PERIODIC NANO-HOLE ARRAYS STRUCTURE INDUCED ON SILICON SURFACE BY DIRECT WRITING WITH FEMTOSECOND LASER

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Regular micro-apparatus covered with periodic nano-hole, nano-ridge and ripple structures on silicon bulk (with crystal orientation of 110) were formed by micro-machining with tightly focused beam of the femtosecond laser with wavelength of 800 nm, repetition rate of 1 kHz and the pulse length of 130 fs in air. Use laser direct writing technology to form periodic double rows nano-holes structures and the laser was focus with a $10^{\times}$ focusing objective lens (NA = 0.3). Investigating the relationship between the width of structures and the speed of processing to provide knowledge of the evolvement of the nano-hole and nano-ridge structures.

Key words: *Femtosecond laser; nano-hole arrays; ripples; direct writing.*

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Introduction

Laser induced periodic structure has been extensively investigated since the beginning of laser processing [1]. Femtosecond pulse laser had been shown to be highly effective for precision processing and surface micro-modification due to its fast material processing speed, minimal thermal and mechanical damage, large scan area and single-step capability, it has been demonstrated to be a promising technology for surface nanostructuring of metals and semiconductors [2–5]. Due to the increasing application of nanostructures in numerous fields, nanostructuring of a variety of materials were gaining widespread importance. In the last two decades, direct laser writing or ablation has emerged as a fast and efficient technique for fabricating micro and nanostructures [6–7]. Ripples or, in general, laser induced periodic ripple structures (LIPSs) was the most typical structure induced by femtosecond laser pulse, and has wide potential applications in the fields of physics, chemistry, and materials, such as enhancing light absorption [8], efficient terahertz radiation generation [9], improving catalytic action [10], and strengthening tribological and hydrophilic properties [11]. LIPSs have been observed substantially near the ablation threshold. In the most common type of surface topography, a periodicity of the wavelength λ of the laser radiation is observed . However, the LIPSs with the periodic just a half of the laser wavelength was also reported [12]. Normally, these regular LIPSs are oriented perpendicular to the direction of the laser electric field and have a spatial period very close to the incident wavelength. The mechanism was generally accepted based on the assumption that they were produced by an intensity modulation, which arose from the interaction of the incident and scattered wave with a surface wave created from any periodic modulation in the surface. More recently, it was reported that transient changes of the optical properties of the laser excited solid play a crucial role in the formation of LIPSs in semiconductors [13]. However, the above mechanism were suitable to interpret the formation of periodic ripple grating structures. In this work, we study the formation of periodic nano-hole arrays structure which had been first reported by Zhang et. al. [14]. The underlying mechanism of nano-hole arrays was elucidate by the numerical calculation the electric field distribution using FDTD. In this experiment, we perform the detailed study of surface structures produced by femtosecond laser treatment of silicon surfaces. The influences of the scanning speed and the orientation of polarization of pulse laser on laser-induced surface topography were studied. Between the range of 20 and 2000 μm/s, the nano-hole arrays could be divided into three regions according to the change tendency of nano-hole depth. We perform the processing for obtain the sequence 2D nano-hole arrays and ripple grating structures by precise control the scanning speed and the pulse energy.

Experimental set-up

Fig.1 is schematic diagram of the experimental set-up. A commercial femtosecond Ti:sapphire laser system was employed in our experiments, which was based on the chirped pulse amplification technique. This system provides laser pulses at wavelength of 800 nm and the pulse length of the laser was about 130 fs. The laser running at a repetition rate of f = 1 kHz, from which we can find the spacing of two sequence pulses $d = v/f$, where v is the speed of scanning. The laser beam was focused on the surface of silicon by a microscope objective lens ($10^{\times}$ objective lens with a numerical aperture NA = 0.3) which according to the spot size about 3.5 μm. The surface of the bulk Si samples was polished and cleaned in an ultrasonic bath of ethanol before processing, so that no structure was detectable by light-optical microscopy, then, it was mounted on x-y-z translation stage which controlled by a computer with a spatial resolution of 100 nm. The sample was moved at scanning speed, ranging between 200 and 20000 μm/s. The applied pulse energy was chosen about 300 nJ which near the ablation threshold of silicon. In order to investigate the influence of polarization, Green lens were sent to change the polarization of the pulse laser. The pulse energy was measured directly before the microscope objective. The samples were processed by the focused laser pulse. In order to form periodic structures, we make sure the pulse spacing d (distance between two successive pulses) was smaller than 4 μm. So the sequence pulses overlap at least 60%. The pulse energy was kept constant during the processing. All experiments were performed in air at normal incidence. The surface morphology of the samples was determined by SEM and AFM.

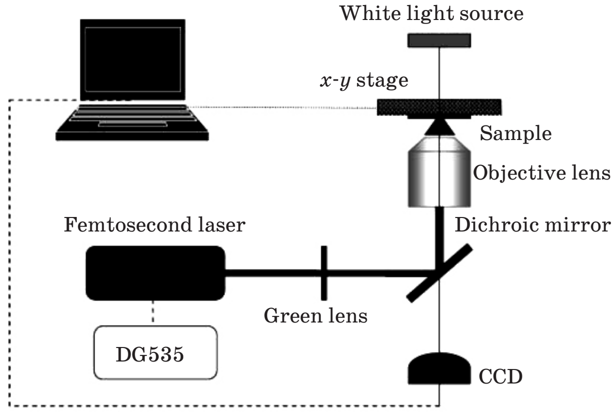

Fig. 1. Schematic diagram of experimental set-up.

Results and discussion

SEM and AFM images of the structure induced on the surface of silicon were shown in Fig. 2. A femtosecond laser was focus by a $10^{\times}$ objective (which with the numerical aperture of 0.3) to form the structures in air. In our experiments, the periodic structures were induced with the direct writing by line scanning with different speed at the certain pulse energy of 300 nJ/pulse which was selected by the previously research [15], the fluence achieve 200 mJ/cm^2 approximately.

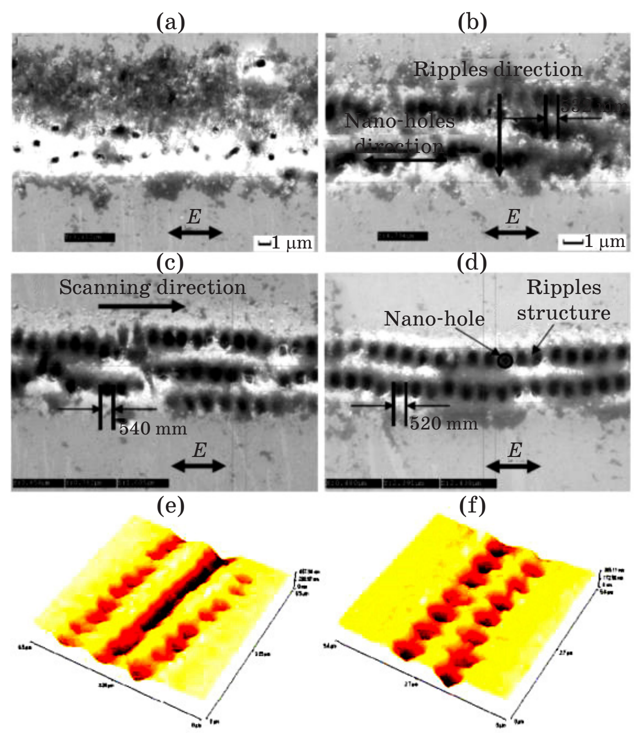

Fig. 2. SEM and AFM micrographs showing nano-hole arrays induced on silicon by a fs laser (λ = 800 nm, f = 1 kHz) at 300 nJ/pulse with different speed, the scanning and polarization with same direction (a) v = 20 μm/s, (b) v = 100 μm/s, (c) v = 200 μm/s, (d) v = 1000 μm/s, (e) the 3D profile of (c), (f) the 3D profile of (d).

As shown in Fig. 2a, one line scan was performed at a scan speed of 20 μm/s, images have been taken directly after ablation without further processing. Some recast or melted matter was observed on the ablation region which covered with disperse nano-hole and melted trace. Periodic structure was dimness and the width of the line channel was about 9.4 μm. When the speed was increased to 100 μm/s, the periodic structure to be more clearer which contain many columns ripples and three line nano-holes as shown in Fig. 2b. Details could be obtained from the image Fig. 2b, the periodic of nano-hole was about 532 nm, also, it was the value of ripple structure period. The orientation of ripple was perpendicular to the polarization, however, the nano-hole and the polarization with the same direction. In addition, with increasing the scanning speed, the width of ablation line decreases to 4.5 μm. With further increase the scan speed to 500 μm/s, the periodic structure was clearer than ever obtained as shown in Fig. 2c. There are three nano-hole chains along the scanning direction which interrupt the ripple columns.

The periodic of the nano-hole and ripple was about 540 nm and the width of the ablation line was about 2.5 μm. Fig. 2d shows very clear image of line scan at the speed of 1000 μm/s. It was interesting that with increasing the scanning speed the number of nano-hole chains were decreased from three to two corresponding the speed increase from 100 to 1000 μm/s, so it shows a strong dependence on the scanning speed. However, the periodic of nano-hole and the ripple was kept constant about 520 nm, it means that the width of the periodic of ripple and the nano-hole was undependence to the scanning speed. Fig. 2e, d show the 3D profile of the periodic structure. It was clearly display the multiline nano-hole structures.

In analogy to the previous case, the width and depth of nano-hole were evaluated as a function of the scanning speed, as plotted in Fig. 3. Ten group data were collected to plot the figure. For the scanning speed range from 20 to 2000 μm/s, the periodic was kept constant which was a little less than the laser wavelength of 800 nm.

However, the depth of the nano-hole could be divided into three tendencies by the speed of 200 and 1000 μm/s. When scanning with speed less than 200 μm/s, the depth of the periodic nano-hole was about 200 nm but with larger fluctuation as shown in Fig. 3. Whereas, if we ablation with greater scanning speed range from 200 to

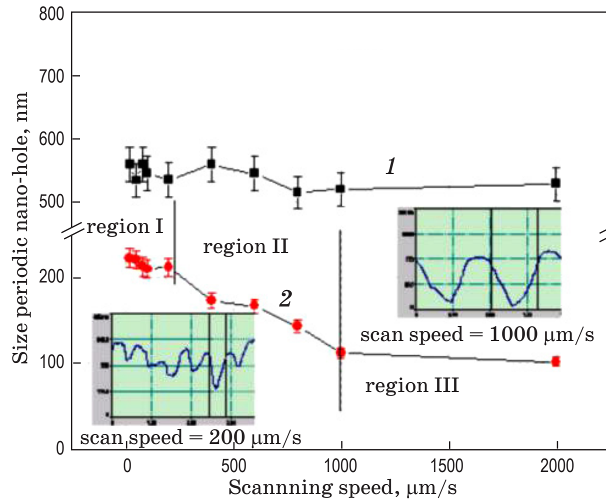

Fig. 3. Width (*1*) and depth (*2*) of the induced nano-hole vary with the different scanning speed.

1000 μm/s, the depth decreases linearly with scanning speed. Finally, with the scanning speed faster than 1000 μm/s, the depth was kept constant about 100 nm and the fluctuation was smaller than ever obtained.

Further experiments had been made using this interesting property of spontaneous formation of periodic structure with fs laser direct writing. For investigate the relationship between the formation nano-hole structure and the polarization, we use a Green lens and a half wave plate to change the orientation of the pulse laser. Then, at the same experiment with the previous research, the nano-hole chains could not be obtained. There is only periodic ripple structures achieved as shown in Fig. 4a, b. With the polarization changed, the nano-hole chains could be obtained stably as shown in Fig. 4c, d. At this experiment, the pulse energy was kept constant of 300 nJ/pulse which was less than the threshold of the silicon, however, the line scanning with different speed. The line scan was composition of a series of sequence points, modulate the scanning speed was equal to change the fluence of irradiation laser pulse. The fs pulse laser with the frequency of 1 KHz and was focused on the silicon surface by an objective lens which with the focus length of 4 mm, so the theoretical laser irradiation spot diameter D_0 was calculated using $D_0 \approx 1.27\lambda f/D$ [16], where f was the effective focal length of the focusing objective lens equal to 4 mm, λ was the wavelength of the laser equal to 800 nm, and D was the laser beam diameter equal to 5 mm. From this formula the theoretical spot size was calculated to be 3.5 μm in diameter. Although, the

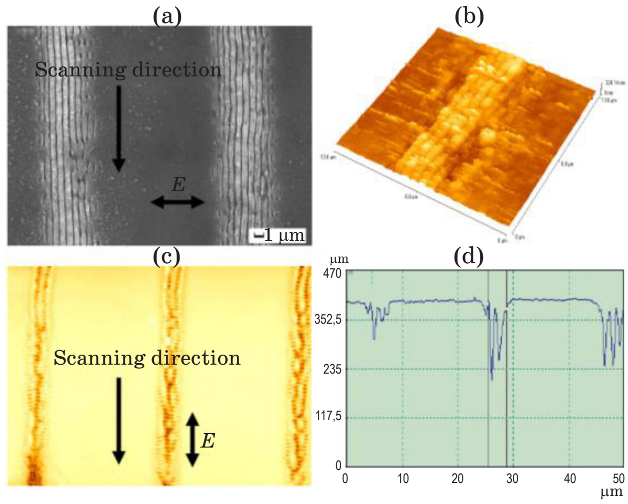

Fig. 4. SEM and AFM micrographs showing nano-hole arrays and ripple structure induced on silicon by a fs laser (λ = 800 nm, *f* = 1 kHz) with 300 nJ/pulse and 2000 µm/s with different polarization orientation (a) and (b) with same direction of scanning and polarization, (c) and (d) with the scanning direction perpendicular to the polarization.

single pulse energy was less than the threshold of the silicon, but through control the scanning speed we could achieve the fluency greater than or less than the threshold. For example, if the scanning speed was 1000 µm/s, the percentage of overlapping of adjacent two points was reach 75%. As shown in Fig. 2a, when the speed was 20 µm/s, per 4 µm region was irradiation with 250 pulses, due to the incubation effect, the surface was melted and no periodic structure was found. Increasing the speed to 100 µm/s, the melted region was observed clearly, however, the periodic nano-hole arrays were obtained also as shown in Fig. 2b. With further increasing the scanning speed to 500 and 1000 µm/s, the number of nano-hole chains were decreased from 3 to 2 and the melted region disappear as shown in Fig. 2c, d. The spontaneous formation of periodic ripples which induced by laser pulse were reported extensive [17, 18]. The ripples structures with two typical characteristics, the direction was perpendicular to the polarization of the pulse and the periodic was at the same scale to the wavelength of the laser. However, it was obviously that the nano-hole arrays and the polarization with the same orientation. So, it could not be simply interpreted by the theory of interference between the incident light/surface wave which was the most popular mechanism to interpret the formation of the periodic ripple structures [19]. In addition, the self-organize theory also could not give the complete explanation for nano-hole arrays which describe as due to the forces induced by the thermal gradient, the melted material moves from the hot regions to colder region, then periodical structures are formed [20–21]. The same periodic structure was obtained by Zhang et. al. recently [14]. They use the finite-difference time-domain technique for numerical calculation of the electric field distribution. The effect was due to local field enhancement in the direction of polarization on the former induced ripple structure. This mechanism could be used to interpret the formation of nano-hole arrays. However, there still exist doubt about the formation of nano-hole arrays, as the former theory describe, emergence nano-hole arrays has a prerequisite for induced periodic ripples structures. As reported there was no evidence for satisfying this condition. In addition, the width of the nano-hole was kept constant nearly, this was opposite to the reported. Although there are many theories to interpret the produce of nanostructures induced with ulra-short pulses, but the formation mechanism of periodic nano-hole arrays also have not yet been thorough elucidated.

Conclusion

In summary we have performed periodic nano-hole arrays and ripples structures on silicon surface by the use of nJ pulse at a repetition rate of 1 KHz in air. 2D nano-hole arrays and ripples grating were direct written by accurate processing modulation through scanning speed. We discuss the formation mechanism of the nano-hole arrays.

It was found that the nano-hole has a strong dependence with the polarization and the irradiation fluence of the pulse laser. These results are of prime interest for potential applications as high density data storage, anti-reflectivity, nanolithography and other optical applications in the nanoscale.

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