6-30-2014

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Anisotropy modulations of femtosecond laser pulse induced periodic surface structures on silicon by adjusting double pulse delay

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Abstract: We demonstrate that the polarization-dependent anisotropy of the laser-induced periodic surface structure (LIPSS) on silicon can be adjusted by designing a femtosecond laser pulse train (800 nm, 50 fs, 1 kHz). By varying the pulse delay from 100 to 1600 fs within a double pulse train to reduce the deposited pulse energy, which weakens the directional surface plasmon polarition (SPP)-laser energy coupling based on the initial formed ripple structure, the polarization-dependent geometrical morphology of the LIPSS evolves from a nearly isotropic circular shape to a somewhat elongated elliptical shape. Meanwhile, the controllable anisotropy of the two-dimensional scanned-line widths with different directions is achieved based on a certain pulse delay combined with the scanning speed. This can effectively realize better control over large-area uniform LIPSS formation. As an example, we further show that the large-area LIPSS can be formed with different scanning times under different pulse delays.

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OCIS codes: (320.5540) Pulse shaping; (320.2250) Femtosecond phenomena; (220.4000) Microstructure fabrication.

References and links
1. Introduction

Laser-induced damage has been intensively studied, not only for its broad industry applications but also for its rich physics [1]. Recently, advances in femtosecond (fs) laser technologies, which operate on the time scale of electron dynamics, offer better perspectives on micro/nanomachining [2,3]. A femtosecond laser induced periodic surface structure (LIPSS), also referred to as a ripple, can offer unique advantages and provide nanometer-scale precision [4–7]. Actually, a LIPSS is a universal phenomenon; and the near-subwavelength LIPSS is widely accepted to be the result of the interaction between the incident light and the surface scattering wave [8,9].
Laser-induced damage is complex [10–13], involving miscellaneous interactions between light and matter, such as laser-induced carrier excitation, carrier thermal ionization, and various thermal and structural events. Increasing capabilities for shaping light in terms of amplitude, phase, and polarization on the time scale of electron dynamics allows a variety of the physical properties of the complex processes to be regulated and processes to be routed in a prespecified direction [12]. Recently, the feasibility of controlling electron dynamics with a fs laser pulse train was theoretically proven [13], offering new possibilities for controlling the transient localized material properties to adjust phase-change mechanisms [14–16]. Many studies have been devoted to pulse train processing parameters, including pulse duration [17,18], the number of pulses per train [18,19], and pulse separation between subpulses [17,20]. More recently, pulse train shaping has been applied to adjust LIPSS formation. For example, Kim et al. studied the influence of the time separation of double femtosecond laser pulse trains on chromium (Cr) films [21]. Kim found that the formation of ripples and nanometer-sized particles could be controlled by time delays using double pulse trains. In addition, Bonse et al. pointed out that the orientation of the LIPSS can be determined by the first subpulse with low fluences and by the second subpulse with high fluences on fused silica upon multiple cross-polarized double-fs-laser-pulse irradiation sequences [22]. Furthermore, the authors have recently shown that the orientations and the periodicities can be controlled by a well-designed, parallel-polarized double fs laser pulse [20]. Such experimental studies with well-designed, double fs laser pulses offer a new method for obtaining controllable nanogratings and provide additional insight into the dynamics and formation mechanisms of a LIPSS. Nevertheless, most of the previous works are mainly devoted to the study of orientation and periodicity dependent on the time delay of the double fs laser pulse. Few studies have addressed the geometrical morphologies of a LIPSS, which is important to better control the ripple formation and further development for a wide range of applications in solar cells [4], waveguides [5], colorization [6], light-extracting surfaces in light-emitting diodes (LEDs) [23], surface-enhanced Raman scattering (SERS) [3], and water-repellent surfaces [24]. Since the ripple applications generally require large-area, uniform LIPSS fabrication, the scanning interval is of great importance. Systematic research of the geometric morphology of the LIPSS enables precise control of the laser scanned line width, which determines the scanning interval during laser-scanning-induced, large-area LIPSS formation. Recently, we reported the anisotropic geometrical morphology of a LIPSS with an ellipse-shaped ablated region under linearly polarized fs laser irradiation [25]. The directions of the major axes are perpendicular to the polarization directions.

In this study, the geometrical morphology of a LIPSS is investigated as a function of the double-fs-laser pulse delays ($\Delta t$) from 100 to 1600 fs. A transition from isotropic morphology to anisotropic morphology is observed when the pulse delay increases. The experiments demonstrate that it is feasible to control the anisotropy of the asymmetric LIPSS under linearly polarized fs laser irradiation by carefully designing pulse trains. This phenomenon is interpreted in terms of the carrier relaxation under the well-designed fs laser pulse train, which modulates the directional surface plasmon polariton (SPP)-laser energy coupling based on the initially formed ripple structures. Furthermore, based on the geometrical morphology control of LIPSS under a well-designed pulse train (double pulse delay), we show the anisotropy of the two-dimensional scanned line width modulations. Finally, we comprehensively illustrate the important role of the scanning interval being determined by the scanned line width on the large-area, uniform LIPSS formation.

2. Experimental setup

The experimental setup is shown schematically 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, a pulse duration of 50 fs, and a 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 fs laser pulse is temporally shaped to be a pulse train consisting of two subpulses with an accurate pulse delay, nearly equal energy distribution, and identical pulse duration between the subpulses by a pulse shaper (BSI MIIPS BOX 640). Meanwhile, the intensity measurement and the real-time spectrum analysis shows no spatial-temporal distortion of the fs laser after being shaped to double fs laser pulse train with different pulse delays. A 640-pixel liquid crystal spatial light modulator is inserted in the Fourier plane of a zero-dispersion stretcher; the detailed principle of pulse shaping is explained by Weiner [26].

The temporally shaped fs laser pulse train travels through the dichroic mirror (DM) and is focused through an achromatic doublet (f = 100 mm) onto the bulk of the sample. A 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 ambient pressure and temperature. After irradiation, the surface morphology is characterized by a scanning electron microscope (SEM). The ablated surface area is quantified subsequently from the corresponding micrographs.

3. Results and discussion

3.1. Continuous modulations of LIPSS geometrical morphology anisotropy based on double fs-laser-pulse delay

To explore the detailed dependence of the surface morphology on the double pulse delay (Δt), the silicon sample is irradiated by the double femtosecond pulse with parallel polarization (0°-polarization) direction. The pulse delay is systematically varied from 100 to 1600 fs. In all cases, the number of the irradiated pulses is 100; and the total average laser energy per train is 0.3 J/cm². The multipulse damage threshold of silicon is around $\Phi_{th} (N = 100) = 0.2$ J/cm² [27]. Therefore, the pulse energy here indicates that only the joint action of both subpulses can modify the surface permanently. Figure 2 shows the dependence of the dimensions (the lengths of the major and minor axes of the ablated ripple structure) and the ablated area (S) on the double-pulse delay (Δt). The inserts (a)-(e) present the corresponding data-based SEM images of LIPSS morphologies generated at various pulse delays. The LIPSS always appears...
as a grating-like distribution with the almost constant periods of 650–750 nm; and the orientation of the LIPSS structures are almost perpendicular to the laser polarization in all cases. The periods and directions of the LIPSS are consistent with the previous study under multiple single-pulse sequences irradiation [28]. Moreover, as the geometrical area marked by the dotted lines shown in the inserts (a)-(e), when the time delay increases from 100 fs to 1600 fs, the geometrical morphology of the LIPSS evolves from a nearly circular shape to a somewhat elongated elliptical shape. This anisotropic elliptical-geometrical morphology of the LIPSS under double pulse train irradiation can be also observed in [29], which explore the rippled area dependence on wide band gap materials after double pulse excitation. Actually, the ablated LIPSS region is surrounded by the heat-affected annular region; and the outer boundary of the heat-affected region always retains a circular shape when time delays are increased. Meanwhile, the dimensions and ablated area of the LIPSS decrease with an increase in the pulse delay, Δt, within a few picoseconds. At delays longer than 1.6 ps, the LIPSS vanishes completely. The dimensions evolvement can also illustrate that the decreasing of the minor axis is quicker than that of the major axis as the pulse delay increases, which gives the LIPSS an ellipse-shaped morphology. Moreover, the major axis of the ellipse-shaped region is along the polarization direction of the linearly polarized fs laser pulse. This preferential ablation effect has been observed recently on silicon under multiple single-pulse sequences irradiation, which the anisotropy is affected by the deposited pulse energy, and the directional SPP scattering along the polarization direction is proposed to interpret this effect [25]. In order to numerically represent the anisotropy evolvement on the time delay of the double fs laser pulse train, the parameter K (anisotropy) is defined 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 inset in the upper right-hand corner of Fig. 2 shows the anisotropy (K) of a LIPSS as a function of the time delays (Δt). The anisotropy parameter K undergoes a rapid decay from approximately 1 to nearly 0.4 when the time delay increases from 100 to 1600 fs, which indicates the aggregation of the geometrical anisotropy. This observation of the LIPSS morphology modulation signifies a strong influence by the time delays of the double fs pulse train on LIPSS structuring.

![Fig. 2. Dimensions (major and minor axes of the LIPSS), ablated area (S), and the anisotropy parameter (K) (upper right-hand corner inset) of the LIPSS as a function of double-fs-pulse delay at an energy of 0.3 J/cm². The number of irradiated laser pulses is 100 shots. The insets (a)-(e) show the SEM images at different time delays. All of the SEM images have the same scale bar.](image-url)
The fabrication of the LIPSS on silicon is considered to be caused by excitation of SPP at the air-silicon interface when the material turns from a semiconducting into a metallic state [8,9,30,31]. Upon high-intensity laser irradiation of silicon with fs laser pulses, electrons can be promoted from the valence (VB) to the conduction band (CB) via linear and nonlinear optical absorption effects. And the SPP excitation occurs when the carrier density reaches a critical value [32]. The process of ablation and damage can be described in terms of the excitation and relaxation of the conduction band electrons (CBEs). As a universal phenomenon in the field of laser ablation, the relaxation of CBEs (mainly carrier diffusion and Auger recombination) [30,31] generated by the first leading pulse of the double pulses is crucial to the whole ablation process and hence affects the resulting ablated rippled area. Here, the decrease in the continuously ablated area which occurs with the increase in the pulse delays can be ascribed to CBE relaxation. In the case of silicon, the bandgap is 1.14 eV; and electrons are mainly ionized in the conduction band through one-photon ionization and two-photon ionization [30]. A previous double-pulse experiment has shown that CBEs excited by the first pulse will relax back to the valence band during the time interval between the two pulses, in terms of diffusion and recombination [33]. Therefore, with the increase in pulse delay, the decreasing number of CBEs reduces the energy coupling by the second pulse to the transiently excited material, leading to the decay of the ablation, which results in the reducing of the rippled area. More recently, the CBEs generation and their relaxation is theoretically researched with sequences of double fs laser pulse, for large double pulse delays, carriers relaxation (mainly carrier diffusion and Auger recombination), which affect the SPP excitation, contributes to the rapid decrease of the ripple area [30,31].

Fig. 3. (a)-(c) SEM images of multiple-shot ablated LIPSS fabricated by different directions of linearly polarized fs laser with single pulse per burst. The pulse energy and pulse number are 0.25 J/cm² and 100, respectively. (d)-(f) and (g)-(i) SEM images of multiple-shot ablated LIPSS fabricated by double fs pulse under different polarization directions with the pulse delay of 300 fs and 600 fs, respectively; and the pulse energy and pulse number are identical with 0.3 J/cm², and 100, respectively. All of the SEM images have the same scale bar.

In order to comprehensively investigate the anisotropy modulation of the LIPSS at given pulse delays, a couple of double-fs-pulse-train-induced ripple structures are fabricated under different polarization directions. For comparison, LIPSS fabrication is also performed under single-fs-pulse irradiation with the same polarization directions as the double fs pulse train. As seen in Figs. 3(a)-3(c), the LIPSSs on silicon, which is irradiated by 100 linearly polarized fs laser pulses with 0.25 J/cm² laser energy, are fabricated with 0°, 45°, and 90°-polarizations. The ablated ripple structures are aligned by a preferential orientation along the polarization
directions, which exhibit elliptical morphologies. The anisotropy parameter $K$ under this condition almost remains unchanged at approximately $0.7 \pm 0.05$. In addition, in the case of double fs-pulse-train irradiation, as shown in Figs. 3(d)-3(i), LIPSSs are fabricated on the silicon sample by 100 double fs pulse bursts with 300 fs [Figs. 3(d)-3(f)] and 600 fs [Figs. 3(g)-3(i)] pulse delays. The average total laser energy per burst is $0.3 \text{ J/cm}^2$. At a pulse delay of 300 fs, the experimental result shows an isotropic polarization-independent geometrical morphology as compared with the structure irradiated at a single pulse per burst. As expected, during the variation of polarization directions, the LIPSSs always exhibit an isotropic circular geometrical morphology, which illustrates anisotropy elimination at a pulse delay of 300 fs. Nevertheless, with a pulse delay of 600 fs, as shown by the geometrical area marked by the dotted line in Figs. 3(g)-3(i), the geometrical morphology of a LIPSS exhibits an ellipse-shaped ablated region under the single fs pulse condition. Moreover, the anisotropy parameter keeps constant about $0.6 \pm 0.05$ at the energy of $0.3 \text{ J/cm}^2$.

After the formation of the initial ripple structures, which behave as a transient localized surface grating, it is expected that the interaction of the laser material surface would be changed, leading to the redistribution of the electric field that significantly affect the subsequent ablation process [25,34,35]. Zhang et al. demonstrated that the morphology of the initially formed ripple structures, which can be determined by the deposited laser energy, plays a crucial role in the subsequent electric field distribution [34]. Meanwhile, the elliptical-shaped ripple structures on silicon under fs laser irradiation is also observed to be affected by the irradiated pulse number, with a transformation to a rather circular feature as with the increasing deposition of the laser energy (Fig. 6 in reference [25]). In this study, the anisotropic geometrical morphologies are pronounced at large delays [Figs. 3(g)-3(i)], while with a nearly circular shape at short pulse delays [Figs. 3(d)-3(f)]. This anisotropic geometrical morphologies modulation effect based on the double pulse delay can be interpreted by the reducing deposited pulse energy as the increases of the double pulse delay. The free electron recombination strongly impacts the free electron density [19,30,31], which affects the resulted pulse energy absorption on surface of the sample. As mentioned above, the carrier relaxation increases with the increase of the pulse delay. Hence, the free electron density decays as the pulse delay increases, resulting in the reducing pulse energy deposition. The low deposited pulse energy results in the directional SPP-laser energy coupling based on the initially formed ripple structures. While, as the deposited pulse energy increases, another characteristic feature of grooves with an opposite orientation emerges on the surface of ripples. This cross-distributed structure is considered to weaken the directional SPP-laser energy coupling, leading to the weakened ellipticity of the LIPSS geometrical morphology. Here, in this study, for short pulse delay, the pulse energy deposited is high enough to form the cross-distributed ripple structure [Figs. 3(d)-3(f)], which weakens the directional SPP-laser energy coupling, leading to the resulted nearly symmetry circular LIPSS geometrical morphology [Figs. 3(d)-3(f)]. For long pulse delays, the reduced deposited pulse energy, results in the fabrication of one-dimensional pre-corrugated grating structures. The strengthened directional SPP-laser energy coupling due to the initially formed ripple structures lead to the elongated anisotropic geometrical morphologies. Thus, by designing the fs laser pulse train (double pulse delay), the continuous modulation of ripple structures can be obtained.

### 3.2. Anisotropy modulation of laser scanning two-dimensional LIPSS lines

The upgrade in material properties is a strong outcome of fs laser irradiation, especially the optical properties in LIPSS that enable material characteristics to be designed on mesoscopic scales. For example, color effect due to the diffraction of light by LIPSS has already been evidenced [6]. This technique proposes new applications for laser marking and new types of identifying codes. Meanwhile, the scanning technique is the main method for information storage, which has been known from the dawn of civilization. Nevertheless the precise control of LIPSS formation under laser scanning still remains a big challenge, since the optical properties of these structures are significantly dependent on beam properties, such as
laser polarization. For instance, Fauchet et al. manifested that the surface ripple structures on crystalline or ion-implanted semiconductors exhibit an incident laser wave-front sensitivity under picosecond laser scanning [36]. Here, based on the aforementioned experimental results, we designed a double-fs-laser-pulse-train scanning experiment (Fig. 4) aimed at precisely modulating the polarization-dependent anisotropic effect on laser scanned linewidth in different scanning directions. Each line is scanned with only one pass, in one direction. This adjustable anisotropy of scanned linewidths along different scanning directions under different time delays provides further control over the differently oriented scanning LIPSS lines. Figure 4 furnishes the anisotropy modulations of the mutually perpendicular LIPSS lines with a pulse delay of 300 fs [Figs. 4(a)-4(b)] and 600 fs [Figs. 4(c)-4(d)]. The scanning directions are given in the SEM images. And the polarization directions are 40° in Figs. 4(a)-4(b) and 90° in Figs. 4(c)-4(d), respectively. The pulse energy and repetition rate are 0.3 J/cm² and 250 Hz, respectively. The scanning speed is 100 μm/s and 200 μm/s in Figs. 4(a), 4(c) and Figs. 4(b), 4(d), respectively. As expected, the LIPSS lines in Figs. 4(a)-4(b) exhibit isotropic widths in the two mutually perpendicular directions for the isotropic LIPSS morphology under static fs laser irradiation. The cross-section marked by the dotted line is consistent with the one by static laser irradiation due to the large laser energy accumulation at the cross-section. The ratio of horizontal writing width to vertical writing width (H/V) is nearly 1 in each scanning speed case. Remarkably, the different widths shown in Figs. 4(a)-4(b) illustrate that it can be controlled by the scanning speed. This anisotropy depression effect can be used to eliminate polarization sensitivity under different scanning directions. As a comparison, the characterized scanning LIPSS lines in Figs. 4(c)-4(d) exhibit anisotropic width with the mutually perpendicular orientations. As shown, the ratio of the H/V is nearly 0.6 ± 0.05, which is consistent with the aforementioned anisotropic geometrical morphology of the LIPSS under static fs laser irradiation.

3.3. Pulse delay dependence large area LIPSS fabrication

Although the formation of LIPSS is a remarkable phenomenon in laser processing, how to precisely control the fabrication of large-area, uniform LIPSS is an essential challenge. As an important parameter, the scanning interval (interval of two adjacent scanning lines) is of great importance in the formation of large-area, uniform ripple structures, which definitely affects
the continuity of the ripples and the processing efficiency. Here, based on the above modulated scanning LIPSS lines technique, the large-area ripple structures can be produced by a simple scanning technique with a fixed scanning interval \((d_1, d_2)\), as the schematic inserts shown in Figs. 5(a) and 5(b). At the fixed pulse energy, repetition rate, and identical scanning directions in the aforementioned experiment, the 1 \(\times\) 0.6 mm\(^2\) area can be produced with various processing times when the pulse delay increases from 100 to 1,600 fs with 0° and 90°-polarization, respectively. Scanning speed is 100 \(\mu\text{m/s}\). Processing efficiency is also related to the polarization directions. As shown in Fig. 5, when the pulse delay is short, the scanning time is almost identical, which indicates an isotropic property. Whereas, it can be found that, the growing of the processing time with 90°-polarization is quicker than that with 0°-polarization as the pulse delay increases. This phenomenon can be understood as the anisotropy scanning line widths effect with pulse delay, as shown in the schematic diagram in Figs. 5(a) and 5(b). Therefore, the processing efficiency and precision can be improved by a well-designed pulse train and the polarization directions, which may add an extra freedom for the large-area LIPSS fabrication.

4. Conclusion

In summary, the geometrical morphology evolution of LIPSS on silicon by double fs laser pulse train has been investigated. The pulse delay can significantly change the polarization-dependent anisotropic morphology of LIPSSs: isotropic morphology is transited to anisotropic morphology as the pulse delay increases from 100 to 1,600 fs. The modulated directional SPP-laser energy coupling by the pulse energy deposited based on the free electron density control under the well-designed fs laser pulse train (double pulse delay) is proposed to interoperate this phenomenon. Furthermore, the polarization sensitivity scanning LIPSS lineswidth modulation under different scanning directions is demonstrated based on the geometrical morphology control of LIPSS under a well-designed pulse train (double pulse delay). As an example, we illustrate the fabrication of large-area uniform LIPSSs based on the anisotropic scanning line width control.

Acknowledgments

This research is supported by the National Basic Research Program of China (973 Program) (Grant 2011CB013000) and National Natural Science Foundation of China (NSFC) (Grant No. 90923039 and 51305030).