

PAPER • OPEN ACCESS

Femtosecond Laser Double Pulses Nanofabrication on Silicon

To cite this article: Jin Zhang *et al* 2019 *IOP Conf. Ser.: Mater. Sci. Eng.* **565** 012018

View the [article online](#) for updates and enhancements.



IOP | ebooks™

Bringing you innovative digital publishing with leading voices to create your essential collection of books in STEM research.

Start exploring the **collection** - download the first chapter of every title for free.

Femtosecond Laser Double Pulses Nanofabrication on Silicon

Jin Zhang^{1*}, Sumei Wang¹, Mengmeng Wang¹ and Zhuyuan Chu¹

¹Laser Micro/Nano Fabrication Laboratory, School of Mechanical Engineering,
Beijing Institute of Technology, Beijing 100081, P.R. China

*Email: zj_1995@126.com

Abstract. The temporal shaping femtosecond (fs) laser is realized by dividing a fs laser pulse into two identical sub-pulses. The double-pulse fs laser can be used to effectively control the initial free electron state such as electron temperature, capacity and density, etc. to improve the surface morphology quality. In this experiment, silicon was fabricated using the double-pulse fs laser. It was found that the average diameter of ablated micro- or nanoholes decreases with the increasing pulse delay (up to 1.5 ps) under the same laser fluence and the phenomenon was explained quantitatively by the plasma model. Furthermore, nanoholes were achieved using the double-pulse fs laser, which cannot be obtained by single pulse laser, and the processing size can be reduced to below 200 nm.

1. Introduction

Depending on unique processing advantages compared to other special processing techniques such as electron beam machining [1], focused ion beam machining [2], and photolithography [3], etc. femtosecond (fs) laser beam machining is an irreplaceable technique in the micro- and nanofabrication field. Due to ultrahigh peak power and ultrashort pulse duration, fs laser processing can be regarded as a ‘cold’ processing. Meanwhile, relying on its non-linear effect [4], fs laser can process any difficult-to-machine material including metal, dielectric and semiconductor, etc. In addition, it is simple to operate, low-cost and can realize three-dimension processing. Because of ultrashort pulse duration of fs laser, only the free electron states such as free electron capacity, temperature and density, etc. can be affected during the laser irradiation [5]. The double-pulse fs laser can control these properties of free electron by adjusting the laser pulse delay [6]. Furthermore, the subsequent physical processes can be affected and the processing quality can be optimized.

Fs laser temporal shaping technique can be applied in various fields of laser processing. In terms of laser modification [7], the absorption rate of fs laser can be improved using the double-pulse fs laser under the same laser fluence. Then the increased absorption rate leads to the formation of more 3-membered ring structures inside fused silica. Based on more 3-membered ring structures, laser-irradiated fused silica is easier to be etched by HF acid and the etching efficiency is greatly improved. In the laser ablation field, the fs laser pulse sequence can be achieved through pulse shaper, and the ionization process of material is changed by the fs laser pulse sequence. Using the fs laser pulse sequence, a high depth-diameter ratio microhole can be processed on the sapphire substrate, which cannot be achieved by an ordinary Gaussian-like single pulse [8].

In this paper, the temporal shaping [9] was realized by dividing a fs Gaussian-like single pulse into two identical sub-pulses with the same fluence. As a semiconductor, silicon is widely applied in the electronic information field. The double-pulse fs laser was used to process the crystal <111> silicon in



this experiment. By changing the pulse delay to control laser-material interaction process, the surface morphology of silicon can be optimized. Compared to the single pulse, it was found that the average diameter of microholes processed by the double pulses is smaller than the single pulse and the diameter became smaller with the increasing pulse delay under the same laser fluence. Using the critical free electron density as the laser ablation criterion [10], the change of laser ablation threshold was predicted by the plasma model [5]. Furthermore, we found that double pulses can achieve more precise machining which cannot be completed under the single pulse, and the diameter of holes can be reduced to below 200 nm.

2. Experimental section

A commercial Spectra Physics Spitfire laser with a central wavelength of 800 nm and a pulse delay of 35 fs was used to ablate the crystal <111> silicon. A single pulse fs laser was divided into two identical sub-pulses with an energy ratio of 1:1 through the beam splitter. The optical path difference of two sub-pulses was adjusted to control the time delay of reaching the silicon surface through the electronic translation stage. The temporal shaping was achieved using the double-pulse fs laser. In this experiment, a single shot pulse was used to ablate these holes on a silicon substrate. The laser was focused by a 10X objective lens. Further, in order to make the hole diameter as small as possible, a 50X objective lens was used to generate the tightly focused fs laser pulse to explore the minimum size. The processing results were observed by cold field scanning electron microscopy (SEM) for detailed data analysis.

3. Results & Discussion

3.1. Double Pulses Nanofabrication

Firstly, a fs laser single pulse was used to ablate the microhole structure on silicon. In order to compare the difference between the double pulses and single pulse, the pulse delay was changed from 0 fs to 2000 fs under the same laser fluence. As shown in Figure 1, microholes were respectively fabricated at the laser fluences of 0.52 J/cm^2 and 0.35 J/cm^2 to ensure the good repeatability. Figure 1a and Figure 1b show the change of surface morphologies at the laser fluence of 0.52 J/cm^2 , and the black area in the center of microholes means that the microholes were ablated. However, as shown in Figure 1c and Figure 1d, the white area in the center of microholes means the formation of bump. The laser irradiated area was modified and the laser fluence of 0.35 J/cm^2 was under the ablation threshold. The formation mechanism of bump was attributed to the fact that the liquid density (2.52 g/cm^3) of silicon is larger than the solid density (2.32 g/cm^3).

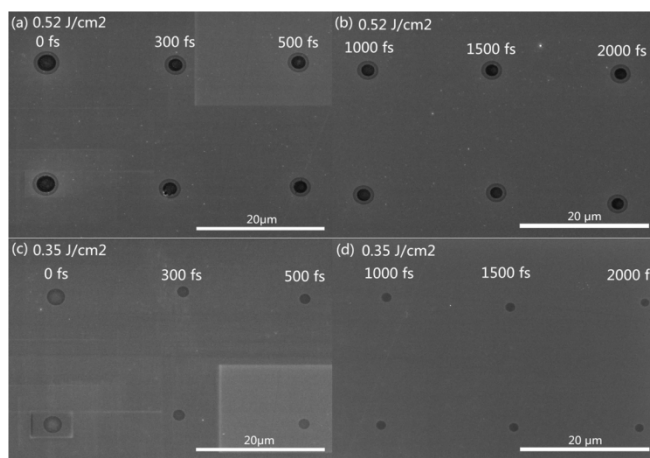


Figure 1. SEM images of silicon surfaces exposed by (a-b) a fs pulse at a fluence of 0.52 J/cm^2 with pulse delays of 0, 300, 500, 1000, 1500 and 2000 fs, respectively, and (c-d) a fs pulse at a fluence of 0.35 J/cm^2 with pulse delays of 0, 300, 500, 1000, 1500 and 2000 fs, respectively. The experimental parameters of holes in each column are uniform.

By counting the diameters of microholes, it was found that the microhole became smaller with the increasing pulse delay and the diameter gradually stabilized as the pulse delay is up to about 1500 fs. Figure 2 shows the trend change of hole diameter as a function of pulse delay.

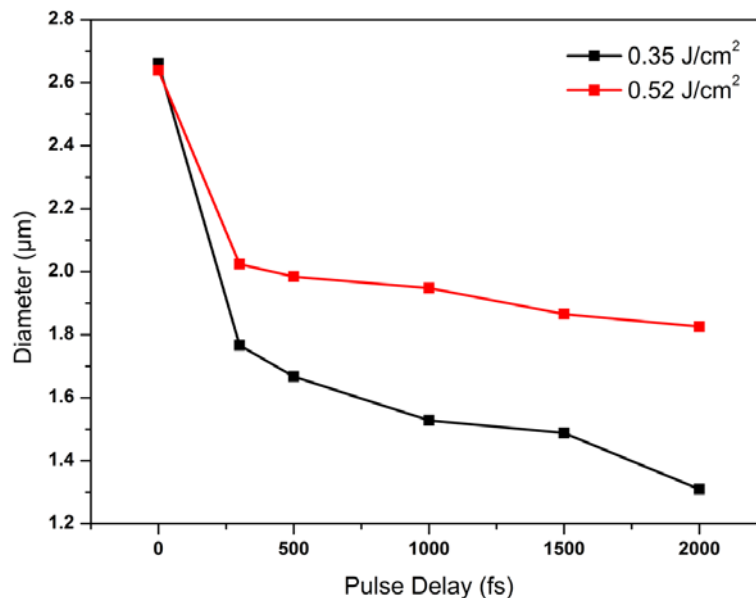


Figure 2. Average diameter of microholes as a function of pulse delay at the double-pulse threshold fluence of 0.52 J/cm² and 0.35 J/cm², respectively.

3.2. Plasma Model Theory

This phenomenon that the hole diameter decreased with the increasing pulse delay under the same laser fluence is also found in the fs laser ablation of fused silica [6]. Compared to the insulator, the properties of semiconductor silicon are similar to fused silica during the process of laser interaction. Therefore, the plasma model can be utilized to describe the laser-silicon interaction. The direct cause of this phenomenon is that the increase of pulse delay lead to the increase of the ablation threshold of silicon. Due to higher ablation threshold of large pulse delay, the diameter of microhole is smaller than the single pulse under the same laser fluence. Further, depending on the critical electron density ablation criterion, the ablation threshold of silicon can be calculated by the plasma model. When the fs laser double pulse irradiates on the silicon surface, the first sub-pulse excites a large amount of free electrons from the valence band of silicon by photoionization, and the free electron density cannot remain constant during the time interval between two sub-pulses. During the time interval, the recombination of free electron occurs due to direct radiation recombination, Auger recombination, and diffusion of free electron [11]. The free electron density decreases through the recombination process. During the second sub-pulse irradiation, these free electrons act as seed electrons to excite more free electrons through impact ionization, and large amounts of free electrons also are excited through photoionization. Therefore, as the pulse delay increases (up to 1500 fs), the free electrons ionized by the first sub-pulse will recombine more so that the total free electron density naturally become less. According to the critical free electron density ablation criterion, the ablation threshold will become larger.

Furthermore, the plasma model was used to simulate the relationship between the ablation threshold of silicon and pulse delay of double pulses quantitatively. Since silicon is a semiconductor, the valence band is only 1.12 eV [12], which is lower than the 1.5 eV of a single photon energy. This means that the ionization is mainly generated by single photon and two-photon absorption [13], and impact ionization can be ignored under the low laser fluence [14]. Same with fused silica, the

recombination term of free electron density can be simplified by $-\frac{n_e}{\tau}$ [15]. The typical equations for calculating the free electron density after the laser irradiation on silicon are given by:

$$\frac{\partial I}{\partial z} = -\alpha_0 I - \beta I^2 - \alpha_{fca} I \quad (1)$$

$$\frac{\partial n_e}{\partial t} = \frac{\alpha_0 I}{\hbar \omega_L} + \frac{\beta I^2}{2\hbar \omega_L} - \frac{n_e}{\tau} \quad (2)$$

where I means the initial optical intensity, z is the spatial coordinate perpendicular to the silicon surface, α_0 and β are single photon and two-photon absorption coefficient, α_{fca} refers to the absorption rate of free electron, n_e is the free electron density and τ describes the relaxation time of free electron, which is about 200 fs for silicon [15].

Different pulse delay means different optical intensity distribution in the time scale. Through the iteration of equation (1) and equation (2), we can calculate the relationship between pulse delay and ablation threshold, which fits well with the experimental data, as shown in Figure 3. The ablation threshold of silicon becomes larger and finally stabilizes with the increasing pulse delay from 0 fs to 2000 fs. Therefore, the average diameter of holes naturally becomes smaller.

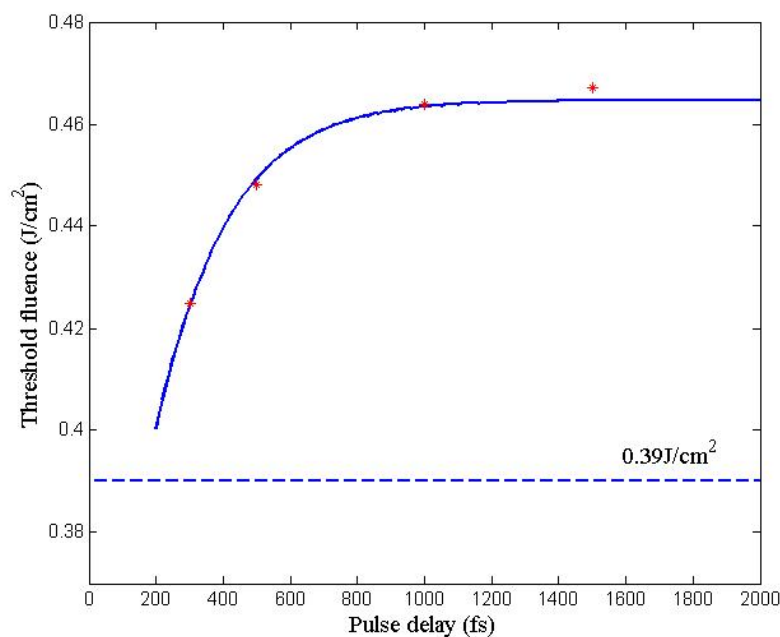


Figure 3. The relationship between ablation threshold and pulse delay is stimulated through the plasma model, and the dot line means the ablation threshold under the single pulse. The asterisks represent the ablation thresholds under different pulse delay.

3.3. Exploration of Minimum Size

In order to ablate enough small hole on silicon using fs laser, a 50X objective lens was used to focus tightly fs laser to obtain waist diameter of 1.57 μm , which is calculated using a method proposed by Liu [16]. By continuously reducing the laser fluence, a hole with diameter 400 nm was processed at laser fluence of 0.52 J/cm^2 under the single pulse, which is larger than the ablation threshold fluence under single pulse, as shown in Figure 4. The smallest hole was processed under single pulse and

silicon cannot be ablated under double pulses at the same laser fluence, which proved the ablation threshold under double pulses was larger than that under single pulse. Using the double pulse, it can be found that the diameter of nanohole was about 325 nm at the laser fluence of 0.62 J/cm^2 and pulse delay of 2000 fs (Figure 5a). A nanohole with diameter of about 200 nm was processed at the laser fluence of 0.57 J/cm^2 and pulse delay of 300 fs, as shown in Figure 5b, which is the smallest hole at present and cannot be achieved under the single pulse.

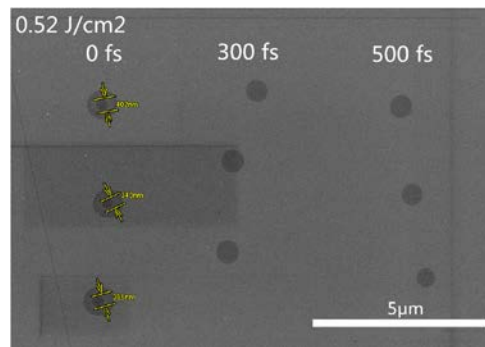


Figure 4. SEM images of silicon surfaces exposed by a fs pulse at a fluence of 0.52 J/cm^2 with pulse delays of 0, 300, and 500 fs, respectively. The experimental parameters of holes in each column are uniform.

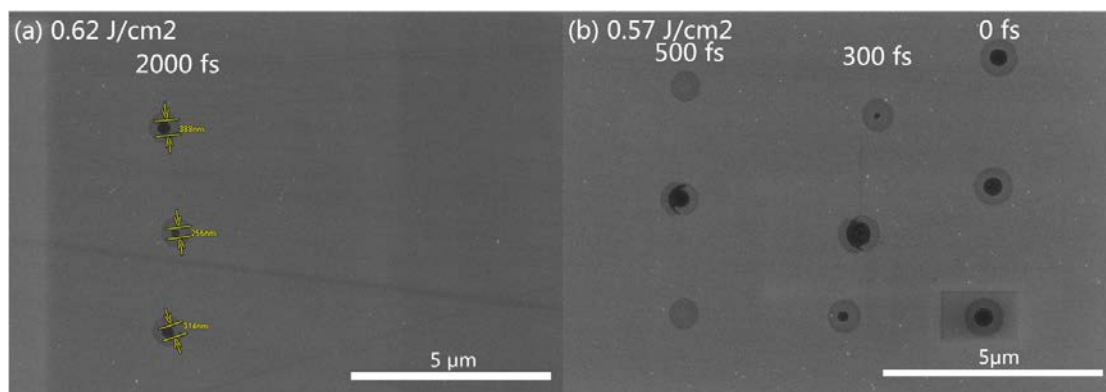


Figure 5. The minimal size of hole fabricated at the laser fluences of 0.62 J/cm^2 (a) and 0.57 J/cm^2 (b), respectively. The experimental parameters of holes in each column are uniform.

4. Conclusion

In conclusion, we use the double-pulse fs laser to achieve the temporal shaping of fs laser. Compared with the traditional single pulse, double pulses can precisely control the properties of free electron such as temperature, capacity and density, etc. by adjusting the pulse delay. By ablating microhole structure on silicon material using double-pulse fs laser, it was found that the average diameter of holes decreases with the increasing pulse delay from 0 fs to 2000 fs under the same laser fluence. Based on the plasma model, this phenomenon can be explained quantitatively from the prospective of ablation threshold. Furthermore, the processing accuracy can be improved using the double-pulse fs laser.

Acknowledgments

The authors gratefully acknowledge the financial support of the National Key R&D Program of China (Grant No. 2017YFB1104300) and the National Natural Science Foundation of China (NSFC) (Grant No. 51575053).

References

- [1] Corbierre M K, Beerens J and Lennox R B 2005 *Chem. Mater.* **17** 5774–9
- [2] Melngailis J, Mondelli A A, Berry I L and Mohondro R 1998 *J. Vac. Sci. Technol. B* **16** 927–57
- [3] Chou S Y, Krauss P R and Renstrom P J 1996 *J. Vac. Sci. Technol. B* **14** 4129–33
- [4] Levis RJ, Menkir GM and Rabitz H 2001 *Science* **292** 709–713
- [5] Jiang L and Tsai H L 2005 *Int. J. Heat Mass Transfer* **48** 487–499
- [6] Jiang L and Tsai H L 2005 *Appl. Phys. Lett.* **87** 151104
- [7] Zhao M J, Hu J, Jiang L, Zhang K H, Liu P J et al. 2015 *Sci. Rep.* **5** 13202
- [8] Javier H, Nadine G, Jan S, Michelina S, Bastian Z, et al. 2015 *ACS Appl. Mater. Interfaces* **7** 6613–6619
- [9] Lan Jiang, An-Dong Wang, Bo Li, Tian-Hong Cui and Yong-Feng Lu. 2018 *Light: Science & Applications* **7** 17134
- [10] Stuart B C, Feit M D, Rubenchik A M, Shore B W and Perry M D 1995 *Phys. Rev. Lett.* **74** 2248–2251
- [11] Sundram K S and Mazur E 2002 *Nature Mater.* **1** 217–224
- [12] Bristow D A, Rotenberg N and Driel M H 2007 *Appl. Phys. Lett.* **90** 191104
- [13] Sokolowski T K and Von der Linde D 2000 *Phys. Rev. B* **61** 2643–2650
- [14] Deng Y P, Xie X H, Xiong H, Leng Y X, Cheng C F, Lu H H, Li R X and Xu Z Z 2005 *Opt. Express* **13** 3096–3103
- [15] Li M, Menon S, Nibarger P J and Gibson G N 1999 *Phys. Rev. Lett.* **82**, 2394–2397
- [16] Liu J M 1982 *Opt. Lett.* **7** 196-198