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Self-organizing microstructures orientation control in femtosecond laser patterning on silicon surface

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Abstract: Self-organizing rippled microstructures are induced on silicon surface by linearly polarized femtosecond laser pulses. At a near threshold fluence, it is observed that ripple orientation is co-determined by the laser polarization direction and laser scanning parameters (scanning direction and scanning speed) in surface patterning process. Under fixed laser polarization, the ripple orientation can be controlled to rotate by about 40° through changing laser scanning parameters. In addition, it is also observed that the ripple morphology is sensitive to the laser scanning direction, and it is an optimal choice to obtain ordered ripple structures when the angle between laser scanning and laser polarization is less than 45°.

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References and links
1. Introduction

Direct writing techniques, including plasma spray, e-beam, focused ion beam and laser beam, are the main methods for material processing and surface patterning for several decades [1]. More recently, femtosecond (fs) pulse laser surface patterning has attracted considerable interest due to its capability in micro/nanostructures fabrication, which may find potential applications in various fields of science and technology [2, 3].

When the focused laser beam irradiates on the material surface, periodic surface structures, which are also called LIPSS or ripple, are induced at the laser fluence near or just above the ablation threshold. This kind of structures was firstly proposed by Birnbaum on various semiconductor surfaces [4]. Since then, different kinds of ripples have been reported on virtually all materials [5–7]. For semiconductor silicon, up to now, there are two distinct kinds of ripples: high spatial frequency ripple [8] and low spatial frequency ripple [9]. The former one with spatial periodicity of about 100 nm, is always induced under high repetition rate [10] or in liquid environment [11]. Whereas in most cases, low spatial frequency ripples are observed with spatial periodicity close to or somewhat smaller than the laser wavelength. For the laser with central wavelength of 800 nm, the previously reported spatial periodicities of the low spatial frequency ripples are between 560 nm to 770 nm [12]. Previous researchers believed that the formation mechanism of this kind of ripple is the result of the interaction between the incident light and the surface scattering wave [13]. Nowadays, Huang et al. revised the simplified scattering model, taking into account the surface plasmons (SPs). They considered that the ripples result from the initially direct SP-laser interference and the subsequent grating-assisted SP-laser coupling [14]. Although the underlying mechanism responsible for the ripples formation remains controversial, it is widely accepted that the ripple orientation is dominated by the polarization state of the incident laser beam, which has been verified by multi-shot experiments [15]. Varlamova et al. investigated the ripples induced on silicon surface by elliptically polarized laser beam, indicating that the ripple orientation is determined by the major axis of the polarization ellipse [16]. For linearly polarized laser beam, the ripple orientation induced on silicon surface is exactly perpendicular to the irradiation beam polarization [17].

Our previous studies also confirm that the ripple orientation is determined by laser polarization direction [18, 19]. However, this widely accepted principle cannot be extended to the surface patterning process. In this study, linearly polarized fs laser pulse is used to write continuous lines on silicon surface, we found that the ripple orientation is not only determined by the laser polarization direction, but also interrelated with the laser scanning direction and the laser scanning speed. This intriguing phenomenon, which is firstly proposed here, is attributed to the ripple structures induced on the initial stage in scanning process, which may act as seeds for subsequent ripple orientation changes. Besides, another two-step experiment is conducted to understand the process of the ripple orientation changes.
2. Experimental setup

Figure 1 shows the schematic diagram of the experimental setup. An amplified, mode-locked titanium-sapphire based laser system (Spectra Physics Inc.) is used for the generation of fs laser pulse with a central wavelength of 800 nm and pulse duration of 50 fs. A half wave plate and a polarizer are combined together to serve as attenuator and the pulse energy is monitored by a calibrated photodiode. The sample is a 1.0 mm thick, single surface polished crystalline silicon with crystal orientation of (1 1 1). The sample is fixed on a 6-axis translation stage (M-840.5DG, PI, Inc.), which can be programmed to move at constant speed. The laser beam is focused onto the sample surface at normal incidence by a 5 × microscope objective (Olympus) with numerical aperture of 0.15. The fs laser repetition rate is adjusted to be 200 Hz. The laser polarization direction is fixed along the X axis. We define $\theta$ as the angle between the laser polarization direction and laser scanning direction. Scanning Electron Microscopy (SEM) is used to inspect the sample surface after fs laser treatment.

![Fig. 1. Schematic diagram of the experimental setup. The laser polarization direction is along X axis. S: laser scanning direction, P: polarizer, HWP: half wave plate.]

3. Results and discussion

In the first set of experiments, the fs laser pulse is scanned over the sample surface at $\theta$ ranging from 0° to 90°, resulting in a series of self-organizing rippled lines. Each line is scanned only once to avoid the energy accumulation. The laser scanning speed ($v$) is 100 $\mu$m/s. Low frequency ripples are formed as shown in Fig. 2(a). When $\theta < 45^\circ$, it is evident that the induced ripple is continuous and straight; while $\theta > 45^\circ$, the ripple becomes discontinuous and curved. This is due to the elliptical shape of the ripple structure, which has been discussed in our previous research [18]. Besides the ripple continuity, we can also observe that the ripple morphologies, including ripple orientation and uniformity, are somewhat different.

Figures 2(b) and 2(c) are partially enlarged view of the lines at $\theta = 0^\circ$ and $\theta = 90^\circ$ in Fig. 2(a). It is worth noting that Fig. 2(c) is rotated vertically for intuitive comparison. It can be seen that the orientations and uniformities of the induced ripples in the two cases are different. In Fig. 2(b), at $\theta = 0^\circ$, the induced ripple has a regular arrangement and clear boundary, and its orientation is exactly perpendicular to the laser polarization direction. While in Fig. 2(c), $\theta = 90^\circ$, the induced ripple structure looks less ordered, and no distinguishable boundary can be observed. The ripple orientation is also chaotic and no longer exactly perpendicular to the laser polarization. Therefore, comparing with the two cases, scanning along the laser polarization direction ($\theta = 0^\circ$) is beneficial to inducing ripple structures with clear boundary and uniform orientation.

Furthermore, in Fig. 2(a), we also find that not all of the ripple orientations are perpendicular to the laser polarization direction. This observation is not consistent with the widely accepted principle and reveals the dependence of the ripple orientation on the laser scanning condition. The following detailed experiments are conducted to explore the influence of laser scanning parameters ($\theta$ and $v$) on the ripple orientation.
Fig. 2. (a) SEM images of ripples on silicon surface at $v = 100 \, \mu m/s$. (b) and (c) are partially enlarged view of lines in (a) at $\theta = 0^\circ$ and $\theta = 90^\circ$. It is worth noting that (c) is rotated vertically.

Figures 3(a)–3(c) depict the SEM images of the lines written on the sample surface at $v = 100 \, \mu m/s$. In Fig. 3(a), $\theta = 0^\circ$, it can be seen that the orientation of the induced ripple (along Y axis) is perpendicular to the laser polarization direction, which is in agreement with the previous researches [19]. In Fig. 3(b), $\theta = 30^\circ$, ordered and distinguishable ripple pattern is also formed. However, the ripple orientation changes unexpectedly, which is no longer perpendicular to the laser polarization direction and rotates by about 20° with respect to the widely assumed ripple direction (Y axis). The rotation of the ripple orientation becomes more obvious when $\theta = 45^\circ$, as shown in Fig. 3(c). The rotation value reaches up to 30°, tending to be perpendicular to the laser scanning direction. When $\theta > 45^\circ$, the rotation value of the induced ripple does not increase any more, but the ripple structures begin to become chaotic (Fig. 2(a)). Hence, $\theta < 45^\circ$ is an optimal choice for ordered ripples fabrication. Under fixed laser polarization and laser scanning speed, the ripple orientation changes with the laser scanning direction, confirming that the ripple orientation is not only determined by the laser polarization, but also interrelated with the laser scanning direction.

Fig. 3. SEM images of the lines written at (a) $\theta = 0^\circ$, (b) $\theta = 30^\circ$, (c) $\theta = 45^\circ$, $v = 100 \, \mu m/s$. The dashed arrow indicates the laser scanning direction. (d), (e) and (f) are SEM images of the lines written at different laser scanning speeds, $\theta = 45^\circ$. The scale bar is 5 μm.
Figures 3(d)–3(f) show the SEM images of the ripples on sample surface at $\theta = 45^\circ$. In Fig. 3(d), $v = 80$ $\mu$m/s, the induced ripple has an ordered arrangement and slight orientation rotation. When $v = 100$ $\mu$m/s, an obvious rotation can be observed as shown in Fig. 3(e), under this experimental condition, the ripple orientation rotates by about $25^\circ$ with respect to Y axis. While $v$ is increased to $120$ $\mu$m/s, the orientation of the induced ripple is nearly perpendicular to the laser scanning direction, as shown in Fig. 3(f). Under fixed laser polarization and laser scanning direction, the ripple orientation rotation increases with the laser scanning speed ($80$ $\mu$m/s - $120$ $\mu$m/s).

In order to quantitatively represent the dependence of the ripple orientation on the laser scanning parameters, we character $\alpha$ as the angle between the ripple orientation and the Y axis. $\alpha = 0^\circ$ denotes that the ripple is perpendicular to the laser polarization, as shown in Fig. 3(a). The relation between $\alpha$ and $\theta$ under different laser scanning speed is plotted in Fig. 4. Each data represents an average of two measured data points and the error bars are shown. At $v = 60$ $\mu$m/s, $\alpha = 0^\circ$ when $\theta$ increases from $0^\circ$ to $90^\circ$, which reveals that under lower scanning speed, the ripple is always perpendicular to the laser polarization direction and nearly unrelated with the scanning direction. However, when the laser scanning speed is increased to $100$ $\mu$m/s, $\alpha$ increases with angle $\theta$ and reaches its maximum at $\theta = 45^\circ$, then gradually drops down. The error bars are greater when $\theta > 45^\circ$ because of the chaotic ripple arrangement, as mentioned in Fig. 2. At $\theta = 45^\circ$, it can be also observed that the measured $\alpha$ increases from about $0^\circ$ to about $40^\circ$ with the laser scanning speed, as shown in Fig. 5. Under this condition, by adjusting the laser scanning speed, the ripple orientation can be controlled to be perpendicular to the laser polarization direction or laser scanning direction.

![Fig. 4. The relation between ripple orientation ($\alpha$) and laser scanning direction ($\theta$) at $v = 60$ $\mu$m/s and $v = 100$ $\mu$m/s.](image)

![Fig. 5. The dependence of the measured $\alpha$ on the laser scanning speed at $\theta = 45^\circ$.](image)
The aforementioned results demonstrated an unusual phenomenon that the ripple orientation can be controlled to rotate by changing the laser scanning parameters under fixed laser polarization, which is not consistent with the previous principle that the ripple orientation is determined by the laser polarization direction. Repeated experiments are conducted on silicon with different crystal orientations to exclude the influence of surface defects and crystal orientation.

Gottmann et al. [20] proposed that the ripple formation is a feedback process, where existing structures lead to polarization depended scattering of the light. If the laser irradiates at the position of a pre-existing surface structures like scratches, the orientation of the induced ripple will also along the scratches. Therefore, we believe that the ripple orientation rotation can be attributed to the ripples induced on the initial stage of the laser scanning process, which acts as the seeds for subsequent ripple line formation. The following is a series of two-step induced ripple spots, aiming to explain the influence of the initially induced ripple on the subsequent ripple formation. Figure 6(a) is the schematic diagram of the experimental process. The laser polarization is still along the X axis and XOY plane represents the sample surface. For the first step, the sample surface is pretreated by 10 laser shots on point O, elliptically shaped ripple structure will be induced on this point [18]. And then for the second step, another 10 laser pulses are shot on point B, C or D by moving the translation stage. The distances between OB, OC and OD are about 2 μm, 4 μm and 6 μm, respectively. The angle between line OB and X axis is about 30°.

Figures 6(b)–6(d) are SEM images of the two-step experimental results. The solid-line ellipse represents the first-step irradiated spot while the dashed-line ellipse represents the second-step irradiated spot. In Fig. 6(b), it can be seen that more than half regions of the two ellipses are overlapping. The orientation of the second-step induced ripple is still perpendicular to the laser polarization. In Fig. 6(c), the center distance between the two-step induced ripple ellipses is increased to about 4 μm. Under this condition, the second-step induced ripple in the dashed-line ellipse rotates by about 16° with respect to the ripple induced on the first-step, which is the beginning of the ripple orientation changes. When the center distance between the two ellipses is increased further to about 6 μm, two ripple structures are almost separated in space and the influence between each other vanishes, so the ripples in the two ellipses are ordered and their orientations are perpendicular to the laser polarization. In addition, similar two-step experiments are also conducted along the X axis and Y axis, but obvious ripple orientation change as shown in Fig. 6(c) has not been observed.

Fig. 6. (a) Schematic diagram of the two-step experimental process. (b), (c) and (d) are SEM images of the ripple structures induced by the two-step method. The scale bars are 1 μm.

The formation of the self-organizing rippled lines in Fig. 3 can be considered as the repetition of the two-step process. The control of the ripple orientation by scan speed and scan direction is mainly attributed to the overlap during the scanning process. When the fs laser is irradiated on sample surface, initially seeding ripple is induced with orientation perpendicular to the polarization. The overlap areas and positions between the seeding ripple and irradiation beam change with the scan speed and scan direction, which could affect the orientation of the
neighboring formed ripples under certain conditions (Fig. 6(c)). This seeding effects play a dominant role for the subsequent ripple formation, leading to the entire ripple orientation changes eventually. As for the rotation in Fig. 6(c), it is attributed to the first-step induced structures that may redistribute the incident optical field, leading to the periodic ablation along specific direction. Moreover, it may be also related with the surface plasmon polariton (SPP). Previous researches reveal that initially structured surface could affect the generation and propagation of the SPP [21, 22]. Hence, when the laser beam irradiates on certain positions of the initial structure, the coupling between specially excited SPP and incident laser beam may result in the ripple orientation changes. The exact mechanism for this phenomenon is still unclear and needs more investigation.

4. Conclusion

In summary, the dependence of ripple orientation on laser scanning parameters, including scanning speed and scanning direction, has been experimentally investigated. For linearly polarized fs laser surface patterning, at a near threshold fluence, the orientation of the self-organizing rippled structure is collaboratively dominated by the laser polarization and laser scanning parameters. For fixed laser polarization, the ripple orientation can be controlled to rotate by about 40° through changing the laser scanning parameters. Another two-step experiment is conducted to explain the process of ripple orientation rotation. Besides, it is also observed that \( \theta < 45^\circ \) is an better choice for ordered ripples (clear boundary and uniform orientation) fabrication, which could promote the development of electro-optic devices. In future, the corresponding theories should be investigated to further understand the mechanisms of ripple formation and orientation rotation.

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