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Continuous modulations of femtosecond laser-induced periodic surface structures and scanned line-widths on silicon by polarization changes

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Abstract: Large-area, uniform laser-induced periodic surface structures (LIPSS) are of wide potential industry applications. The continuity and processing precision of LIPSS are mainly determined by the scanning intervals of adjacent scanning lines. Therefore, continuous modulations of LIPSS and scanned line-widths within one laser scanning pass are of great significance. This study proposes that by varying the laser (800 nm, 50 fs, 1 kHz) polarization direction, LIPSS and the scanned line-widths on a silicon (111) surface can be continuously modulated with high precision. It shows that the scanned line-width reaches the maximum when the polarization direction is perpendicular to the scanning direction. As an application example, the experiments show large-area, uniform LIPSS can be fabricated by controlling the scanning intervals based on the one-pass scanned line-widths. The simulation shows that the initially formed LIPSS structures induce directional surface plasmon polaritons (SPP) scattering along the laser polarization direction, which strengthens the subsequently anisotropic LIPSS fabrication. The simulation results are in good agreement with the experiments, which both support the conclusions of continuous modulations of the LIPSS and scanned line-widths.

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References and links


1. Introduction

Although traditional nanolithography is of great potentials for nanoscale devices fabrication, the process is very complicated [1]. Self-organized structures provide simpler and cheaper ways [2]. A recent developed method of self-structuring, laser induced periodic surface structures (LIPSS, also referred to as ripples) have been extensively studied for various materials, including semiconductors [3,4], metals [5,6], and dielectrics [7–10], for their...
promising applications in solar cells [3], waveguides [7], colorization [5,11], light extracting surfaces in light emitting diodes (LED) [12], surface enhanced Raman scattering (SERS) [13], and water-repellent surfaces [14,15]. All the applications require the formation of well-defined large-area, uniform LIPSS. However, high precision formation of LIPSS remains a big challenge, which limits industry applications. Compared to other physical and chemical methods for preparations of large-area, uniform nanoscale structures [16], direct laser-scanning-induced LIPSS on a material’s surface using femtosecond (fs) pulses is quite simple and efficient, which open new possibilities for nanofabrication [11,17–19]. As a key parameter, the scanning interval is of great importance in the formation of large-area, uniform LIPSS, which significantly affects the continuity of the LIPSS and the processing efficiency. Therefore, it is important to study the scanned line-width, which determines the scanning intervals during the laser-scanning-induced large-area LIPSS fabrication.

Previous studies on LIPSS under irradiation of fs laser pulses showed that the formation of LIPSS primarily correlates to the material properties as well as the irradiation conditions, such as the laser wavelength [20], the laser fluence [10], the pulse duration [9], the pulse number [4], and the incident angle [4,6]. More recently, we explored the periodicity and orientation of the LIPSS modulation by a well-designed fs laser pulse train [21], which provides a new method for obtaining controllable, smaller nanogratings. Recent investigations reveal that laser polarization, as an additional controlling freedom, also plays an important role in the formation of LIPSS. For example, Reif and Varlamova et al. studied the influence of variable laser polarizations of incidence on fluoride crystals [22,23]. Varlamova found that different ripple orientations could be created with elliptically polarized femtosecond lasers and with spherical nanoparticles produced by circular polarization. In contrast to the aforementioned observations, Wang et al. pointed out that regular ripple structures slant by 45° with respect to linear polarization and can be produced under the irradiation of circularly polarized fs laser pulses [24]. Recently, two-dimensional (2D) LIPSS have been formed by arbitrary designed fs vector laser fields on the surface of silicon [25]. Most studies have been focused on the effect of fs laser polarization on LIPSS orientations. However, there have been few studies addressing the laser-scanning LIPSS line morphology with only one pass, which enables precise control of large-area, uniform LIPSS formation.

In this study, the scanned line-widths modulation based on polarization changes is investigated. It is shown the scanned line-width reaches the maximum when the polarization direction is perpendicular to the scanning direction. The minimum scanned line-width is obtained with the polarization direction parallel to the scanning direction. Meanwhile, when the polarization direction (α) is tuned between 0° and 90°, the scanned line-width can be flexibly modulated continuously. To show this polarization-dependent scanned line-width application, we further show the large-area, uniform LIPSS formation by controlling the scanning directions and polarization directions, which greatly improves the processing efficiency and precision. We try to interpret this phenomenon in terms of the anisotropic SPP scattered by the initially formed LIPSS. The simulation also confirms the dominant role of the initially formed ripples. Furthermore, based on the numerical simulation, the geometrical morphology of the LIPSS under static fs laser irradiation can be continuously modulated by polarization directions.

2. Experimental setup

In the experimental arrangement (as shown in Fig. 1), the laser source is a Ti:sapphire laser regenerative amplifier system, which provides a fundamental Gaussian mode with a central wavelength of 800 nm, pulse duration of 50 fs, and repetition rate of 1 kHz. An achromatic half-wave plate and a linear polarizer are used to control the laser fluence incident on the sample surface. Another half-wave plate is used to change the polarization direction of the incident laser pulses. The pulse number (N) delivered to the sample is controlled by a fast mechanical shutter synchronized with the laser repetition rate. The laser light travels through the dichroic mirror (DM) and is focused through an achromatic doublet (f = 100mm) on the surface of the sample. The focal spot size (D) (width at the waist defined by 1/e² point) of the
Gaussian beam, which is close to the beam diameter in the sample processing plane under good focused conditions, is measured as 60 μm. The highly polished silicon (111) sample (10 mm × 10 mm × 1 mm) is mounted on a computer-controlled, six-axis moving stage (M-840.5DG, PI, Inc.) with a positioning accuracy of 1 μm in the x and y directions and 0.5 μm in the z direction. To observe the fabrication process, a charge coupled device (CCD) camera along with a white-light source is used to image the sample surface. All experiments are carried out in air at room temperature. After irradiation, the surface morphology is characterized by a scanning electron microscope (SEM).

![Diagram of experimental setup](image)

**Fig. 1.** Schematic diagram of the experimental setup. The insert depicts the relative angle between directions of the linearly polarized fs laser and the sample coordinate. HWP: half-wave plate; P: polarizer; S1: shutter; WS: white-light source; BS: beam splitter; L1: convex lens; DM: dichroic mirror; L2: achromatic doublet; S2: sample.

### 3. Results and discussion

#### 3.1. Continuous modulations of scanned line-width for potential large-area uniform LIPSS formation

Figure 2 shows the scanned line-width as a function of laser polarization direction. The average laser fluence is 0.75 J/cm² of the Gaussian beam, which is slightly higher than the single-shot ablation threshold (0.637 J/cm²) [26]. The repetition rate and scanning speed are 250 Hz, and 500 μm/s, respectively. Additionally, each scanned line has only one pass of the laser with an identical scanning direction. The LIPSS always appear as grating-like distributions. As shown in Fig. 2 inserts, a range of scanned lines are produced and the widths decrease as the polarization ranging from 0° to 90°. During the variation of polarization directions, the LIPSS periods remain almost unchanged with approximately 650–750 nm; and the orientations of the LIPSS structures are almost perpendicular to the polarization direction in all cases. The periods and directions of the LIPSS are consistent with previous study [27]. At an angle of α = 0°, at which the laser polarization is perpendicular to the scanning direction, the scanned line-width reaches the maximum. While, at an angle of α = 90°, at which the laser polarization is parallel to the scanning direction, the minimum scanned line-width is obtained. The ratio of minimum width with 90°-polarization along the maximum width of LIPSS with 0°-polarization is about 0.77 at the given fluence.
Using the proposed method, we can precisely control large-area, uniform LIPSS formation by properly adjusting the interval of two adjacent scanning lines. Therefore, appropriate intervals of the scanning lines are prerequisite for the formation of a large-area, uniform LIPSS. In this study, we control the scanning interval corresponding to the scanned line-width, which is shown in Fig. 2. As shown in Fig. 3, the $1 \times 1$ mm$^2$ area can be produced with various processing times, in which the polarization directions range from 0° to 90°, at the fixed pulse energy, repetition rate, scanning speed, and identical scanning directions in the aforementioned experiment, with different polarization-based scanning intervals based on the above research results. When the polarization direction ($\alpha = 0°$) is perpendicular to the scanning direction, the scanning time can be reduced from 220 s to 170 s compared with the scanning time at 90°-polarization, as the scanning interval is the maximum. While, with the polarization parallel to the scanning direction, the large-area uniform LIPSS processing is the most time-consuming. The processing time ratio of [170 s (0°)]/[220 s (90°)] is 0.77, which is consistent with the aforementioned scanned line-width of the orthogonally polarized directions. Therefore, the processing efficiency and precision can be improved by optimizing the scanning directions and the polarization directions. Meanwhile, by precisely selecting the scanning interval based on the scanned line-width with only one pass, the respective scanning lines and the “overhatch” effect [17] can be eliminated, so that the continuity of the LIPSS can be precisely controlled, which is very beneficial in applications.
3.2 Numerical simulation and potential theoretical analysis

Up to now, the mechanism of the LIPSS formation is still a subject of discussion and is usually related to the optical or mechanical reaction of irradiated surfaces, which may involve, e.g., collective electronic behavior, selective energy deposition, or the capillarity phenomena. It is now widely accepted that the excitation and propagation of SPP plays a crucial role in the formation of LIPSS [4,8]. To explain the above proposed polarization-dependent scanned line-width of LIPSS modulation, SPP scattering is proposed in this study. It is expected that the interaction of the laser with the material surface will be changed after the formation of initial ripples, which leads to the redistribution of the incident laser that may significantly affect the subsequent ablation process [28]. According to a previous study [29], the initially formed ripples, which behave as a surface grating, may facilitate the coupling between the incident laser light and the surface plasmon wave. Meanwhile, Garrelie et al. experimentally demonstrated the effect of an initial structured surface on the role of SPP, which highlights its important role in LIPSS formation [30]. Furthermore, research shows that for linearly polarized fs irradiation, directional SPP scattering is anticipated [31], strengthening the subsequent LIPSS fabrication.
In order to comprehensively explain the physical mechanism, we have calculated the electric field distribution on a grooved surface (i.e., the surface with periodic ripples) by using a commercial finite-element-method software, COMSOL MultiPhysics. It is assumed that three grooves have been produced on the surface of the silicon along they axis as a result of initial ripple formation. The geometry of the grooves can be described by four parameters, $l$, $w$, $h$, and $d$, which denote the length, width, depth, and the period of the grooves, as depicted in Fig. 4(a). Under the irradiation of fs laser pulses, the normal state of the complex refractive index of silicon is dramatically modified due to the high density of photogenerated carriers [28,32]. Since the laser fluences in this study are just above the damage threshold of silicon, the real part of the refractive index remains nearly unchanged while a significant increase in $\kappa$ is expected. Therefore, the complex refractive index of silicon is chosen to be $n = 3.4 + i0.5$ [32]. The fixed complex refractive index makes our numerical simulations a semiqualitative theoretical analysis to the anisotropic phenomenon. In the numerical simulation, the laser beam polarized along the $x$ axis is normally incident on the silicon surface, and the electric field ($E$) distribution on the surface of the grooves is shown in Fig. 4(b). The diameter of the laser beam is assumed to be much larger than the area with grooves. Based on experimental observations, we fixed $w$, $d$, $h$, and $l$ to be 0.4, 0.7, 0.1 and 2 $\mu$m. The fringes formed induce the subsequent SPP scattering. Equally strengthened SPP are scattered toward the left and right sides of the grooves, which are perpendicular (parallel to the polarization direction) to the orientation of ripples. Therefore, along the laser beam polarization direction, the formation of the subsequent ripple structures is strengthened due to the enhancement of the SPP scattering. However, in the perpendicular direction, the LIPSS formation is difficult due to the weakening of the SPP scattering. This strengthened directional SPP scattering leads to the elongated ablated areas with surface ripple structures along the laser polarization so that the maximum line widths can be achieved when the polarization direction is perpendicular to the scanning direction. Figure 4(c) shows a schematic illustration for the scanned line-width modulations with partial polarization directions. It illustrates that during the variation of polarization directions, the LIPSS formation keeps directional formation along the laser polarization directions all the cases, which lead to the directional line width.

3.3. Continuous modulations of the LIPSS geometrical morphology under static fs laser irradiation

Based on the above simulation, we can deduce an anisotropic elliptical geometrical morphology of LIPSS, which preferential to the polarization-direction under static fs laser irradiation. Therefore, the geometrical morphology of the LIPSS can be continuously modulated by polarization directions. To demonstrate the tendency LIPSS fabrication along the laser polarization directions, we further perform the LIPSS formation under different polarizations. Figure 5(a-g) contains SEM images of the LIPSS on silicon irradiated by 200 linearly polarized fs laser pulses. The average laser fluence is 0.8 J/cm$^2$. As expected, the LIPSS exhibits an anisotropic elliptical geometrical morphology. However, the intensity measurements reveal that the laser beam is close to the symmetric Gaussian distribution, and no significant changes are observed by varying the polarization directions. During the variation of polarization directions, the LIPSS periods remain almost unchanged with the laser-scanning induced LIPSS formation, and the orientations of the LIPSS structures are almost perpendicular to the polarization direction in all cases. Nevertheless, as shown by the geometrical area marked by the dotted line in Fig. 5(a-g), the geometrical morphology of LIPSS strongly depends on the laser polarization. As a matter of fact, one can note that the major axis of the ellipse-shaped LIPSS tends to take on a preferential orientation parallel to the direction of laser polarization, which is agreement with the above theoretical deduction. For instance, at a polarization angle of $\alpha = 0^\circ$, the major axis of the ellipse-shaped LIPSS is vertical, as shown in Fig. 5(a). However, when $\alpha$ is rotated by 45$^\circ$, the major axis orientation of the ellipse-shaped LIPSS is also slanted by 45$^\circ$. In order to numerically represent this anisotropy, we define a parameter $K$ (anisotropy) as the ratio of $L_1$ to $L_2$, where $L_1$ and $L_2$ are
the length of axes with orientation perpendicular and parallel to the laser polarization direction, respectively. The parameter $K$ of the LIPSS almost remains unchanged at approximately 0.63 to 0.68 during the variations in polarization direction. As a comparison, for circularly polarized fs laser irradiation, regular ripple structures are produced under the same processing parameters, but the ripple orientation seems to slant by 45° with respect to the linear polarization case, which is consistent with previous studies [33,34]. In spite of these observations of slantwise orientation, no convincing relevant explanations have been achieved. As seen in Fig. 5(h), the overall morphology of LIPSS is nearly circular. Its period is almost consistent with those irradiated by the linearly polarized pulses.

During our investigation, we varied the pulse numbers at a fixed fluence. Figure 6 shows the dimensions and anisotropy ($K$) of LIPSS as a function of irradiated pulse numbers ($N$). We found that the dimensions of the major (or minor) axis are growing as the pulse numbers increase. The parameter $K$ increases from 0.54 to 0.9 with the increase in pulse number, which indicates the decreasing ellipticity of LIPSS. The SEM images [Fig. 6 inserts (a)-(h)] show the development of LIPSS as the number of laser pulses increases. For $N = 50$, LIPSS are formed [Fig. 3(a)] with an ellipse-shaped geometrical morphology. When the number of irradiation pulses is increased to 150 [Fig. 6(c)], another characteristic feature develops in the form of grooves with the periods of 1.6-2.7 $\mu$m [as indicated in the magnified image of Fig. 6(d)]. Their orientation is parallel to the polarization direction. This has been described in previous studies [35,36], which attribute its formation to the capillary waves. If the pulse number is raised to 900, as shown in Fig. 6(h), LIPSS vanish; meanwhile, the ablated area evolves into a nearly circular morphology. This observed decrease of ellipticity with increasing pulse numbers is also attributed to the anisotropic SPP scattering. As pulse numbers increase, the opposite-oriented coarsened ripples are fabricated on the top of initiate LIPSS. This induces the opposite anisotropic SPP scattering enhancement, which weakens the initially anisotropic SPP scattering enhancement, thereby leading to the weakened ellipticity of ablated areas with LIPSS.
Fig. 6. Dimensions and the anisotropy parameter ($K$) of the LIPSS ellipse-shaped region as a function of irradiated pulse numbers. The laser pulse energy is $0.8 \text{ J/cm}^2$ in all cases. Inserts (a) - (h) SEM images of LIPSS with different pulse numbers. All the images share the same scale bar.

4. Conclusion

This study proposes an effective new method to continuously modulate fs laser LIPSS and scanned line-widths by polarization changes. When the laser polarization is perpendicular to the scanning direction, the maximum scanned line-width is obtained. When the polarization direction is parallel to the scanning direction, the scanned line-width reaches its minimum. The simulation results show that the directional scattering of SPP by the initially formed surface ripple structures strengthens the subsequent LIPSS fabrication along the laser polarization, which interprets the aforementioned phenomenon. Furthermore, the simulation is in agreement with the experimental results of anisotropic geometrical morphologies of LIPSS under static fs laser irradiation with different polarizations, which strongly supports the proposed method of continuous modulations of LIPSS and scanned line-widths. The effect of pulse number on the anisotropic morphology of LIPSS is also investigated. The asymmetry (ellipticity) decreases as the pulse number increases.

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