EARLY STAGES OF NANOSECOND PULSED-LASER GROWTH OF SILICON PILLARS IN VACUUM

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Dense arrays of high-aspect silicon microcolumns and microcones are formed by cumulative nanosecond pulsed excimer laser irradiation of single-crystal silicon in vacuum. The early stages of pulsed-laser growth of silicon microcolumns were analysed and compared as the number of KrF laser shots was increased from 1 to 100 shots. Scanning electron microscopy observation (SEM) shows that the beginning of micron growth occurs at about 1-5 shots irradiation by the appearance of small dome-like features. Additional laser shots leads to the formation of deep pits on either side of the dome and ring segments. In addition, it has been shown that the silicon microcolumns initially grow rapidly as the number of laser shots increases but growth appears to halt when they reach a certain length. Moreover, the phenomenon of the significant redeposition of ablated material observed at the tips and edges of the pillars was shown only for ranges of shots. This striking feature remains is discussed.

1. Introduction
Several studies of pulsed laser ablation process of silicon target by nanosecond or sub picosecond pulses have been performed in order to understand its surface modification mechanism by cumulative laser irradiation [1-3]. It has been shown that the silicon surface develop ripples with submicron periodicity when it is irradiated by nanosecond laser pulses with fluence close to the melting threshold [4]. Laser pulses with even higher fluence induce laser ablation of silicon with a crater on the surface surrounded by irregular cones protruding above the surface [5-6]. Most of the models used to explain the development of conical structures assume that the structures are formed by preferential removal of the material surrounding the cones [7]. In this paper, the early stages of pulsed-laser growth of silicon microcolumns were studied by performing a series of ablation experiments with increasing the number of KrF laser shots from 1 to 100 shots.

2. Experimental
The experiments were carried out in a high vacuum pumped to a base pressure of 10⁻⁶ mbar using a turbomolecular pump. The laser source was a KrF excimer (Lambda Physics compex 102) laser operating at 248 nm with pulse duration of 25 ns and a repetition rate of 10Hz. The Si rotating target was irradiated with 45 laser beam incident angle at different laser pulse. The emitted energy per pulse was fixed at 120 mJ. More details on the experimental were given elsewhere [8]. The typical number of pulsed delivered used in the experiments varies between 1 and 100. The rotation is 10 tr/min. The surface morphology of the ablated silicon surface is investigated using JEOL JSM 6360 LV Scanning Electron Microscope (SEM).

3. Results
The evolution of the columnar structures was studied by performing a series of ablation experiments with an increasing number of laser shots. Figure 1 shows the morphological changes induced on the Si surface after different laser shots from 1 to 50. SEM analyses show that after only the first laser shot (Fig.1a), the original flat surface is clearly modified by the formation of disordered cavities (pits or pores) that form a random labyrinth by coalescence and the location of domes within it with triangular or tetragonal-like base shape. Further laser irradiation to 5 shots (Fig.1b) revealed the appearance of new cavities and small domelike features. The later mark the beginning of microcone as reported by D.H. Lowndes et al. [9]. Additional laser shots to 10 shots induce the formation of deep cavities and pits on either side of dome. These observations have also been observed during sub-PS laser ablation of Si target [10]. The dimensions of the later (height and base) are increased as shown in Fig.1c. The increase of the laser shots to 20 shows that the laser irradiation produces conical spikes randomly positioned on the sample. In addition, it clearly appears that the cones are sharp and wear a clear spherical cap on top (Fig. 1d). An examination of Fig. 1d reveals significant redeposition of ablated material at the edges of the microcolumns. Pedraza et al. [6] revealed in their study a significant redeposition of ablated material near the edges and just outside of the laser irradiated region and...
suggested that a redeposition mechanism must be active during microcolumn growth. This redeposition phenomenon becomes more pronounced as the laser shots increase as shown in Fig.1e where a redeposition of the ablated matter at the tips of the pillars is also shown, which confirm the result presented by cited authors. This morphology is found similar with that observed by several authors for etching in SF₆ at 780-800 nm [11-12 and 9].

One can note that the pillars shown in Fig.1e average 43 µm height and 24 µm base width. The packing density is 74. 10³ cm⁻².

![Fig 1: SEM images of the morphological changes induced on the Si surface after different laser shots from 1 to 50, 1a) 1 shots, 1b) 5 shots, 1c) 10 shots, 1d) 20 shots, 1e) 30 shots.](image)

The height and base of the microcolumns have been measured by cleavage of the sample and examination in cross section. Fig. 2 depicts the variation of the average height of the microcolumns as a function of the number of shots from 1 to 50. It shows that the pillar height increases first slowly from 3 to 7 µm in the range 1 to 15 shots and rapidly above the later value to reach a value of 30 µm then tends to stabilize at a value of 60 µm for 50 shots.

![Fig 2: The variation of the average height of the microcolumns as a function of the number of shots from 1 to 50.](image)

Fig. 3 depicts the base width of the micocolumns as a function of the number of laser shots where it shows a same behaviour as observed for the height pillar variation. The base width increases first slowly from 7.4 to 9.3 µm, in the range 1-15 shots and rapidly to attain 24 µm for a number of laser shots of 30 shots.

These results clearly indicate that above 15-20 laser shots, the ablation mechanism of silicon is modified; the ablation and redeposition rate is enhanced.

![Fig 3: The base width of the micocolumns as a function of the number of laser shots.](image)

Fig. 4 plots a further geometrical parameter, the average separation between cones, as a function of the number of laser shots. The shape of the curve shows that the average separation distance decreases in two steps. The first step from 40 µm to 25 µm for 1 and 30 laser shots, respectively and the second one from 25 to 10 µm for 30 and 40 laser shots, respectively and then stabilizes for number of laser shots above 40.
The experiments carried out to reveal a succession of surface morphological changes as the number of laser shots is increased, indicate it is possible to identify the principal features of the mechanism that microcolumns are grown.

Based on the SEM observation of pillar evolution presented here it is clear that the formation of microcolumns occurs in two stages as reported by several authors [6, 13, and 14]. The first stage is the formation of microhole or pit which Pedraza et al. have shown necessarily precedes microcolumn formation. The presence of small domelike features actually marks the beginning of the microcolumns growth. As the number of laser shot increases, these pits or grooves become important which leads in turn upward growth of the dome to form a microcone. The later grows by redeposition of material that in ablated out of the adjacent etch pits. The silicon source for microcolumn growth is the laser-ablated silicon-rich vapour.

A.W.Bailey et al. [15] Stated that silicon-rich molecules are produced from the initially formed grooves or pits where etching is enhanced because the laser energy is concentrated as the beam undergoes multiple reflections from the steep walls.

In addition, based on the presented result (see Figs. 1 to 4) it is clear that only after a sufficient number of laser shot have succeeded in the creation of some initial features (in this work it corresponds to that produced by about 15-20 laser shots) then silicon microcolumns grow rapidly by a “catalyst-free” vapour-liquid-solid (VLS) growth mechanism. The later mechanism takes over and drives the subsequent growth of mature microcolumn as suggested by several authors [1,6].

4. Conclusion

The changes in silicon surface morphology were systematically analyzed and compared as the number of pulsed KrF (248nm) laser shots was varied for 1 to 100 in vacuum.

The results presented here provide interesting insight into the early surface evolution as well as the nucleation and subsequent growth mechanism of the microcolumns.

It has been shown that the nucleation of microcolumns is inhomogeneous; as the number of laser shots increase, new microcolumns initially grow slowly but growth increases rapidly when about 20 laser shots are reached. From SEM observation it is evident that the growth mechanism from microcolumns must involve redeposition of ablated silicon.

In addition, it is proposed, in agreement with several works, that growth occurs through a combination of pulsed-laser melting of the columns and walls and redeposition there of the intense flux of Si-rich vapour produced by ablation from especially grooves or pits.

5. References