_v, J/cm^{3}K	2	[17]
Surface tension coefficient σ, mN/m	850	[9]
Melting temperature T_m, K	1685	[18]

Recently, Emel'yanov and Babak [17], developed a model for pulsed laser formation of periodic surface structures on a semiconductor and demonstrated its feasibility for explaining the origin of fs-formed spikes of the silicon reported in [6]. They qualitatively explained the dependence of the applied laser fluence for formation of periodic surface structures. Their analyses show that the formation of structures is due to the rapid solidification of the melted layer accompanied by capture of point defects. At high applied fluence, the whole initially molten layer is evaporated per pulse and smooth ablated surface is obtained. It seems that the overall behaviors of the observed picosecond laser-induced surface morphologies of silicon are consistent with prediction of the model [17]. Further investigations are required to estimate the observed microspike spacings at applied fluence levels close to the melting threshold of silicon for picosecond pulses.

5. Conclusion

In conclusion, the picosecond laser-induced formation of conical microstructures of silicon has been studied at different applied laser fluence levels. At laser fluence of 1.3 -1.5 J/cm2, the conical spikes surrounded by very deep microholes are produced by cumulative pulsed laser irradiation of silicon samples in air. Our analysis shows that at fluence levels close to melting threshold of silicon, the experimental condition for the formation of conical microspikes are provided. At higher applied laser fluence levels, no conical spikes formed and smooth surface is obtained in irradiated region of the silicon targets. A qualitative discussion has been presented for the experimental observation of conical microstructure formation of silicon.

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