Optimal proportional relation between laser power and pulse number for the fabrication of surface-microstructured silicon

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We experimentally demonstrate that, under the same laser fluence, there exists an optimal proportional relation between the laser power and pulse number for the fabrication of surface-microstructured silicon. During this fabrication process, the pulse number represents the interaction time between the laser and the silicon, which determines the depth of energy transferred into the inner part of the material, while the laser power determines the ablation and volatilization rate of the silicon. The proper combination of laser power and pulse number can ablate the material on the silicon surface effectively and have enough time to transfer the energy into the deep layer, which can produce microstructured silicon with a high spike. In addition, we compare the absorptance of samples etched by different combinations of laser power and pulse number; the corresponding results further prove the existence of an optimal proportional relation. © 2011 Optical Society of America

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1. Introduction

Single-crystal silicon irradiated by the femtosecond laser pulses in a gas medium can generate a new material, “black silicon” [1–3]. This material, with special microstructures, has high absorptance and photoelectric energy conversion efficiency in a wide wavelength range (0.2–$2.5\,\mu$m) [4], which can be widely applied in solar cells [5], terahertz emission [6], sensors, and optoelectronic detectors [7,8]. It is found that its absorption property is strongly dependent on the morphologies formed on the surface. Therefore, many experimental parameters have been adjusted to obtain different surface-microstructured silicon, including laser fluence [9], pulse width [10], polarization [11], pulse number [12], gas medium, and gas pressure [13]. We note that the laser fluence Φ (energy per unit area) can be written as $\Phi = (P{\cdot}t)/S = (P{\cdot}m)/(f{\cdot}S)$, where P is the laser power, t is the interaction time between the laser and the silicon, S is the irradiated area of the laser beam, m is the irradiated pulse number, and f is the laser repetition frequency. This formulation shows that laser fluence is proportional to the laser power and pulse number. Additionally, it has been known that both the laser power and pulse number can effectively affect the fabrication of surface-microstructured silicon [14]. However, whether the laser power or the pulse number mainly determines the surface morphologies and the absorption property of the microstructured silicon is still unexplored. Therefore, under the same laser fluence, the measurement and analysis of the morphologies formed on the silicon surface by using different combinations of laser power and pulse number are expected.

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In this paper, we investigated the effect of different combinations of laser power and pulse number on the formation of surface-microstructured silicon under the same laser fluence. The result shows that there exists an optimal proportional relation between the laser power and pulse number for the fabrication of surface-microstructured silicon, which can affect the surface morphology, absorptance, and photoelectric property dramatically.

2. Experiment

In the experiments, a Ti:sapphire regenerative amplifier produced the 800 nm, 45 fs pulses at a 1 kHz repetition rate. The laser beam was focused with a convex lens ($f = 100$ cm) and delivered into the vacuum chamber through a 0.4 mm thick silica window. The vacuum chamber, backfilled with SF_6 (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 (phosphor doped and n type with resistivity between 0.01–0.02 Ω cm) in the vacuum chamber was put vertical to the incident direction of the laser pulses, and the beam waist radius of each spot on the sample surface was set about 150 μm by choosing the distance between the wafer and the laser focus. Additionally, laser fluence of $2.8\,\mathrm{kJ/m^2}$ was used in our whole experiment, where the spatial profile of the laser pulses was nearly Gaussian. Furthermore, in order to realize the intensity adjustment, we used a circular variable metallic ND filter to provide linear and adjustable attenuation by rotation. In addition, the pulse number was controlled by a beam shutter (SH05, Thorlabs). After irradiation, the morphologies of the samples were observed and analyzed by using a scanning electron microscope (SEM).

3. Experimental Results and Discussion

At the pressure of 500 Torr, surface-microstructured silicon was fabricated by a set of different combinations of laser power and pulse number with the same laser fluence. The corresponding experimental results are shown in Fig. 1. Clearly we can see that, as the laser power increases synchronously but does not exceed 600 mW together with the decrease of the pulse number [see Figs. 1(a) and 1(b)], the spike height and the interval between spikes gradually increase and the droplet-shaped balls at the tips of the spikes become more and more clear and round. While the laser power is larger than 800 mW, one flat area begins to emerge in the central part of the irradiated region [see Fig. 1(c)]. When the laser power increases and the pulse number decreases further, the range of the flat area gradually enlarges, and then it reaches the maximum at the laser power of 1200 mW and the pulse number of 167 [see Fig. 1(e)]. Additionally, the spikes near the flat area are not standard cones, but they have some distortion in shape. Interestingly, as the laser power is larger than 1200 mW and the pulse number is less than 167 with the same laser fluence, the range of flat area gradually narrows and finally disappears at the laser power of 1800 mW and pulse number of 111 [see Fig. 1(h)].

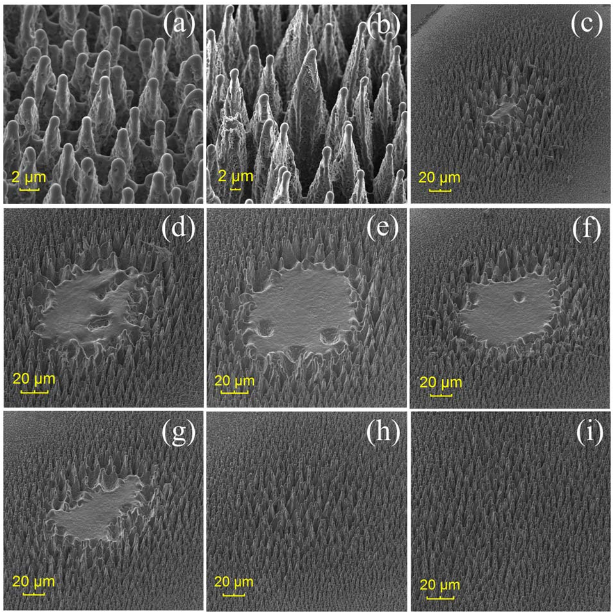

Fig. 1. (Color online) SEM photos of the surface-microstructured silicon produced by (a) 400 mW, 500 pulses; (b) 600 mW, 333 pulses; (c) 800 mW, 250 pulses; (d) 1000 mW, 200 pulses; (e) 1200 mW, 167 pulses; (f) 1400 mW, 143 pulses; (g) 1600 mW, 125 pulses; (h) 1800 mW, 111 pulses; (i) 2000 mW, 100 pulses. All pictures are viewed at an angle of 45° from the surface normal.

From Ref [14], we know that when the laser irradiates on the silicon surface and the energy of each pulse is absorbed by the sample, the energy density is greatest at the surface and then decreases for the deep layer. This density profile causes the topmost layer to be ablated away and the material beneath it to melt and then resolidify to form the microstructured surface. During this process, the pulse number represents the interaction time between the laser and the silicon, which determines the depth of energy transferred into the inner part of the material, while the laser power determines the ablation and volatilization degree of the surface material. We can see that only the proper ablation and enough interaction time can produce the microstructured silicon with a high spike. Therefore, under the same laser fluence, the phenomenon observed in our experiment can be understood from two aspects as follows: first, when the pulse number is large enough, the laser energy can be transferred into the deep layer of the silicon in a timely manner. Therefore, under the effect of laser ablation and volatilization of the surface material, spikes can be formed effectively. Additionally, at the increment of laser power, the spike height can increase synchronously [see Figs. 1(a) and 1(b)]; second, when the pulse number is not enough for all the energy transferring into the depth of the silicon, the energy partly accumulates at the surface. As the accumulated energy can still melt the material that is near the surface; then a flat area can be observed on the silicon surface [see Figs. 1(c)–1(g)]. While the pulse number decreases to a point that can only support a very small melt, the flat area cannot appear again [see Figs. 1(h) and 1(i)]. At the same time, the decreasing pulse number induces the decrease of spike heights.

According to the discussion above, we assume that for the experiments of Figs. 1(c)–1(g), if we only increase the pulse number while keeping the laser power unchanged, the energy gathered on the silicon surface can be transferred into the inner part of the material well. Then the flat area may disappear, and the formed spike height should be higher than the case of the lower pulse number with the same laser power. While the laser power is high and the pulse number is excessive, the material may be excessive ablated and then volatilized, which may form a big hole in the center of the irradiated region. In order to prove this, we did the corresponding experiments, and the results are shown in Fig. 2. As a comparison, taking Fig. 1(d) as example, when the laser power is the same while the pulse number increases to 1000, the flat area disappears completely [see Fig. 2(a)]. This directly proves that the pulse number affects the energy transfer depth, and then it determines the microstructured morphology formed on the silicon surface. On the other hand, when the pulse number increases to 4000, which is excessive for the maximum energy that can transfer into the inner part of the material, the high laser power leads to the ablation and volatilization of the surface material continuously, which finally forms a hole in the center of the irradiated area. Considering all these experimental results together, we can conclude that there exists an optimal proportional relation between laser power and pulse number for the fabrication of surface-microstructured silicon.

Additionally, in order to prove the effect of this proportional relation between laser power and pulse number on the absorption property of microstructured silicon, we measured the absorptance of samples etched by different combinations of laser power and pulse number. The measurements were performed with a Lambda 750 s UV/Vis/NIR spectrophotometer (PerkinElmer) equipped with an integral spherical detector that integrates all transmitted or reflected light. The absorptance ($A = 1 - R - T$) of samples is determined by the transmittance (T) and reflectance (R) [15]. The corresponding results are shown in Fig. 3. We can see that the different combinations of laser power and pulse number finally result in the different absorption property of samples. Furthermore, from our experiment, we also find the absorptivity is proportional to the height of the spikes, which can be controlled by using different combinations of laser power and pulse number. All these prove that under the same laser fluence, the laser power and pulse number have an optimal proportional relation for the fabrication of surface-microstructured silicon and its absorption property.

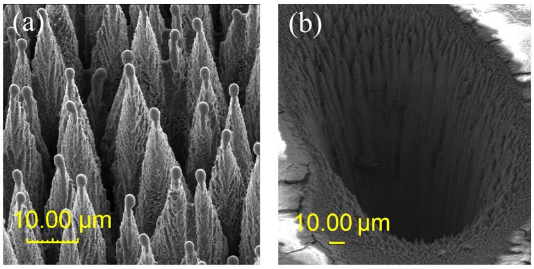

Fig. 2. (Color online) SEM photos of the surface-microstructured silicon produced by 1000 mW (a) 1000 and (b) 4000 pulses. The other parameters are the same as those in Fig. 1.

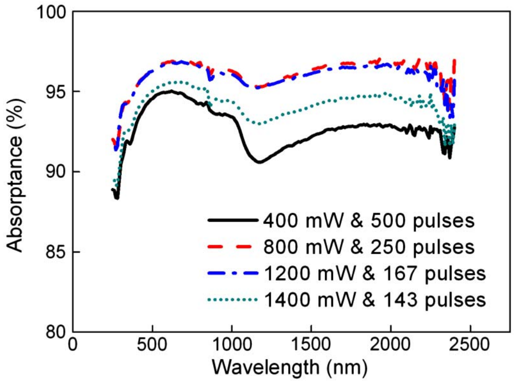

Fig. 3. (Color online) Absorption curves of surface-microstructured silicon etched by the femtosecond laser pulses with the parameters of 400 mW, 500 pulses (solid curve); 800 mW, 250 pulses (dashed curve); 1200 mW, 167 pulses (dashed–dotted curve); 1600 mW, 125 pulses (dotted curve).

4. Conclusion

In conclusion, we experimentally investigated the dependence of microstructured silicon fabrication on the proportional relation between laser power and pulse number. It is demonstrated that under the same laser fluence, there exists an optimal proportional relation between the laser power and the pulse number for the fabrication of surface-microstructured silicon. During this process, the pulse number represents the interaction time between the laser and silicon, which determines the depth of energy transferred into silicon, while the laser power determines the ablation and volatilization degree of the surface material. The proper combination can ablate the material on the silicon surface effectively and have enough time to pass the laser energy into the depth of the silicon, which can produce microstructured silicon with a high spike. In addition, we compared the absorptance of samples etched by different combinations of laser power and pulse number; the corresponding results further prove the existence of this optimal proportional relation.

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