Differences in the evolution of surface-microstructured silicon fabricated by femtosecond laser pulses with different wavelength

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We experimentally investigate the differences in the evolution of surface-microstructured silicon fabricated by femtosecond laser pulses with different wavelength as a function of irradiated laser energy. The results show that when laser energy absorbed by the silicon material is the same, laser pulses with a shorter wavelength can form the surface-microstructured silicon with less laser energy, while the corresponding spike height is much lower than that of laser pulses with a longer wavelength. This is because the penetration depth of the laser pulses increases exponentially at the increase of the laser wavelength. Additionally, for two laser pulses with the certain wavelength and the certain absorption efficiency of silicon, the proportional relations between their formed spike height and irradiated laser energy should be determined. In particular, the average spike height is 3 times with 8 times corresponding energy for 800 nm laser pulses than that of 400 nm. These results are a benefit for the fast and optimum-morphology preparation of microstructured silicon. © 2012 Optical Society of America

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1. Introduction

Various microstructures can be formed on silicon surface by femtosecond laser pulses irradiating in ambient gas. Surface-microstructured silicon has remarkably enhanced light absorption in a wide wavelength range (0.2–2.5 μm) [1,2], which can be widely applied in solar cells [3], terahertz emission [4], sensors, and optoelectronic detectors [5–7]. It is found that remarkable absorption property is strongly related to the morphologies formed on the silicon surface. Therefore, much effort has been devoted to microstructured silicon fabrication with different surface morphology to obtain a larger photoelectric absorption coefficient. In past work, many experimental parameters, including laser fluence [8,9], pulse width [10,11], polarization [12], pulse number [13], gas medium [14], and gas pressure [15], have been adjusted to obtain different surface-microstructured silicon, and the corresponding results present different absorption properties. As laser pulses with different wavelength can form a distinct ripple pattern on the silicon surface in the initial stage and they have different penetration depth for the same silicon material, the fabricated surface microstructures should also be different. Nevertheless, the difference in the evolution of microstructures fabricated by femtosecond laser pulses with different wavelength remains unknown.

In this paper, we experimentally investigate the differences in the evolution of microstructures fabricated by femtosecond laser pulses with different wavelength as a function of irradiated laser energy.

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The results show that when laser energy absorbed by the silicon material is the same, laser pulses with a shorter wavelength can form surface-microstructured silicon with less laser energy, while the corresponding average spike height (the height of conical spike formed on the silicon surface) may be much lower than that of laser pulses with the longer wavelength. Additionally, for two laser pulses with the certain wavelength and the certain absorption efficiency of silicon, the proportional relations between their formed spike height and irradiated laser energy should be determined. All these come from the penetration depth of laser pulses increasing exponentially with the increasing of laser wavelength and the wavelength dependence of the absorption ability of silicon. These results are benefit for the fast and optimum-morphology preparation of microstructured silicon.

2. Theory

In order to investigate the relation between the wavelength of the incident laser and the microstructure morphology, we first analyze the process of microstructure formation [16]: when the laser beam irradiates on the silicon surface, the interference between the incident beam and light scattered by minor surface results in inhomogeneous energy deposition. When the incident energy exceeds both the ablation and melting thresholds for silicon, ablation and melt occur at nonuniform depth, creating capillary waves with the wavelength of the laser. At the increase of ablation and melting time, these capillary waves gradually become the ripple pattern, then a quasi-periodic array of beads. The further material ablation on the sides of the beads creates the conical structures with increasing height. During this process, we can find that the formation of conic spikes originates from the ablation and melt occurred in every small space between two ripple points. According to the energy density profile of the irradiated laser [16], each small space can be approximated to a cone. Then, the energy per unit volume of these cones, J_V, can be written as

$$J_V = \frac{J_\lambda \times A_\lambda}{\frac{1}{3}\pi R^2 d}, \tag{1}$$

where J_λ is the incident laser energy, whose value is proportional to the product of average laser power and incident pulse number [9]; A_λ is the absorption efficiency of a single crystal silicon for the laser beam; R is the radius of cone; and d is the penetration depth of energy, i.e., the height of spikes formed eventually. According to the analysis above, we can see that both R and d are determined by the wavelength of the incident laser. For two laser pulses with wavelengths of λ_1 and λ_2: $R_{\lambda 1}/R_{\lambda 2} = \lambda_1/\lambda_2$. The longer the laser wavelength is, the deeper the penetration distance, d [17]. Therefore, when laser energy absorbed by the silicon material ($J_\lambda \times A_\lambda$) is the same, the energy per unit volume is inversely related to the laser wavelength. In particular, for laser pulses with the shorter wavelength, its energy gathered in a smaller conical region, it is easy to reach the ablation and melting threshold and then form the microstructures easily and earlier. While, for laser pulses with the longer wavelength, their energy can go deeper into the silicon and then distribute in a larger conical region. Once the energy per unit volume reaches the ablation and melting threshold, the fabricated spikes can be much higher than that of laser pulses with the shorter wavelength.

On the other hand, the ablation and melting thresholds should be the same for microstructure formation with the same silicon material. Therefore, when the energy per unit volume is the same for two laser pulses with different wavelength ($J_{V\lambda 1} = J_{V\lambda 2}$) and it exceeds both the ablation and melting thresholds, then microstructures on the silicon surface can be formed simultaneously. Then, Eq. (1) can be written as

$$\frac{J_{\lambda_1} \times A_{\lambda 1}}{J_{\lambda_2} \times A_{\lambda 2}} = \frac{\lambda_1^2 \times d_1}{\lambda_2^2 \times d_2}. \tag{2}$$

This equation shows that without considering the other effects, there is a proportional relation between the irradiated laser energy and the formed spike height for the laser pulses with a certain wavelength and absorption efficiency. This determinate proportional relation can be used for the spike height prediction of microstructures fabricated by laser pulses with different wavelengths. For example, if we use 800 and 400 nm laser pulses to fabricate microstructures on silicon material, by substituting the parameters $A_{800} \approx 60\%$, $A_{400} \approx 40\%$ [8] and $R_{800} = 2R_{400}$, then Eq. (2) can be written as

$$\frac{J_{800}}{8J_{400}} \approx \frac{d_{800}}{3d_{400}}, \tag{3}$$

where J_{800} and J_{400} represent the incident energy of 800 and 400 nm laser pulses, d_{800} and d_{400} represent the penetration depth of 800 and 400 nm laser pulses, respectively. Eq. (3) proves that when the energy of 800 nm laser pulses is 8 times than that of 400 nm laser pulses, the average spike height of 800 nm laser pulses should be about 3 times than that of 400 nm laser pulses. In order to prove all these discussions above, we did the corresponding experiments as follows.

3. Experiment

The experimental setup is schematically shown in Fig. 1. In the experiments, we used a Ti:sapphire regenerative amplifier to produce 800 nm, 45 fs pulses at a 1 kHz repetition rate, and the spatial profile of laser spot was nearly Gaussian. The laser beam was focused with a convex lens ($f = 100$ cm) and delivered into the vacuum chamber through a 0.4 mm thick window. One flip mirror was used for two different light sources to separately interact with the sample: (i) when the flip mirror was flipped up,

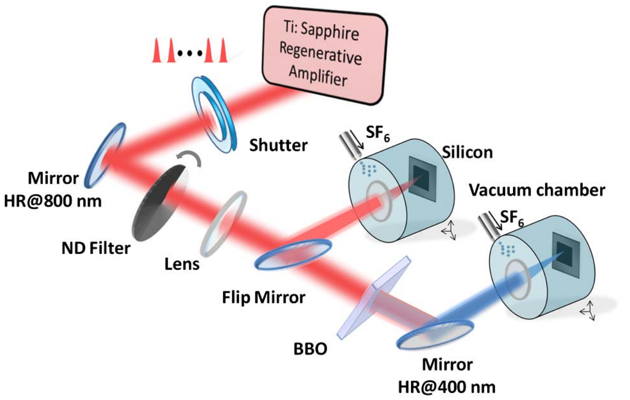

Fig. 1. (Color online) Experimental setup for the surface-microstructured silicon separately fabricated by the 800 and 400 nm laser beam. When the flip mirror was flipped up, the 800 nm laser beam directly interacted with the silicon, while when the flip mirror was flipped down, the 400 nm laser beam can be obtained and then interact with the silicon.

the 800 nm laser beam was directly incident to the sample and (ii) when the flip mirror was flipped down, the 800 nm laser beam was reflected into the 0.2 mm thick beta-barium borate (BBO) crystal (type I), and then the laser pulses with a center wavelength at 400 nm can be obtained. Additionally, we used several mirrors with high-reflection coating at 400 nm and antireflection coating at 800 nm to filter the residual 800 nm laser beam and then obtain the pure 400 nm laser beam. As the thickness of the BBO crystal was very thin, the pulse duration of 400 and 800 nm laser pulses can be considered as the same. The vacuum chamber (the base pressure was less than 10^{-4} Torr) was fixed on a three-axis translation stage to realize the three-dimensional movements. The (100) silicon wafer (n type with phosphor doped and resistivity between 0.01–0.02 Ω cm) fixed in the vacuum chamber was placed vertical to the incident direction of laser pulses and moved with the movement of the vacuum chamber. In addition, the silicon wafer was put before the laser focal spot in order to avoid the damage of high laser power to the window. Correspondingly, the laser spot on the sample surface was monitored by a CCD beam profiler (WinCamD-UCD12), and the diameter of each spot was set about 300 μm in the whole experiment by choosing the distance between the silicon wafer and the laser focus. Furthermore, in order to realize the intensity adjustment, we used a neutral density (ND) filter to provide linear, adjustable attenuation of injected light by rotation. In addition, the pulse number experienced by the sample was controlled by a beam shutter (SH05, Thorlabs). Finally, the fabricated microstructured silicon was analyzed by a scanning electron microscope (SEM, Tescan, VEGA II).

4. Experimental Results and Discussion

With ambient gases of SF_6 at the pressure of 500 Torr, microstructured silicon was fabricated by 800/400 cm laser pulses with different laser energy, in which the pulse number was fixed at 1000 for all the experiments and the detected laser power was changed. The corresponding average heights change of spikes formed on the silicon surface are shown in Fig. 2.

It can be seen that the 400 nm laser pulses can obtain the conical microstructures because the laser energy of 50 mJ [Fig. 3(b1)], and then the spike heights increase gradually with the increase of laser energy. After reaching a maximal height of ~15 μm at laser energy of 175 mJ [Fig. 3(b2)], the average spike height begins to decrease. As compared to it, the conical spikes fabricated by 800 nm laser pulses can only be effectively formed when the laser energy is close to 400 mJ and the maximal height is ~45 μm at the laser energy of 1400 mJ [Fig. 3(a2)]. Interestingly, when the energy of the 800 nm laser pulses is 8 times than that of the 400 nm laser pulses, the formed spike height of the 800 nm laser pulses is always about 3 times than that of 400 nm laser pulses, which is exactly consistent with the theoretical analysis mentioned above. Furthermore, according to the experimental data of 800 nm laser pulses, we also simulate the energy versus spike heights for the 400 nm laser pulses by decreasing 8 times energy and 3 times spike height. The corresponding results

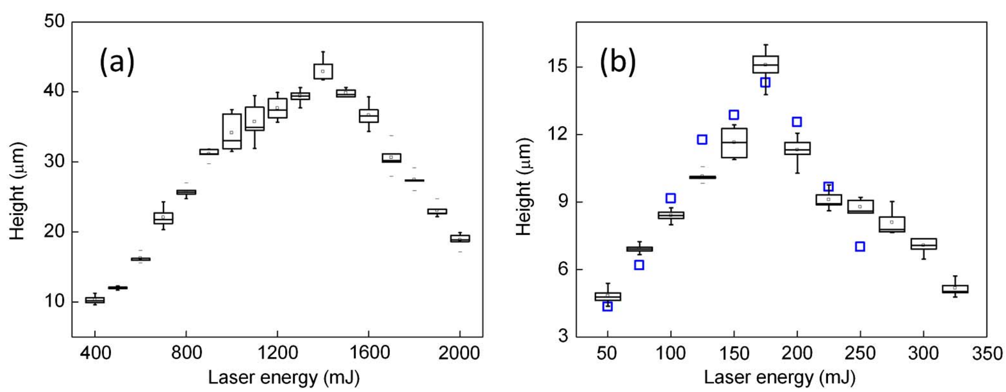

Fig. 2. (Color online) Average height of spikes fabricated by the (a) 800 and (b) 400 nm laser pulses as a function of laser energy. Dividing the energy and spike height by (a) 8 and 3, respectively, gives the calculated spike height as a function of the irradiated energy of 400 nm laser pulses [the blue squares in (b)].

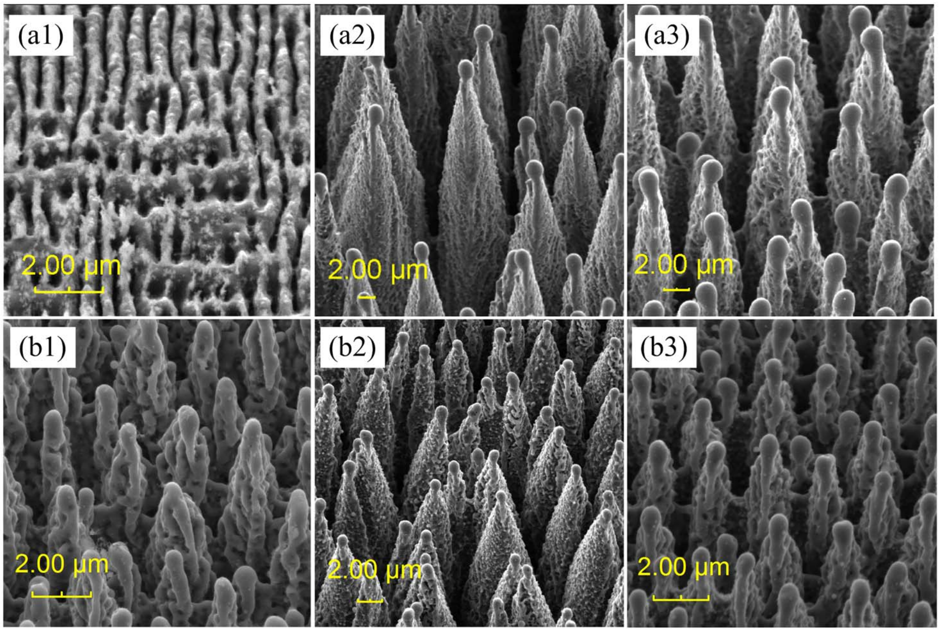

Fig. 3. (Color online) SEMs of surface-microstructured silicon fabricated by (a) 800 and (b) 400 nm laser pulses with the laser energy of (a1) 200, (a2) 1400, (a3) 2000, (b1) 50, (b2) 175, (b3) 325 mJ. The sample is viewed at 45° from the surface normal.

are shown in Fig. 2(b) with the square and dotted line. Therefore, all these phenomenon are consistent with our theoretical analysis.

Furthermore, Figs. 2(a) and 2(b) show that both microstructures fabricated by these two laser pulses have a similar variation of spike heights, i.e., an increase first and a decrease later at the increase of the laser energy. It has been known that the proper increasing laser energy can promote material ablation on the sides of the beads and then create conical structures with increasing height. When the irradiated laser energy is too large, the pulse number that determines the interaction time between the laser and silicon may not be enough for all the laser energy to transfer into the deep layer of silicon. This leads part of the energy to accumulate on the topmost layer. When the accumulated energy reaches a threshold and causes excessive ablation of the surface material, these beadlike structures are suppressed to be formed by initially hundreds of laser pulses in the early stage. Thus, the conical structures just can be established by the later laser pulses in the later stage, which finally leads to the decreases of the effective number of interaction laser pulses and then the average spike height. Overall, a turning point can be observed in the curve of the height variation of microstructures versus the laser energy.

5. Conclusion

In conclusion, we experimentally investigated the differences in the evolution of microstructures fabricated by femtosecond laser pulses with different wavelength as a function of irradiated laser energy. The results show that when laser energy absorbed by the silicon material is the same, laser pulses with a shorter wavelength can form the surface-microstructured silicon with less laser energy, while the corresponding spike height may be much lower than that of laser pulses with the longer wavelength. The reason is that the penetration depth of laser pulses increases exponentially with the increasing of the laser wavelength. Furthermore, for two laser pulses with the certain wavelength and absorption efficiency of silicon, the proportional relations between their formed spike height and irradiated laser energy should be determined. In particular, the average spike height is 3 times with 8 times corresponding energy for 800 nm laser pulses than that of 400 nm. These results are a benefit for the fast and optimum-morphology preparation of microstructured silicon.

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