Femtosecond laser induced damaging inside fused silica detected by a single-pulse ultrafast measurement system

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Abstract: The dynamics of damage inside the fused silica induced by a femtosecond laser has been characterized by a single-pulse ultrafast measurement system that is built upon the pump-probe mechanism. Our investigation disclosed the quantitative relationship between the size of the damaged area and the pulse energy. The dynamic measurement experiments showed that the radial size of the damaged area increased rapidly from 0 to 21 µm within ~10 ps before stabilizing at 21 µm with the pulse energy of 1.1 mJ, which follows the rule of Boltzmann function. Moreover, we demonstrated that the structure inside the damaged area kept changing for about 200 ps before the formation of a double-void structure. The developed system alongside the proposed analysis method is expected to be of great importance in understanding the dynamics of laser-induced damage process in laser micromachining.

1. Introduction

Over the past decades, femtosecond laser-based micromachining has played an important role in the field of micromanufacturing and material processing [1–4]. The strong electromagnetic field associated with the femtosecond laser pulse-matter-interaction can be utilized to remove or change both transparent [3] and opaque materials [5]. For a transparent material, a series of linear and nonlinear effects, including diffraction, group velocity, free-carrier absorption, plasma defocusing, multiphoton absorption and self-focusing, will occur in the foci. The material in the foci absorbs the photon energy, after which electrons transfer from the valence band to the conduction band, leading to a strong plasma in the material. As a result, permanent damage is generated in the foci [6]. The feature of changing the local properties of transparent materials makes femtosecond laser popular in diverse fields, including optical memories, color centers, waveguides, and diffractive optical elements, etc. [7–9]. Typical structural damage induced by femtosecond lasers in transparent materials includes filaments and voids, the formation of which depends on the focusing conditions and the pulse energy. Filamentous structures can be generated along the direction of optical axis via the effect of self-focusing and plasma defocusing at the condition of weak focusing for high-energy pulse input [10–12]. Conversely, voids are generated in the foci due to the micro-explosion when the laser pulse with lower energy is tightly focused inside the material [13–15].

As the growing demand of laser writing, drilling, and cladding is stimulating global demand for high-precision laser manufacturing tools, understanding the dynamics and mechanism of damage formation alongside its morphology is of great importance. To characterize the ultrafast dynamics of the damage inside the transparent material, the ultrafast shadow imaging method, the interferometric method, and the laser holography method were reported [16,17]. The pump-probe-based ultrafast shadow imaging method has been utilized in the detection of the dynamic...
evolution process of plasma [18–21], shock waves [22–24], the filamentous structure [25,26], optical breakdown [18,26], and the spatial distribution of refractive index inside transparent materials [27,28]. In order to obtain the phase shift of the probe light resulting from passing through the damage region, the interferometric imaging method has been proposed [18,29]. Y. Hayasaki et al. used the time-resolved interferometer to investigate the dynamics of the probe light phase shift and the spatial distribution of the refractive index change in the damage area [30–32]. In addition, the laser digital holography method has been used to measure the spatiotemporal distribution of the laser-induced free-electron and the refractive index change in transparent material induced by ultrastar laser [33,34]. These methods provide abundant information on the laser induced damage in transparent materials through repeated measurements. However, these discrete measurement methods cannot capture the continuous dynamic evolution of the damaged area due to inevitable fluctuation of the loading condition and defects in the material. Recently, Y. Yao et al. proposed an ultrafast photography with a hyperspectral camera that enables picosecond-level temporal resolution and nanometer-level spatial resolution of dynamic measurements for ultrastar processes within a single-shot [35]. At the same time, we have demonstrated a time-resolved pump-probe technique based on diffraction which is capable to characterize the dynamics of femtosecond laser-induced plasma within a single measurement by fitting of diffraction fringes [36], which inspires us to utilize it to observe the dynamic damaging process inside the materials. Compared with the ultrafast photography strategy, whose temporal resolution is usually limited by spectral band numbers of the hyperspectral camera, the proposed method sacrifices one-dimensional imaging information, but it is easier to obtain higher temporal resolution.

In this paper, we implemented a set of experiments and found a double-void structure with cracks and filamentary modification in the damaged area inside the fused silica, and further made the efforts to disclose the quantitative relationship between the size of the damaged area and the pumping pulse energy. Furthermore, the measured spatiotemporal evolution of the femtosecond laser induced damage inside the fused silica revealed that the temporal evolution of damage size conforms to the law described by the Boltzmann function. The staged evolution characteristics of the damaged structure were revealed, that is, the radial dimension of the damage increased from 0 to 21 $\mu$m in $\sim 10$ ps and finally stabilized at 21 $\mu$m, while the formation of the void took about 200 ps.

2. Experiments

Details of the single-shot time-resolved experimental setup has been described in our recent work, which acquires the plasma dynamics by fitting of diffraction fringes [36]. Differently, in this work the spatial-temporal resolved dynamics of the single pulse-induced damage can be directly obtained through the measured spectrum without fitting process. As shown in Fig. 1(a), Gaussian limit pulses with the duration of 35 fs and the central wavelength of 800 nm are delivered from a Ti: sapphire laser (Newport Corporation, SOL-ACE35F1K-HP). The repetition rates of the output pulse of the system are set as 10 Hz. The spectral width of the output beam is 31.7 nm. In the system, the output femtosecond laser is split into two beams (pump and probe) by a non-polarization beam splitter. A beam expander (GBE05-B, Thorlabs) in the pump arm was used to change the size of the output beam. A mechanical shutter (SH1/M, Thorlabs) with open time of 0.1 s was used to ensure that only one 35 fs pulse was used in a single measurement. The pump beam is used to induce the damage inside the fused silica through a 50× high power focusing objective with a damage threshold of 15.0 J/cm$^2$ (LMH-50X-850, 0.65 NA, Thorlabs), which is safe under the pulse energies used in the experiments. The probe beam is a picosecond linear chirped pulse that obtained through a home-made grating-based pulse stretcher. The linear relationship between wavelength $\lambda$ and time $t$ of the stretched pulse can be expressed as $\lambda = 0.1124*t+786.2$, where 0.1124 is the linear chirped coefficient with unit of
nm/ps. The details of the pulse stretcher can be found in our previous work [37]. The size of the surface-polished fused silica sample is 6 mm × 6 mm × 1.5 mm. The sample was mounted on a three-axis translation stage. The focal plane was set at 300 µm below the front surface of the fused silica. An achromatic lens L1 (ACA254-075-B, Thorlabs) with focal length of 75 mm is employed to focus the probe beam, resulting in a spot size about 190 µm at the detection point, which completely covers the diameter of the damaged area, as shown in Fig. 1(b). Through another achromatic lens L2 (ACA254-050-B, Thorlabs) with focal length of 50 mm, the damage is imaged onto the slit (16 µm width) of an imaging spectrometer (Horiba, iHR550), and finally a spatiotemporal spectrum will be recorded on the CCD detector (Horiba, Syncer-1024×256). With the above configuration, the detecting spatial range is ~180 µm and the detecting time range is ~282 ps in a measurement. The time resolution of the system is ~0.28 ps.

![Diagram](image_url)

**Fig. 1.** (a) The schematic diagram of the optical configuration. (b) Schematic diagram of the pump-probe mechanism in the damage area and the temporal spatial spectrum acquired from a single chirped pulse. NPBS: non-polarized beam splitter; P: polarizer; FW: neutral filter wheel; L: lens; M: mirror; S: mechanical shutter; BE: beam expander; O: objective (50×).

### 3. Results and discussion

#### 3.1. Characteristics of the laser-induced damage inside the fused silica

The interaction between the femtosecond laser and the material will lead to irreversible damage to the material when the laser pulse energy exceeds the damage threshold. Here, a femtosecond laser pulse was focused 300 µm beneath the front surface of the fused silica by a focusing objective with NA = 0.65. Figure 2 presents the optical images of the damaged area inside the fused silica observed by an optical microscope with the input pulse energy of 0.58 mJ, 1.1 mJ, 1.64 mJ, 1.9 mJ, 2.16 mJ, and 2.36 mJ. The red arrow denotes the propagating direction of the laser pulse. It can be seen that a double-void damage was formed in the foci. Three types of damage morphology can be found in the damaged area, i.e., the voids, the cracks, and the filamentary
damage. To analyze the formation of such a composite damage morphological feature, it is important to consider nonlinear propagation effects in the transparent medium. The critical power for self-focusing in fused silica was reported to be $\sim 4.2 \text{ MW}$ [38]. Here, the peak power value for the lowest input pulse energy used in the experiment (0.58 mJ) is calculated to be 15.9 GW, which has taken the Fresnel losses ($\sim 4\%$ [39]) into account at the air-silica interface. Obviously, self-focusing effect will appear during the pulse propagation in the fused silica. Therefore, the weak balance between the self-focusing and the plasma defocusing effect can be considered as one of the mechanisms leading to the phenomenon of multi-focus along the optical axis [40], which results in the formation of two voids and filamentous channel on the propagation path. As for the cracks around the voids and the filamentary damage, two rational interpretation can be considered. On the one hand, the cracks could be caused by the plasma heating and secondary effects of volumetric material ablation induced by the high laser pulse intensities used in the experiments. On the other hand, the thermal effect of the plasma and the stress waves generated during the formation of voids would lead to the generation of cracks inside the material. In addition, it is worth mentioning that the damage shown in Fig. 2 is not strictly symmetrical along the direction of pulse propagation, which may be caused by the slight misalignment of the sample surface and the optical axis of the objective.

![Fig. 2. The optical images of the damaged area inside the fused silica induced by a single laser pulse with input energy of 0.58 mJ, 1.1 mJ, 1.64 mJ, 1.9 mJ, 2.16 mJ and 2.36 mJ.](image)

Furthermore, the relationship between the size characteristics of the damage and the input pulse energy was discussed. Figure 3(a) and 3(b) present the pulse energy dependence of the radial size $d$ and the length $L$ of the damaged area. The black balls are the measured data and the red curves are the fitted results with $R_0^2 = 0.975$ and $R_1^2 = 0.989$, respectively. The corresponding fitting formulas have been given in the figure. The results indicates that the radial size and the length of the damage increases linearly with the increase of pulse energy within the measurement range. Comparing the changes of the radial size and the length of the damaged area, it can be seen that the length changes faster than the radial size. The intercept of the fitted straight line indicated that the linear fitting results are only applicable to the energy range presented in this work. It can be speculated that when the pulse energy drops to a certain value, there exists a sharp drop of the damage size, and no damage would occur when the energy drops below the damage threshold. Furthermore, the pulse energy dependence of the distance between the two voids $\Delta L$ were analyzed, as shown in Fig. 3(c). The red curve is the result of Boltzmann fitting of $\Delta L$ with $R^2 = 0.994$. The corresponding fitting formula has been given in the figure. Interestingly, the relationship between $\Delta L$ and the pulse energy conforms to a law described by the Boltzmann function under the pulse loading conditions in this work. As the energy increases, it increases slowly in the range of 0.58 mJ $\sim 1.64$ mJ at first, then increases rapidly in the range of 1.64 mJ $\sim 2.16$ mJ. Finally, the growth rate decreases and $\Delta L$ tends to stabilize after 2.16 mJ. Although missing the physical meaningful insights, the relationship achieved can be useful in controllable manufacturing. The relationship between the size characteristics of the damage and the pulse
energy can provide us an approximate quantitative solution of the input pulse energy to generate the expected damage structure with specific size.

Fig. 3. The pulse energy dependence of (a) the radial size $d$, (b) the length $L$ and (c) the distance between the two voids $\Delta L$ of the damaged area. The black balls are the measured data, and the red curves are the fitting results.

3.2. Dynamics of the damage

What we have shown above is the morphological characteristics of the permanent damage inside the fused silica induced by the femtosecond laser. In this section, we will discuss the formation dynamics of such damage. The static spectrum without pump and the dynamic spectrum under pump ($E_{in} = 1.1$ mJ) were recorded on the CCD camera, as shown in Fig. 4(a) and 4(b), in which the vertical and the horizontal axes represent the spatial dimension and the wavelength dimension, respectively. The changes in the spectrum caused by the formation of damage can be clearly seen in Fig. 4(b). In order to minimize the effect of the background noise in the dynamic spectrum and obtain higher signal-to-noise ratio, the spatiotemporal spectrum that acquired through the difference of the static spectrum and the dynamic spectrum was shown in Fig. 4(c), where $\Delta I = I_1 - I_0$. The spatial dimension has been converted into the actual size and the wavelength dimension has been mapped into the time domain through the linear relationship of the chirped pulse. The moment $t = 0$ in the spectrums is defined as the position when the damage appears. The dynamic process of the damage development inside the fused silica can be observed simultaneously from the spatial and temporal domain in Fig. 4(c). Moreover, for closer inspection of the initial stage of the damage evolution, which reflects the dynamic increase of the radial size of the damage area, the initial stage presented in Fig. 4(c) has been zoomed in, as shown in Fig. 4(d). Results show that it takes about 10 ps to form the damaged area with a stable radial size. Moreover, it can be observed in Fig. 4(c) and 4(d) that a negative absorption stripe appeared in the central row of the spectrum. The appearance of this stripe can be attributed to the generation of plasma and the formation of void structure. Details on the temporal evolution of stripe intensity are presented and discussed later in Fig. 6.

The details of the dynamic evolution of the radial size were extracted from the measured spectrum, as shown in Fig. 5(a). The black balls were the measured data and the red curve represents the Boltzmann function fitting result with $R^2 = 0.998$. The function has the following expression,

$$d = \frac{d_1 - d_2}{1 + \exp((t - t_0)/a)} + d_2, \tag{1}$$

where $d_1$ and $d_2$ are the final value and the initial value of the radial size, respectively. $t_0$ represents the time when $d = (d_1 + d_2)/2$, and $a$ is a constant. It can be found that the radial size of the damage increases from 0 to 21 µm before stabilizes at 21 µm within the time range of 0 ~ 10 ps. Furthermore, the final radial size of the damage can be observed directly from the spectrum
Fig. 4. Spectral images of the probe beam recorded by CCD in the case of (a) without pump pulse and (b) with pump pulse. (c) The difference spectrum of (a) and (b) that reflects the beginning and the evolution of the laser induced damage. (d) A zoomed in view of the initial stage of damage evolution. The spatial domain has been converted to the actual size and the wavelength has been mapped into the time domain in (c) and (d).

obtained after the damaged structure had been stably formed, as shown in Fig. 5(b). The diameter of the damage area read from Fig. 5(b) is consistent with the result shown in Fig. 2, which depicts that the dynamic spectrum shown in Fig. 4(c) has covered the entire evolution of the damage.

Fig. 5. (a) The radial size of the damaged area extracted from the measured spectrum in the initial stage of evolution. The black balls represent the measured data and the red line represents the Boltzmann function fitting curve with $R^2 = 0.998$. (b) The spectrum obtained after the damage formation.

The evolution of the radial dimension lasted about 10 ps before reaching a steady state, but the change inside the damaged area still continues. Figure 6 shows the intensity evolution in the center row extracted from the dynamic spectrum in Fig. 4(c), which depicts the dynamics of the change inside the damage. The entire process in the measurement range can be divided into three stages. The first stage is a rapid transition status lasting about 4 ps. The value of $\Delta I/I_0$ experiences a rapid increase and decrease, as shown in the inset of Fig. 6. During this stage, the
energy of pump pulse is absorbed by the material in the focal area, which result in the formation (∼2 ps) and rapid recombination (∼2 ps) of plasma. The second stage is a slow decrease stage of ∆I/I₀, which lasted about 196 ps. The rapid recombination of plasma in the first stage leads to the decrease of plasma, which in turns reduces the recombination rate. We now discuss the possible reasons for the evolution of transmission intensity. The rapid increase may be due to the strong optical Kerr effect in the foci induced by the high pulse intensity. As for the decrease of ∆I/I₀, on the one hand, the transmission intensity will be weakened due to the generation of a large amount of plasma. The plasma inside the fused silica can be regarded as a plasma mirror or a diverging lens, thereby reducing the value of ∆I/I₀. On the other hand, from the perspective of material modification, the plasma recombination process is accompanied by the release and transfer of energy, which induces a rise in the temperature of the material around the foci. As a result, the material melted and resolidified, and eventually the void structure was formed, as shown in Fig. 2. Obviously, the formation of the damaged structure will reduce the transmission intensity of the probe light. The last stage is the steady one, which indicates that the damaged structure has been stably formed. The result of the temporal evolution of the transmission intensity revealed that the time required for the formation of void structures inside the damaged area is ∼200 ps.

![Graph](image)

**Fig. 6.** The intensity (∆I/I₀) evolution of the central area of the probe beam, which intuitively reflects the evolution process of the central part of the damage. The evolution can be divided into three stages, stage I: the rapid transition stage; stage II: the slow decrease stage; stage III: the stable stage. The inset shows the closer inspection of stage I.

### 4. Conclusion

In this paper, a double-void structural damage has been obtained by focusing a femtosecond laser pulse inside the fused silica. The characteristics of the damage morphology and the quantitative relationship between the size of the damaged area and the pulse energy have been studied in detail. The obtained quantitative relationship is expected useful to achieve the controllable manufacturing. The dynamics of the femtosecond laser-induced damage inside the fused silica has been measured by a single-pulse method. The evolution dynamics of the damage area, including the variation of radial size, void formation, and steady-state of the damage have been successfully characterized. A rapid increase (10 ps scale) of the radial dimension from 0 to 21 μm with the pulse energy of 1.1 mJ has been revealed from the measured spectrum, which conforms to the law described by the Boltzmann function. Meanwhile, the results showed that the formation of the void took about 200 ps. In conclusion, the proposed method alongside the in-house developed system has been demonstrated as a useful tool for the dynamic detection of laser-induced damage in laser micromachining.

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References


