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Femtosecond laser-induced periodic surface structure on fused silica surface

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ABSTRACT

Single and 10-pulse laser-induced surface damage behaviors of fused silica were investigated using a linearly polarized 40-fs Ti: sapphire laser system operating at a central wavelength of 800 nm. After the sample was subjected to single-shot laser irradiation, it displayed a typically damaged crater, and two types of laser-induced periodic surface structures (LIPSS) on the surface of the damage crater were found under 10-pulse laser irradiation. By increasing the laser fluence, the LIPSS orientation rotated by 90°, and the period changed from 300 nm to 700 nm. Under near-threshold laser irradiation, an intermittent LIPSS, whose orientation was perpendicular to the laser beam polarization direction and average spatial period was around 300 nm, was found on the damaged area. Another type of LIPSS, which was parallel to the laser beam polarization and had a period of approximately 700 nm, was continuously formed at the bottom of the ablation crater when the laser fluence exceeded the damage threshold of material. A model based on the theory of interference between the incident laser and the surface scattered wave was adopted to explain the formation mechanism of the LIPSS.

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1. Introduction

With the development of femtosecond (fs) laser system, numerous practical applications have been developed in fields that involve material removal with submicron precision, such as micromachining, electronics, and data storage. The formation of laser-induced periodic surface structures (LIPSS) has gained significant attention in the past few decades [1–16] because of its precise micromachining with nanometric resolution. In previous publications, two different types of LIPSS, the low spatial frequency LIPSS (LSFL) [1,3–6,17] and the high spatial frequency LIPSS (HSFL) [1–6], were discovered on fused silica surface. These distinct types of LIPSS were either parallel [1,3–5] or perpendicular [1–5,17,18] oriented to the laser beam polarization direction with a linearly polarized fs laser irradiation in air under normal incidence. The period of the LSFL is close to or somewhat smaller than the laser wavelength, whereas HSFL often have spatial periods that are significantly smaller than the laser wavelength. Höhm et al. [1] found that HSFL will transform into LSFL with the increase of laser fluence

on quartz surface under 10-pulse fs laser irradiation, but the critical fluence was significantly higher than the laser-induced damage threshold (LIDT). With fs laser pulse train irradiation, Jiang et al. [6] found that LSFL will transform into HSFL with different laser delay time on fused silica surface. The formation mechanisms of LIPSS were still controversially discussed in the literature, and different models, such as interference [1,19–21], self-organization [8,9], second harmonic generation (SHG) [22,23], excitation of surface plasmon polaritons [24,25], and Coulomb explosion [26], have been proposed. Therefore, the major question was open in the area of LIPSS, which required more experimental and theoretical studies.

In this study, the surface damage behavior of fused silica under single and multi-pulse fs laser irradiation was explored, and an interesting LIPSS phenomenon generated by 10-pulse fs laser irradiation was found. With the increase of laser fluence, the average period of the LIPSS was changed from 300 nm to 700 nm, and the orientation of the LIPSS was changed from a perpendicular orientation to a parallel orientation compared with the laser polarization direction. In other words, HSFL will transform into LSFL with the increase of laser fluence under 10-pulse laser irradiation, and the transition fluence is close to the LIDT of fused silica. More significantly, the LIPSS evolution on fused silica surface results from an interference process.

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2. Experiment

A commercial chirped pulse Ti: sapphire fs laser system was used to generate a linearly polarized laser pulse with pulse duration of 40-fs and central wavelength of 800 nm [27]. A mechanical shutter isolated necessary pulses from the output of 1000 Hz pulse train. A neutral density filter functioned as the variable attenuator to regulate the pulse energy, and the energy was detected by an energy meter from a split-off portion of the beam. The effective spot diameter ($1/e^2$) employed was close to 150 μm , which was measured by a beam analyzer. The laser beam was normally irradiated on the surface of the sample in the atmosphere. The damage process was monitored in situ with a surface image system.

Experiments were performed on 1-cm thick fused silica samples that exhibited less than 0.5 nm rms surface roughness after an ultrasonic bath. The single-pulse damage test was achieved by irradiating only one pulse onto one sample site with 10 sites tested under one fixed fluence step. For the multi-pulse experiment, 10 pulses with fixed laser fluence were irradiated onto the sample. Damage in this study was defined as irreversible modification detected by the Leica optical microscope (magnification: 100 \times). After laser irradiation, the damage morphology was observed by the field emission scanning electron microscope (FE-SEM, Zeiss Auriga S40). The depth profile of the sample was tested using an optical profiler (Wyko NT9100, Bruker).

3. Results and discussion

The LIDT of fused silica under fs laser irradiation was determined by measuring the size of the crater versus the pulse fluence and then extrapolating to zero based on the relation of the crater area and the laser fluence, which was similar to the ablation threshold fluence [28]. Fig. 1 shows the relationship between the crater area and the laser fluence, and the abscissa intercept of the fitting line

is the LIDT. The relative error of LIDT determination is estimated at 10%, which is mainly caused by the uncertainty of the crater size measurement and the fluctuation of laser energy. Therefore, the LIDTs of fused silica under single and 10-pulse fs laser irradiations are $\sim 2.11 \pm 0.21 \text{ J/cm}^2$ and $\sim 1.06 \pm 0.11 \text{ J/cm}^2$, respectively.

The surface morphology of the damage crater was measured to determine the characterization of fs laser damage. Fig. 2 shows the SEM images and horizontal cross-section profiler information of the fused silica surface after single fs laser pulse irradiation with peak fluence of 7.06 J/cm^2 . The damage morphology of the whole crater is illustrated in Fig. 2(a). The figure shows that the damage phenomenon is very slight, and only an annular rim can be observed around the crater. SEM image at higher magnification focusing on the edge of the crater reveals that the crater is very rough (Fig. 2(b)). The depth profiler shows that the maximum depth of the crater is approximately 135 nm (Fig. 2(c)).

Fig. 3 shows the SEM images of fused silica surface after 10-pulse fs laser irradiation with different laser fluences. The LIPSS are generated on the surface of the damage craters. The variation law of the period and orientation of the LIPSS versus the laser fluence is shown in Fig. 4. The general fs laser damage craters (Fig. 3(b1 and c1)) are generated when the laser fluences exceed the LIDT, whereas a special damage morphology (Fig. 3(a1)) is found when the laser fluence decreases to 1.01 J/cm^2 . Such damage behavior is slight and only occurs on the surface layer of the sample. The crater consists of an intermittent LIPSS (HSFL) (Fig. 3(a2)), whose orientation is perpendicular to the laser beam polarization and average spatial period is around 300 nm. Intermittent HSFL are also found around the crater when the laser fluence is 1.10 J/cm^2 (Fig. 3(b2)) and will disappear if the fluence is even higher. Meanwhile, another type of LIPSS (LSFL) is found on the surface of the crater when the laser fluence exceeds the LIDT (Figs. 3(c2) and 4). Such type of LIPSS is continuously formed at the bottom of the crater. The orientation of this LIPSS is parallel to the laser beam polarization, and the average spatial period is approximately 700 nm. HSFL are generated at the onset of the damage, and HSFL will transform into LSFL when the laser fluence exceeds the LIDT. The orientation and the period of the two types of LIPSS are not the same.

In the experiments conducted in this study, the two types of LIPSS have different orientations, but the average spatial periods of the LIPSS are comparable with the laser wavelength. Thus, the phenomenon can be analyzed with the theory based on the interference between the incident fs laser and some type of surface-scattered electromagnetic wave [19,29]. This theory can be simply explained as follows. The first few laser pulses damage the sample and produce a rough surface. When the subsequent pulses reach the damaged surface, the rough surface will scatter the incident fs laser. The interference between the incident fs laser and the scatter light will result in the generation of the regions with periodic greater or lower amplitude (LIPSS). This interference effect will lead to inhomogeneous absorption energy within the bulk of the material. If this absorption energy exhibits sharp peaks, the LIPSS can be formed. Based on the theory of Sipe et al. [19] and Bonse et al. [20], the inhomogeneous absorption energy A can be quantitatively calculated to verify the period and orientation of the LIPSS as follows:

$$A(\mathbf{k}) \propto \eta(\mathbf{k})|b(\mathbf{k})| \cos(\delta) \quad (1)$$

where $\mathbf{k}(|\mathbf{k}| = 2\pi/\Lambda)$ is the wave vector of the LIPSS, Λ is the period of the LIPSS, and δ is the phase difference. The efficacy factor that describes the efficacy with which the surface roughness at \mathbf{k} leads to absorption is denoted by η , and b represents a measure of amplitude of the surface roughness at \mathbf{k} [19,20]. Interference theory indicates that the efficacy factor η can exhibit sharp peaks at certain LIPSS wave vectors \mathbf{k} values.

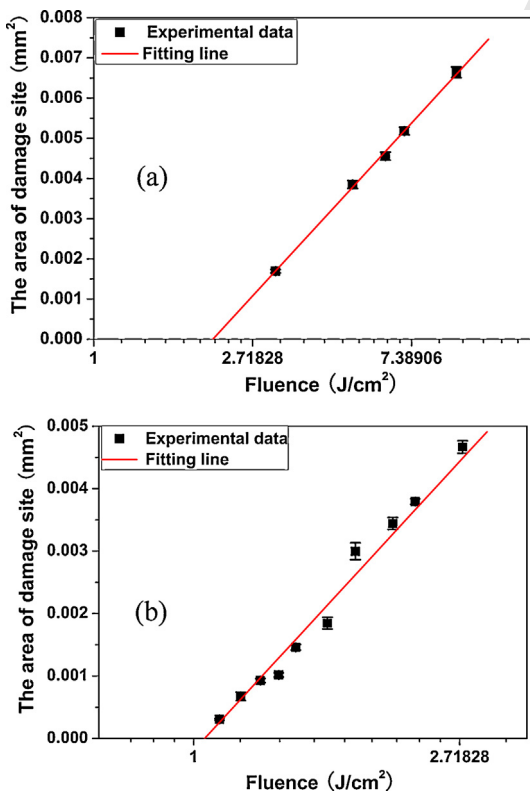


Fig. 1. Graphs for the determination of the LIDT from experimental data. (a) 1-on-1; (b) 10-on-1.

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