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Femtosecond laser-induced cross-periodic structures on a crystalline silicon surface under low pulse number irradiation

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ABSTRACT

A cross-patterned surface periodic structure in femtosecond laser processing of crystalline silicon was revealed under a relatively low shots ($4 < N < 10$) with the pulse energy slightly higher than the ablation threshold. The experimental results indicated that the cross-pattern was composed of mutually orthogonal periodic structures (ripples). Ripples with a direction perpendicular to laser polarization ($R_{\perp}$) spread in the whole laser-modified region, with the periodicity around 780 nm which was close to the central wavelength of the laser. Other ripples with a direction parallel to laser polarization ($R_{\parallel}$) were found to be distributed between two of the adjacent ripples $R_{\perp}$, with a periodicity about the sub-wavelength of the irradiated laser, 390 nm. The geometrical morphology of two mutually orthogonal ripples under static femtosecond laser irradiation could be continuously rotated as the polarization directions changed, but the periodicity remained almost unchanged. The underlying physical mechanism was revealed by numerical simulations based on the finite element method. It was found that the incubation effect with multiple shots, together with the redistributed electric field after initial ablation, plays a crucial role in the generation of the cross-patterned periodic surface structures.

1. Introduction

Laser-induced periodic surface structures (LIPSS, also referred to as ripples), were first observed on germanium by Birnbaum in 1965 [1]. After that, ripples were produced on a range of solid materials, including metals, semiconductors and dielectrics under the irradiation of nanosecond, picosecond or continuous laser fields at various wavelengths [2–5]. Recently, the ripples induced by a femtosecond (fs) laser have become more attractive due to the wide range of potential applications in many fields, such as solar cells [6], colorization [7–9], and waveguides [10]. In most of these applications, the formation of well-defined, uniform submicroscale even nanoscale ripples is needed.

In general, according to the periods of ripples and the incident laser wavelength (λ), LIPSS can be divided in to two distinct types, low spatial frequency LIPSS (LSFL) and high spatial frequency LIPSS (HSFL) [11,12]. LSFL consist of spatial periods (Λ) close to the irradiation wavelength ($\lambda > \Lambda > \lambda/2$), which are widely considered to be the result of the interaction between an incident wave and a surface scattered wave [2,3,5]. Additionally, Bonse et al. reported the surface plasmon polaritons (SPP) play a dominant role during the initial stage of near-wavelength-sized periodic surface structures in fs-laser irradiated silicon [13], and the formation of a periodic surface relief can be reinforced by a resonant excitation of SPPs in the grating like LSFL, leading to a red shift in the SPP resonance and consequently to a decrease in the LSFL period with an increasing number of laser pulses [14]. The spatial periods of HSFL are significantly smaller than the irradiation laser wavelength ($\Lambda < \lambda/2$) and their formation mechanism is still under investigation [12,15–18]. So far, several mechanisms have been proposed to explain the formation of HSFL induced by fs laser pulses, such as self-organization [19], second harmonic generation [12,17,20] and Coulomb explosion [21], etc. Very recently, we investigated shaped fs laser-pulse-induced double-grating including HFSL structures adjustments based on electron dynamics control [22]. Moreover, compared to previous studies that showed interesting results about the HSFL formation at a high repetition rate or in a water environment [15,20,23], a few works related to the formation of HSFL have been carried out on silicon by using a fs laser in ambient air at a low repetition rate. Recently, the effects of the initially formed structures under fs laser ablation on the surface of the materials

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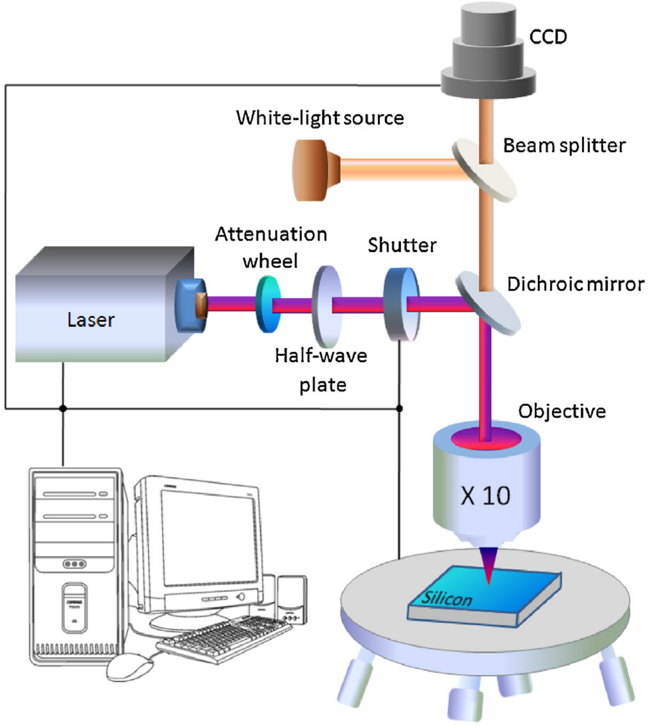

Fig. 1. Schematic diagram of the experimental setup for fabrication of cross-periodic structures on the surface of silicon.

have been investigated. It was expected that the initially formed structures would significantly influence the field intensity distribution of the incident laser, which in turn could affect the subsequent ablation process [24].

In this paper, we report on a type of surface nanostructure on the surface of crystalline silicon, which consists of two mutually perpendicular ripple structures (LSFL and HSFL), namely laser-induced cross-periodic surface structures (LICS). The geometrical morphology of LICS generated by static fs laser irradiation was subject to the laser polarization direction, whose changes could lead to continuous rotation. It was found that the orientation of LSFL was always perpendicular to the laser polarization direction ($R_\perp$), whereas, the orientation of HSFL was parallel to the laser polarization direction ($R_\parallel$).

2. Experimental details

The schematic of the experimental setup is depicted in Fig. 1. In our experiments, the laser source was a Ti: sapphire laser regenerative amplifier system, which provided a fundamental Gaussian mode with a central wavelength of 800 nm, a pulse duration of 50 fs, and a repetition rate of 1 kHz. The intensity measurements reveal that the laser beam is close to the symmetric Gaussian distribution, and no significant changes were observed by varying the polarization directions. A continuous attenuation wheel (a neutral density filters wheel) was used to control the laser fluence incident on the sample surface. The half-wave plate was used to change the polarization direction of the incident laser pulses. The laser pulse number delivered to the sample was controlled by a fast mechanical shutter synchronized with the laser repetition rate. The laser light with the diameter ~3.5 mm traveled through the dichroic mirror (DM) and was focused through a 10× microscope objective (Olympus, NA = 0.3, the focal length ~10 mm) on the surface of the sample and the beam spot size is calculated to be about 3 µm. The highly polished silicon (1 1 1) sample (10 mm × 10 mm × 1 mm) was mounted on a computer-controlled, six-axis moving stage (M-840.5DG, PI, Inc.) with a position 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 was used to image the sample surface. All experiments were carried out in air at room temperature. After irradiation, the sample was treated by ultrasonic bath for 5 min both in deionized water and acetone in order to clean the residual ejections of material off of the surface. Morphological characteristics were investigated by a scanning electron microscope (SEM) and atomic force microscope (AFM).

3. Results and discussion

3.1. Cross-periodic surface structures evolution with the increasing of fluences

Fig. 2 shows SEM images of the cross-patterned periodic surface structures induced by five fs laser pulses irradiation on the surface of the silicon. It can be seen that the morphology of the LICS exhibited a strong dependence on laser fluence (*F*). In this study, the threshold fluence (F_{th}) is determined as the minimal fluence for a five pulses that just creates damage on the silicon surface observed by SEM system. Hence, the threshold fluence (F_{th}) about 0.185 J/cm^2 was obtained after our measurement. It is slightly lower compared to the value of 0.2 J/cm^2 reported previous study by Bonse et al. [25]. They studied that laser-induced modification and ablation of silicon surfaces with laser pulse duration in the range between 5 fs and 400 fs and found several physical processes resulting in clearly distinguishable morphological features with different fluences and pulse number [25]. For a laser fluence smaller than 0.19 J/cm^2, no distinct nanostructures were produced, as shown in Fig. 2a. The dark region belongs to annealing or modified region, which is probably oxidation or amorphization silicon by fs laser ablation according to Bonse et al. studies [25]. When *F* reached 0.2 J/cm^2, one row comprised of two nanoholes was fabricated, as shown in Fig. 2b. The nanoholes appeared to be circular in shape with a diameter of ~150 nm. The spacing of the two adjacent nanoholes was measured and was about a half of a laser wavelength, ~400 nm, which represents the formation of HSFL. When the laser fluence, *F*, was increased to 0.22 J/cm^2, the nanoholes became larger and deeper because the incident light was mainly localized in the nanoholes, as shown in Fig. 2c. With a further increase in *F* to 0.25 J/cm^2, we observed a nanohole array composed of three rows separated by ~780 nm, which was slightly smaller than the laser wavelength. This means LSFL was initially produced, which can be identified in Fig. 2d. When *F* was raised to 0.28 J/cm^2, we obtained a distinct nanohole array comprised of four nanohole chains, as shown in Fig. 2e. In fact, it apparently revealed that the morphology of LSFL spread in the whole irradiation area, and HSFL was just produced in two neighboring LSFL ripples. When the laser fluence was larger than 0.3 J/cm^2, we just obtained LFSL; and the HFSL between two LSFLs vanished, the LICS pattern evolved into LIPSS structures, as shown in Fig. 2f. Fig. 2g illustrates the period and ablation depth of HSFL with different laser fluences. As can easily be seen, the period almost keeps constant; and the depth of the ablation region increases nearly linearly with the corresponding increases in the laser fluence. For example, the average depth of the ablation area is 50, 70, 100 and 150 nm for 0.2, 0.22, 0.25 and 0.28 J/cm^2, respectively.

3.2. Polarization dependence of LICS

It has been shown that the alignment of the ripples depends strongly on the polarization of the laser light [26,27]. When we rotated the laser polarization from $\theta = 0°$, $\theta = 45°$, $\theta = 90°$,