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Effects of femtosecond laser pulse width on the formation of microstructured silicon

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We experimentally investigated the properties of surface microstructured silicon fabricated by 15 and 130 fs laser pulses. By changing parameters of femtosecond laser pulses, including laser flux, actual pulse acting time, and laser peak intensity, we found that the average height of spikes on the surface of microstructured silicon are only determined by the laser peak intensity. These results are important for the preparation and structure control of microstructured silicon.

OCIS codes: (140.3380) Laser materials; (140.3330) Laser damage; (160.6030) Silica; (160.0160) Materials.

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1. INTRODUCTION

Many microsized surface features have been observed on silicon surfaces after the irradiation of ion beams or laser pulses. It is found that femtosecond laser pulses can change the properties of silicon materials dramatically [1–3]. Mazur and co-workers previously found that femtosecond-laser-formed structures exhibit below-bandgap light absorption and photocurrent generation. These remarkable optoelectronic properties are attributed to sulfur impurities in a microcrystalline surface layer [4,5]. Importantly, the light absorption efficiency in a wide wavelength range (200–2500 nm) can be enhanced to more than 90%, which can be applied in solar cells and optoelectronic detectors [6–10] etc. Therefore, many efforts have been made to investigate the surface morphology and absorption property of microstructured silicon under different laser parameters and fabrication conditions, including laser fluence, laser polarization, laser wavelength, gas medium, and gas pressure [11–16]. However, no investigation focused on the relation between the surface morphology and the laser pulse width. Considering that laser pulses with different pulse widths have different interaction times with silicon materials, the surface microstructure fabricated by different pulse width lasers as well as their optical properties may be different.

In this paper, we focus our research on the microspike structure fabricated on silicon substrates by using laser pulses with different pulse widths. The average spike heights of microstructured silicon are compared in SF_6 ambient atmosphere. The relation between microstructures and laser parameters are discussed in detail.

2. EXPERIMENT SETUP

The corresponding experimental setup is shown in Fig. 1. Laser pulses were produced by a Ti:sapphire regenerative amplifier, typically 800 nm, 1 kHz, and 130 fs. The laser beam was focused into the hollow fiber tube (filled with 1 atm argon), where the frequency spectrum of the laser pulses was extended to 600–1000 nm. Finally, after compression of the chirp mirrors, laser pulses with a pulse width of 15 fs and a pulse energy of 0.6 mJ were obtained [17]. After being focused by a convex lens ($f = 100$ cm), the laser beam was delivered into the vacuum chamber through a 0.4 mm window. The vacuum chamber was fixed on a two-axis translation stage to realize the two-dimensional movement and backfilled with SF_6 (the base pressure was less than 10^{-4} Torr). The intensity of the incident laser beam was adjusted using a circular variable metallic neutral-density filter, and the pulse number was controlled by a beam shutter. The silicon wafer (n-type phosphor-doped, resistivity of 0.01–0.02 $\Omega \cdot$ cm, and crystal orientation of $\langle 100 \rangle$) was cleaned through deionized water, methanol, and acetone to remove the impurities and organic matter. Then it was mounted in the vacuum chamber with its face oriented vertically toward the direction of the laser beam. After irradiation, the microstructure image was observed using a scanning electron microscope (SEM). The heights of the spikes were measured from the SEM pictures, and a factor of $\sqrt{2}$ was multiplied because the sample was titled at a 45° angle for the measurement. Each value of the spike height was the average result of five points, and the measuring error was about 1 μm.

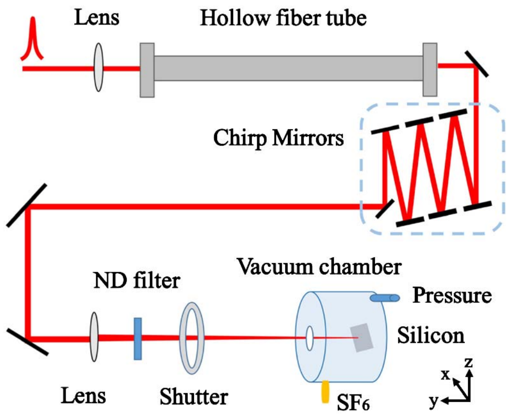

Fig. 1. Fabrication system of microstructured silicon.

3. EXPERIMENTAL RESULTS AND DISCUSSION

A. Under the Same Laser Flux

For the laser flux φ (energy per unit area) and the laser peak intensity P_0, their formulas can be written as

$$\varphi = P * t/S = P * m/(f * S), \quad (1)$$

$$P_0 = \varphi/\tau = (P * m)/(f * S * \tau), \quad (2)$$

where P is the laser average power, t is the laser lasting time, m is the irradiated pulse number, f is the laser repetition rate, S is the irradiated area of the laser beam on the silicon wafer surface, and τ is the pulse width. The laser lasting time (t) can be represented as $t = m/f$, which means the actual lasting time of the pulses. For example, if the pulse number is controlled by the beam shutter as 500 and the laser repetition rate is 1000, the laser lasting time t should be 0.5 s.

First, we want to make sure that the laser flux φ or the laser peak intensity P_0 determines the formation of the silicon surface microstructure. From Eqs. (1) and (2), we can see that the only difference between them is the pulse width τ. Therefore, we choose the following two groups of parameters to fabricate the microstructure silicon samples:

(G1) $\tau = 15$ fs, $P = 650$ mW, and $m = 200$ pulses versus $\tau = 130$ fs, $P = 650$ mW, and $m = 200$ pulses;
(G2) $\tau = 15$ fs, $P = 650$ mW, and $m = 500$ pulses versus $\tau = 130$ fs, $P = 650$ mW, and $m = 500$ pulses.

The surface morphologies of the fabricated microstructured silicon are presented in Fig. 2. We can see that although samples are fabricated under the same laser flux, the corresponding SEM images are completely different. The average spike heights of the microstructures fabricated by 15 fs laser pulses in Figs. 2(a) and 2(c) were 20.3 μm and 28.5 μm, respectively. The average spike heights of the microstructures fabricated by 130 fs laser pulse in Figs. 2(b) and 2(d) were 3.8 μm and 5.0 μm, respectively. Therefore, laser flux is not the determining factor for the size of laser-fabricated microstructures.

B. Under the Same Pulse Acting Time

The aforementioned expression $t = m/f$ gives the laser lasting time, while during the t range, the actual pulse acting time

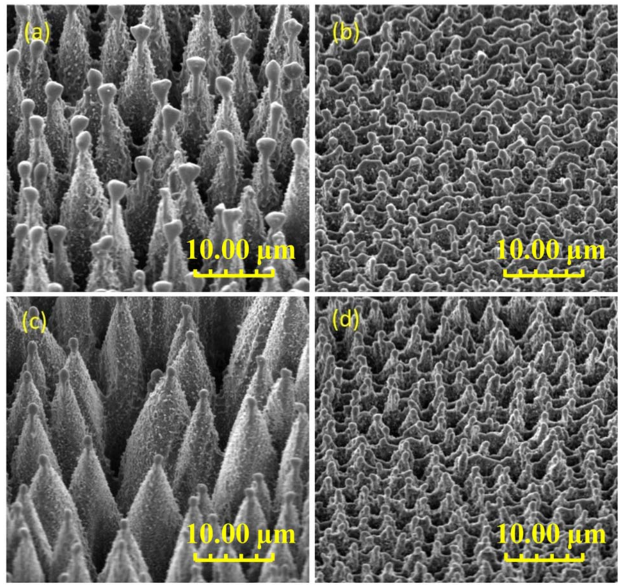

Fig. 2. SEM images of microstructured silicon fabricated by laser pulses with different pulse widths. (a) Pulse width, 15 fs; power, 650 mW; pulse number, 200. (b) Pulse width, 130 fs; power, 650 mW; pulse number, 200. (c) Pulse width, 15 fs; power, 650 mW; pulse number, 500. (d) Pulse width, 130 fs; power, 650 mW; pulse number, 500. Each SEM image is taken at a 45° angle to the surface normal.

upon a silicon wafer *should be* $t * f * \tau = m * \tau$. Therefore, the equation $m_{15\,\text{fs}} * \tau_{15\,\text{fs}} = m_{130\,\text{fs}} * \tau_{130\,\text{fs}}$ is valid when the actual pulse acting time is the same. We realize the same pulse acting time in two cases by changing the number of pulses via controlling the beam shutter. The pulse number of 15 fs is chosen as 130/15 times that of the 130 fs laser pulses, with all the other parameters the same:

(G3) $\tau = 15$ fs, $P = 650$ mW, and $m = 4330$ pulses versus $\tau = 130$ fs, $P = 650$ mW, and $m = 500$ pulses.

The corresponding SEM images are shown in Fig. 3. The microstructure fabricated by the 15 fs laser has a much larger scale than that of 130 fs, and a sunken hole is formed in the center of the silicon sample [see Fig. 3(a)]. The average height of the spikes of the microstructure fabricated by the 130 fs laser

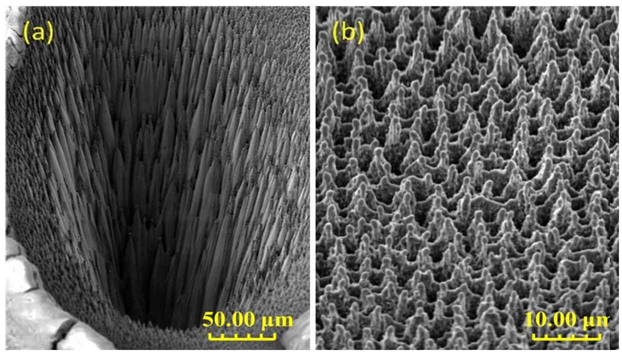

Fig. 3. SEM images under the same actual pulse acting time. (a) Pulse width, 15 fs; power, 650 mW; pulse number, 4330. (b) Pulse width, 130 fs; power, 650 mW; pulse number, 500.

is about 5.0 μm [see Fig. 3(b)]. Therefore, when the actual pulse acting time is the same, the sizes of microstructures fabricated by femtosecond lasers with different pulse widths are still not similar.

C. Under the Same Laser Peak Intensity

Based on Eq. (2), $\varphi_{130\,\text{fs}}/\tau_{130\,\text{fs}} = \varphi_{15\,\text{fs}}/\tau_{15\,\text{fs}}$ means their laser peak intensities are equal. Here, we choose the following two sets of parameters to fabricate the microstructure silicon samples:

(G4) $\tau = 15$ fs, $P = 580$ mW, and $m = 200$ pulses versus $\tau = 130$ fs, $P = 580$ mW, and $m = 1732$ pulses;
(G5) $\tau = 15$ fs, $P = 580$ mW, and $m = 300$ pulses versus $\tau = 130$ fs, $P = 580$ mW, and $m = 2598$ pulses.

As shown in Figs. 4(a) and 4(b), the average heights of the spikes fabricated by 15 fs and 130 fs laser pulses are 13.3 μm and 14.1 μm, respectively. Additionally, the shapes of microstructures are also similar to each other. In Figs. 4(c) and 4(d), when the pulse number of the 15 fs laser is 300 and the pulse number of the 130 fs laser is 2598, the average heights of the spikes are 13.4 μm and 13.8 μm, respectively. It seems that the laser peak intensity determines the size of the microstructures fabricated by femtosecond laser pulses.

To further prove the decisive role of the laser peak intensity for the size of the microstructures, a series of experiments under the same laser peak intensity were conducted. The laser power was set to as constant (580 mW), and the pulse number of 15 fs was 20, 50, 100, …500 (the interval is every 50 pulses). The pulse number of the 130 fs was always 130/15 times that of 15 fs. The average spike heights as functions of pulse numbers are shown in Fig. 5. We can see that the average heights of the spikes in two cases are always close to each other. Therefore, the laser peak intensity solely determines the size of the fabricated silicon microstructures.

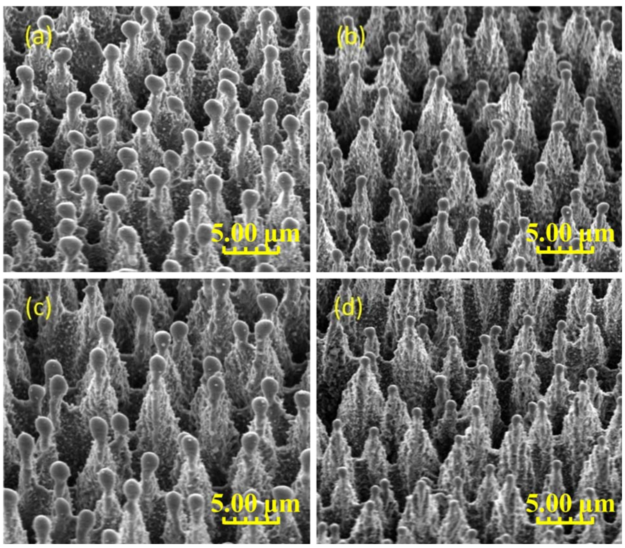

Fig. 4. SEM images upon the same laser peak intensity. (a) Pulse width, 15 fs; power, 580 mW; pulse number, 200. (b) Pulse width, 130 fs; power, 580 mW; pulse number, 1732. (c) Pulse width, 15 fs; power, 580 mW; pulse number, 300. (d) Pulse width, 130 fs; power, 580 mW; pulse number, 2598.

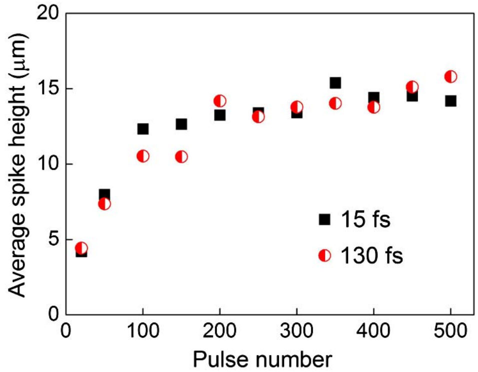

Fig. 5. Change of the average spike height as a function of the laser pulse number.

The above comparative experiments prove that the laser peak intensity has critical influence on the fabrication of the microstructure. The mechanism of the interaction between the laser and the silicon wafer is very complex, but the basic process is started from the excitation of electrons. The solid internal thermal process occurs after it absorbs energy from the laser. The electrons of the solid are emitted to the upper state by absorbing the photons. It takes 10^{-13} s(τ_{e-e}) to finish this process [18]. At this moment, the temperature of the electrons is higher than the temperature of the lattice, and the system is in the equilibrium state. Then, the system achieves balance after energy is released by electrons with high temperature in the excited state is transferred to the lattice through electron–phonon interaction, and this process takes 10^{-12} s(τ_{e-p}). Finally, it takes 10^{-11} s to transmit the energy absorbed by the solid surface into the solid inside.

For a femtosecond laser pulse with a pulse width of 130 fs (it is far less than τ_{e-p}), there is not enough time to establish the thermal balance between the excited electron and the lattice, i.e., the energy from the laser pulses cannot be transferred into the solid inside in time. As a result, a lot of energy accumulates on the silicon surface and then interacts with the surrounding material, which leads to the ablation and volatilization of the surface material. The narrower the laser pulse width, the higher the laser peak intensity, the more silicon material is ablated and volatilized, and finally the size of the microstructure is bigger. In addition, considering the spatial profile of the laser pulses is nearly Gaussian, i.e., the energy distribution at the beam center is the highest, and then the transverse energy distribution is well approximated by a Gaussian function. At the same time, we know that the higher the laser peak intensity, the higher the ablation and volatilization rate of the silicon material. Therefore, when the Gaussian-shape laser beam is incident on the silicon surface, the distribution of material ablation is also Gaussian shaped, i.e., the material in the center ablates quicker than the surrounding material. This induces a hole to be formed in the center of the irradiated area. At first, the depth of hole is too small to be observed. As the peak intensity of the irradiated pulse increases, the depth of the hole gradually increases and finally a sunken hole can be observed clearly. At this moment, the average height of the spikes cannot be measured exactly, again because the bottom of hole is too hard to distinguish. Therefore, the laser peak intensity represents the transient energy on the silicon surface, which determines the degree of silicon etching and becomes the only determining factor for the formation of microstructures.

4. CONCLUSION

We experimentally investigated the effect of pulse width of femtosecond laser pulses on microstructure fabrication. We found that under the same laser flux or the same actual pulse lasting time, the average height of the spikes fabricated by 15 fs laser pulses is much higher than that of 130 fs laser pulses. More importantly, our study reveals that the laser peak intensity has a determinative effect on the formation of silicon surface microstructures. These results are meaningful for the material fabrication of solar cells, sensors, photoelectron devices, and interrelated interdisciplinary subjects.

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