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RESEARCH

Growth of laser damage in fused silica and CaF_2 under 263 nm laser irradiation

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Abstract

This study examined the growth of the laser-damage performance in optical components under the fourth harmonic of Nd:glass laser irradiation (263 nm). The damage-growth threshold of the optical component was relatively low under 263 nm laser irradiation compared to 351 nm irradiation, owing to the higher energy level of 4ω photons, and depended on the material characteristics. The preliminary growth of laser damage in fused silica and CaF_2 under 263 nm laser irradiation is reported in this article. The damage growth coefficients of these two materials were obtained by continuously irradiating the optical components using a 263 nm laser with a pulse width of $\tau = 5$ ns. The damage growth threshold of fused silica is lower than that of CaF_2 because of differences between the materials. The damage characteristics, including the damage morphology and bulk damage, were analyzed.

1 Introduction

The fourth-harmonic laser (263 nm) has great potential for high-power laser systems and has been widely studied [1, 2]; however, the relatively lower damage threshold of optics limits the applications of the 4ω laser [3]. Materials commonly used in high-power laser systems include fused silica, fluoride, and K9. K9 has a lower transmittance in the ultraviolet band, whereas fused silica and CaF_2 are increasingly used in deep-ultraviolet optical components because of their excellent optical and mechanical properties in the ultraviolet band [4–6]. Methods have been proposed for improving the laser damage threshold of optical components [7, 8]. For example, fused silica and CaF_2 have been employed to fabricate optical windows for use in laser systems. However, laser-induced damage and the growth of laser damage are major factors that limit the lifetime of optical components, resulting in transmission losses and additional damage to the optics due to beam modulation. The initial damage thresholds for the fused silica and CaF_2 have been determined using laser-induced damage tests [9]. The initial damage threshold of fused silica is 2~2.5 J/cm^2, and the initial damage threshold under 263 nm irradiation is far lower than that under 351 nm irradiation for the same material. The damage threshold of CaF_2 is higher than that of fused silica: ~5 J/cm^2. The initial damage threshold and damage growth are

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typically used to estimate the optical damage characteristics. The damage growth threshold is generally determined by extrapolating the curve of the damage probability with the energy density to the zero-damage probability [10]. This study focuses on the growth of the laser damage and the damage characteristics of fused silica and CaF_2 after 263 nm laser irradiation. A 263 nm laser was obtained using the non-critical phase-matched fourth harmonic generation of Nd:glass lasers in partially deuterated KDP crystals [11].

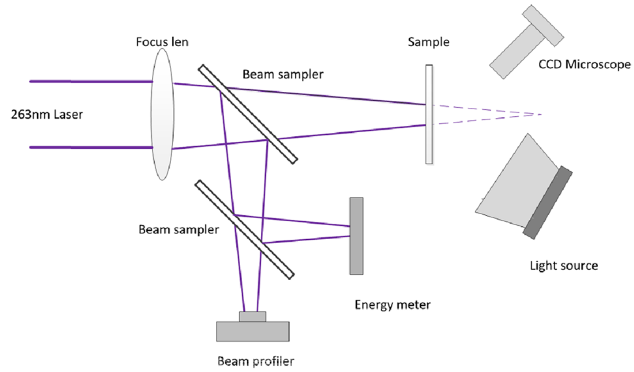


Fig. 2 Layout of the laser-induced damage test

2 Experiment

2.1 Experimental condition

A Nd:YLF laser (1,053 nm) with a pulse width of 10 ns was used as the pump laser because of its availability in our laboratory. To obtain the required 263 nm laser, a 10 mm long BBO crystal was used as a doubler crystal to obtain a 527 nm laser from the 1,053 nm Nd:YLF laser, and a 10 mm long 70% deuterated KD*P crystal was used to convert the second-harmonic laser (527 nm) into a 263 nm laser, then perform spectral filtering to remove residual first and second harmonic energy. The width of 263nm laser radiation is 5ns, and the time beam waveform is shown in Fig. 1, the diameter of the focused spot on the sample surface is 0.53mm. This laser was used to test laser damage growth. Figure 2 shows the experimental layout. A focusing lens was used to obtain sufficient laser fluence. Two beam samplers were used to monitor the energy and beam quality simultaneously. The sample was placed after the first beam sampler, and a charge-coupled device (CCD) microscope(Resolution of 4.5μm) was placed behind the sample to capture images of the damage to the sample in real time. Moreover, the microscope magnification was calibrated according to Groups 0–4 of the 1951USAF resolution test chart with 1.41 line pairs per millimeter. Thus, the pixel sizes of the damaged images were obtained. A light source was used to illuminate the sample to improve the monitoring with the microscope. Adjusting the fundamental frequency output through wave plates and polarizers to increase the energy output of the 263 nm laser from 0.02 to 40 mJ, and the samples were exposed to different laser intensities. Furthermore, the experimental device was placed in a clean laboratory.

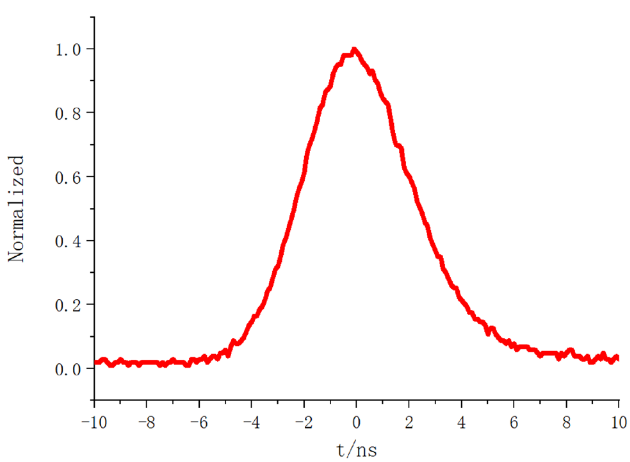


Fig. 1 Temporal waveform of the 263 nm laser

2.2 Samples

In our experiments, Sample 1 was fused silica (type Heraeus-suprasil 312), which was a super-polished, 9 mm thick, 50 mm^2 square. Sample 2 was CaF_2, which was round, 40 mm in diameter, and 11 mm thick,Table 1 shows the material characteristics of two materials [12](Before conducting damage testing, each sample was cleaned twice). These two samples, were prepared to test the damage behaviors, and the average damage growth area was calculated to illustrate the damage properties of the optical surface.

2.3 Experimental design

We aimed to determine the growth rate of laser damage on the exit surface under 263 nm laser irradiation. In this experiment, we utilized a fourth-harmonic laser to irradiate the samples, allowing them to grow until their diameter reached 3 mms. A minimum of 35 shots were administered under the working fluences to ensure no further growth of the damage sites. In some cases, up to

Table 1 Material characteristics of fused silica and CaF_2

Material	Fused silica	CaF_2
Two-photon absorption coefficient (cm/GW)	0.045	< 0.02
Band gap (eV)	9	12

100 shots were applied at each site until zero growth was observed, the initial recording point is when the surface of the sample is initially damaged. The frequency of the fourth-harmonic laser was set at 0.1 Hz. Following each laser irradiation, the lateral area of the damaged site was observed using a CCD microscope, and the damage area was calculated by processing the damage images obtained under the microscope.

The damage growth coefficient is used to evaluate the rate of area growth which is influenced by factors such as laser parameters, material properties, and initial damage site morphology. For an initial damage site under continuous laser irradiation with a fixed flux density, the size of the damage site generally increases with the number of shots. In general, the area of the damage sites on the incident surface of the component increased linearly, whereas the area of the damage points on the outgoing surface increased exponentially with the frequency of laser irradiation. Furthermore, the growth coefficient of damage on the rear surface was greater than that on the incident surface. The damage-growth threshold is the flux density corresponding to the zero damage growth coefficient. For the measurement of damage threshold, we statistically analyze the probability of damage at different energy levels, and then numerically fit the damage probability with energy. The energy density at which the fitted probability is 0 is used as the initial damage threshold. During the experiment, we used the 1-on-1 method for testing. Under the same laser flux, each point only received radiation once. After irradiating N points, we calculated the probability of damage and then changed the laser flux for the experiment until all points were undamaged at that flux.

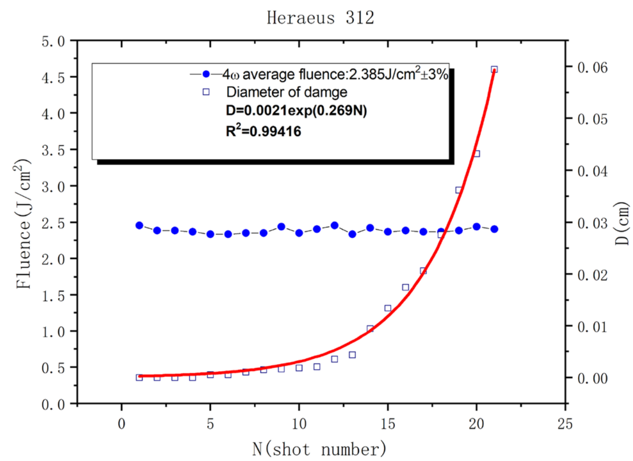


Fig. 4 Damage-growth behavior of fused silica, showing exponential growth with respect to the number of shots

3 Experimental results

3.1 Damage-growth thresholds of different materials

The growth of the laser damage for fused silica was tested first. A typical laser-damage growth site on the exit surface was observed while the fluence of the 263 nm laser irradiation was maintained at 2.385 J/cm^2, as shown in Fig. 3. Figure 3 illustrates the changes in the rear damage sites on the exit surface after different numbers of shots. Initially, the damage sites exhibited slow changes, but around the twelfth shot, the damage area began to increase rapidly. After 25 irradiations, the growth of the damage sites became negligible.

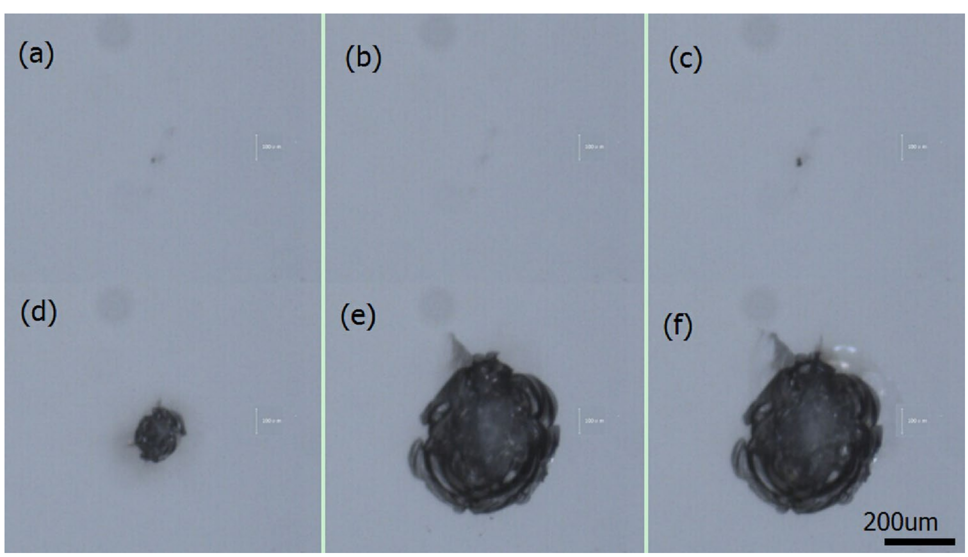


Fig. 3 Changes in the damage sites of fused silica with respect to the number of shots: **a** initial damage site, **b** damage site irradiated 6 times, **c** damage site irradiated 12 times, **d** damage site irradiated 18 times, **e** damage site irradiated 25 times, **f** damage site irradiated 30 times

The diameter of the damage site on the fused silica was calculated using the measured area by assuming a circular equivalent area. A growth plot of the effective diameter of the fused silica under a laser fluence of 2.385 J/cm^2 with respect to the number of shots is shown in Fig. 4. The average fluences in the area surrounding the growth site are also plotted. The data were fitted to an exponential curve [13] given by

$$D = D_0 e^{\alpha N} \tag{1}$$

where D denotes the effective diameter of the damage, N is the number of shots, and α is the growth coefficient. The exponential fit of the data is shown in Fig. 4, along with the R-squared value for this fit. All tested sites exhibited comparable fits to the data, and the growth coefficient is shown in Fig. 4.

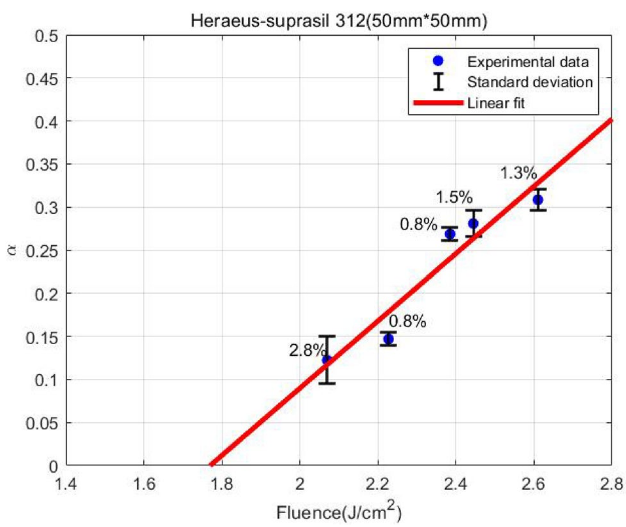


Fig. 5 Growth coefficients of fused silica for a laser fluence of 263 nm (The standard deviation of each set of data is marked in the figure)

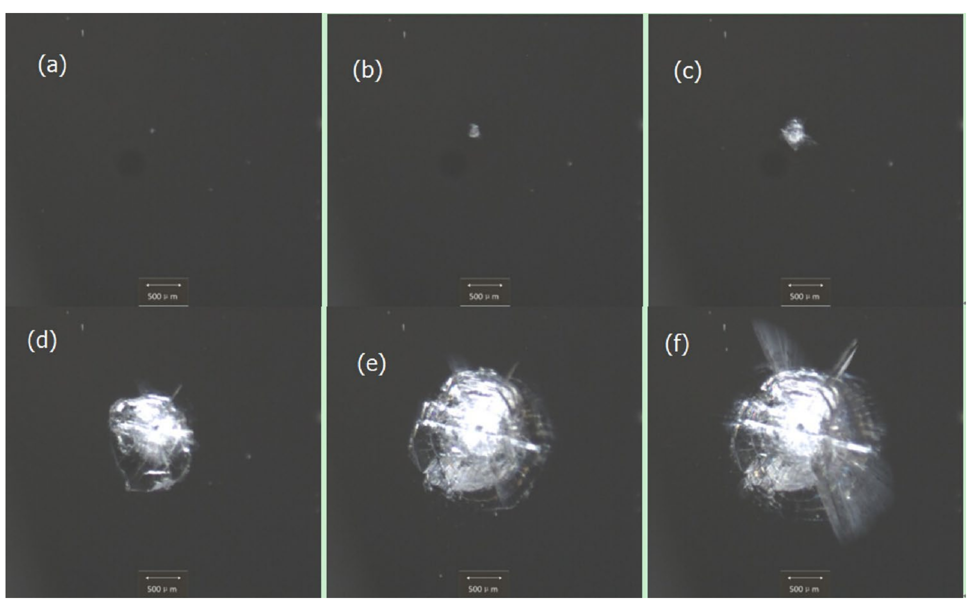
The growth of laser damage on the fused silica was tested under different laser fluences, and the growth coefficients at different laser fluences were obtained. Plots of the growth coefficient with respect to the fluence for many sites (We conducted 10–20 repeated experiments at each point, and the same for Fig. 8) shot with different fluences show the threshold behavior for growth as a function of the total fluence, as shown in Fig. 5.

As shown in Fig. 5, the linear fit to the data, $\alpha = 0.3903F - 0.69072$ with $R^2 = 0.88269$, predicted a growth threshold of 1.77 J/cm^2, although some sites did not grow beyond the growth threshold (not shown in the figure). Next, the growth of laser damage on CaF_2 was tested. A typical laser-damage growth site on the rear surface obtained while the fluence of the 263 nm laser irradiation was maintained at 5.39 J/cm^2 is shown in Fig. 6. The figure shows the changes in the rear damage sites after different numbers of shots.

A comparison between the damage morphologies on the exit surfaces of CaF_2 and fused silica revealed that the damage characteristics differed between the two materials (Fig. 7). The diameter of the damaged site in CaF_2 is calculated using the measured area. The change in the effective diameter of the damage site of CaF_2 under a laser fluence of 5.39 J/cm^2 with respect to the number of shots is shown in Fig. 8.

The growth of laser damage on CaF_2 was also tested under different laser fluences. The relationship between the growth coefficients and laser fluence was determined, as shown in Fig. 8, and the growth threshold of

Fig. 6 Changes in the damage sites of CaF_2 with respect to the number of shots: **a** damage site irradiated 3 times, **b** damage site irradiated 6 times, **c** damage site irradiated 8 times, **d** damage site irradiated 10 times, **e** damage site irradiated 15 times, **f** damage site irradiated 25 times



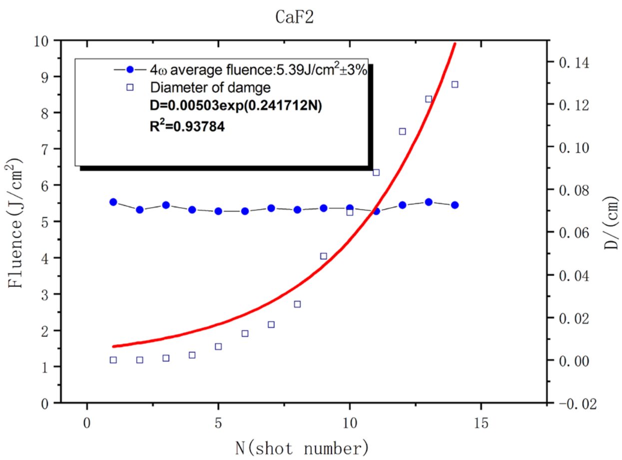


Fig. 7 Damage-growth behavior of CaF_2, showing exponential growth with respect to the number of shots

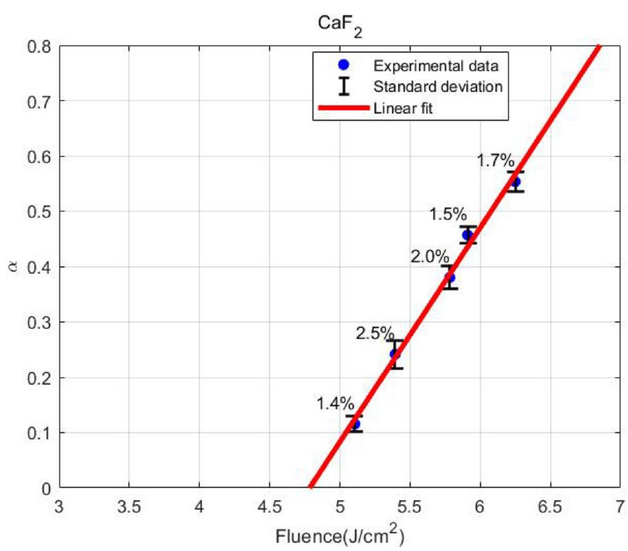


Fig. 8 Growth coefficients of CaF_2 for a laser fluence of 263 nm (the standard deviation of each set of data is marked in the figure)

CaF_2 is observed in this plot. Research has shown that an increase in the damage threshold of CaF_2 takes the form of an exponential function [14]. The linear fit for the data, $\alpha = 0.36113F - 1.71603$ with $R^2 = 0.93545$, shown in Fig. 8, predicts a growth threshold of 4.75 J/cm^2.

The growth thresholds of fused silica and CaF_2 were obtained using a laser-induced test. A comparison of these two materials is presented in Table 2. The initial thresholds and the growth coefficient α for the two samples are also listed in Table 2. We observe that the initial damage threshold of CaF_2 of ~5.1 J/cm^2 as well as the growth threshold of CaF_2 were higher than those of fused silica (~1.77 J/cm^2). The refractive index changed, and a color center formed when the fused silica was exposed to UV irradiation; therefore, the initial damage threshold of CaF_2 was higher than that of fused silica. In addition, from the growth coefficient α, it can be seen that the curve slope of CaF_2 is lower than that of fused silica, and when α reaches 0.4, the laser flux of CaF_2 material is about 6J/cm^2, and that of fused silica is about 3J/cm^2, that is, CaF_2 has better anti damage ability than fused silica under the irradiation of the same laser flux. The damage-growth threshold of fused silica under 263 nm irradiation was far lower than that under 351 nm irradiation (~5.1 J/cm^2) [9]. One reason for this is that the wavelength at 263 nm is shorter than that at 351 nm; thus, the photon energy is higher. Optical materials are more easily damaged by a 263 nm laser than a 355 nm laser [5]. Another reason is that a significantly greater nonlinear phenomenon occurred after the samples were exposed to a shorter-wavelength laser. For example, two-photon absorption forms transient defects and the self-focusing effect leads to excessive local energy [15, 16]. In addition, according to the two-photon absorption coefficient we provided earlier, the two-photon absorption coefficient of fused silica is also higher than CaF_2 at 263 nm, this means that the performance of calcium fluoride is better than that of fused silica under strong light, so calcium fluoride is more suitable for high-power laser systems.

3.2 Damage characteristics of different materials

Laser damage to materials can be divided into surface damage and bulk damage. Surface damage of materials mainly comes from surface defects and light absorption of impurities, while body damage is mainly caused by body defects and intrinsic damage of materials. The differences in the damage characteristics of the exit surfaces of the two samples were examined using a CCD microscope. The damage was observed using a high-precision optical microscope for accurate analysis. Figure 9 shows the damage to the exit surfaces of the two optical materials.

Table 2 Damage thresholds and extrapolated damage-growth thresholds of fused silica and CaF2

Laser irradiation	263 nm at 5ns	263 nm at 5ns
Material	Fused silica	CaF_2
Damage threshold (J/cm^2)	~2.5	~5.1
Damage-growth threshold (J/cm^2)	~1.8	~4.8
The growth coefficient	0.3903F - 0.69072	0.36113 F - 1.71603

Figure 9a shows the damage sites on the fused silica under a 263 nm laser fluence of 2.33 J/cm^2. Bulk damage did not occur in this sample and visible features of the damage were observed on the exit surface of the fused silica. The middle of the damage site was molten, and the fused silica exhibited evident signs of ablation and impact compression. There are many melting nodules in the central melting area, and the material in the damaged area is in a high-temperature melting and gasification state during the damage process. Figure 9b shows the damage sites on CaF_2 under a 263 nm laser fluence of 5.39 J/cm^2. Radial and circumferential cracks were more obvious in CaF_2 than in fused silica, and cleavage was the main form of damage in CaF_2 because of the thermodynamic properties of this material. The damage of CaF_2 material is more complex than that of fused silica. There are many regular geometric shapes in its damage pit, which is due to the fact that CaF_2 material is a soft and brittle crystal material, and brittle fracture is manifested as cleavage fracture, cracks propagate along the cleavage plane. As a crystal material, CaF_2 is prone to residual defects such as scratches and impurities on the surface during the grinding and polishing process, so the processing of CaF_2 materials has a great impact on its damage. Compared with fused silica, it can be found that under the same conditions, under the current processing technology, the surface damage of CaF_2 is better than that of fused silica.

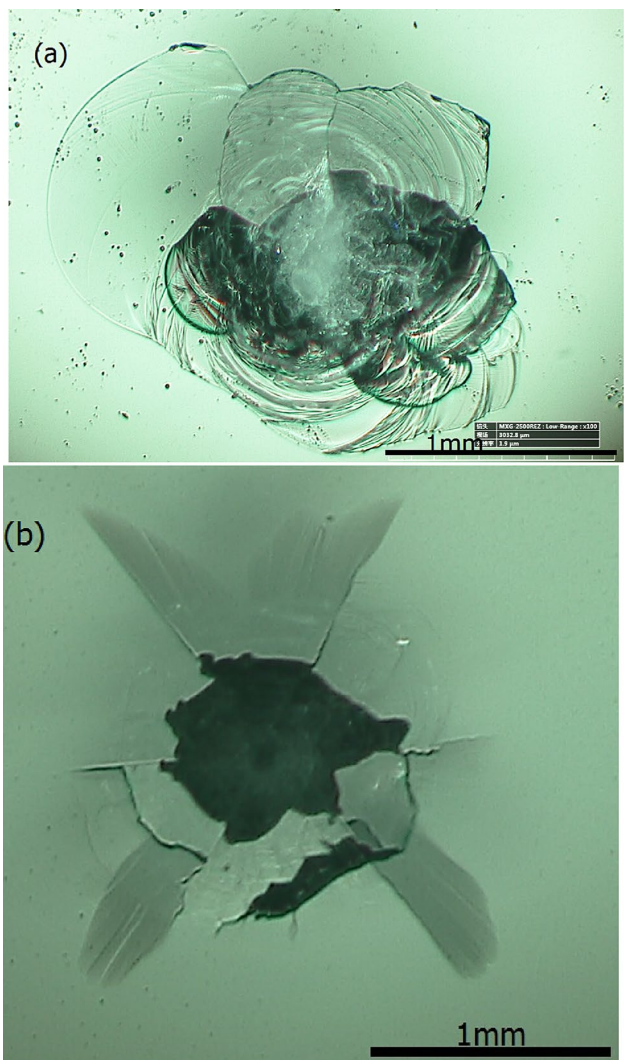


Fig. 9 Damage to the exit surfaces of the two samples: **a** fused silica (the lower right corner shows the camera parameters) **b** CaF_2

Under the same flux irradiation, fused silica has bulk damage but CaF_2 does not. Since the side polishing of fused silica is convenient for us to observe the bulk damage, we have carried out bulk damage experiments on fused silica. Figures 10, 11, 12 and 13 show the damage characteristics observed on the side of the sample under different laser fluences. Four damage sites, irradiated by different fluences, were observed, all of which were irradiated 30 times, except for the damage site irradiated by the fluence of 2.96 J/cm^2, which was irradiated four times.

The sample was irradiated at a fluence of 2.33 J/cm^2, which was slightly higher than the damage threshold. Damage occurred on the exit surface of the fused silica, whereas the front surface and bulk were not damaged. When the sample was irradiated with a higher fluence of 2.38 J/cm^2, bulk damage appeared near the exit surface and filamentary damage was observed (Fig. 10). When the sample was irradiated at a fluence of 2.44 J/cm^2,

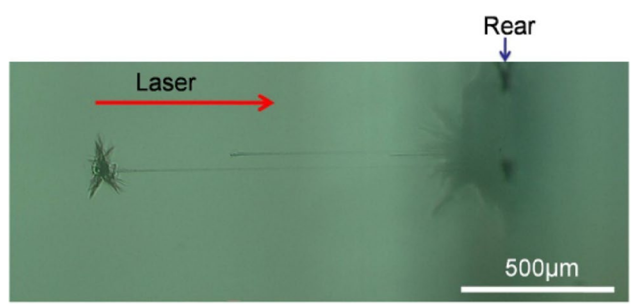


Fig. 10 Damage characteristics of fused silica under a laser fluence of 2.38 J/cm^2

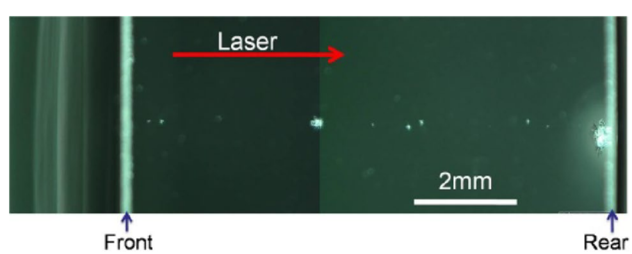


Fig. 11 Damage characteristics of fused silica under a laser fluence of 2.44 J/cm^2

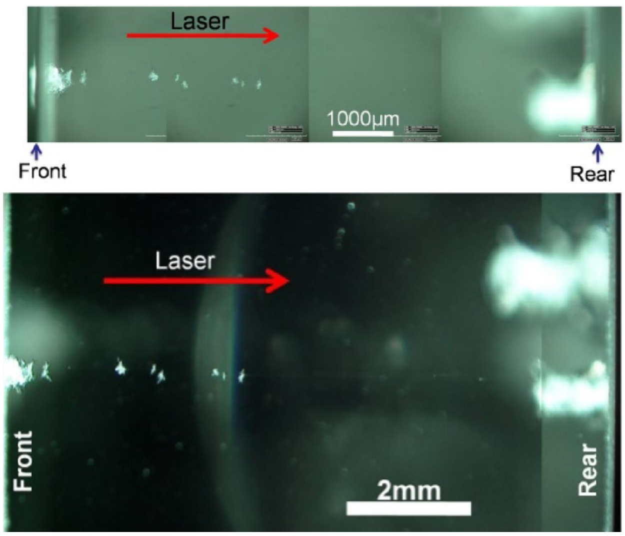


Fig. 12 Damage characteristics of fused silica under a laser fluence of 2.75 J/cm^2

Fig. 13 Damage characteristics of fused silica under a laser fluence of 2.96 J/cm^2

filamentary damage appeared in the front of the fused silica and nearly throughout the sample (Fig. 11). When the laser fluence was higher than 2.75 J/cm^2, both the front and exit surfaces were damaged, filamentary damage occurred throughout the sample, and bulk damage was evident (Fig. 12). When the sample was irradiated with a laser fluence of 2.96 J/cm^2 four times, both the front and exit surfaces were damaged, and filamentary damage occurred (Fig. 13). Due to the optimization of the current process, the bulk defect density of the material is very low, so the intrinsic damage threshold of the material can be approximately regarded as the bulk damage threshold. Because the refractive index of fused silica changes under the irradiation of ultraviolet laser, resulting in the self focusing effect, and damage occurs inside the material after the focused laser exceeds the intrinsic damage threshold of the material. Compared with fused silica, CaF_2 has a wider band gap, which makes multiphoton ionization speed lower. At the same time, as a crystal material, CaF_2 has a periodic arrangement of its internal structure. Compared with fused silica, an amorphous material, CaF_2 has a lower density of structural defects in its body. The initial electrons of avalanche ionization are provided by defects and free electrons ionized by multiphoton ionization. Therefore, CaF_2 has fewer initial electrons and a lower electron avalanche rate. Therefore, the intrinsic damage threshold of CaF_2 is higher than fused silica, so fused silica is more prone to bulk damage under the same laser flux.

4 Conclusion

Samples of different materials were exposed to a 263 nm laser, and the growth of laser damage in these materials was tested. In our experiment, the growth coefficients for different laser fluences were determined to predict the damage-growth thresholds of the two materials. The estimated damage growth thresholds of CaF_2 and fused silica were 4.7 and 1.7 J/cm^2, respectively. The damage surfaces of the two materials were also analyzed. The variations in the damage characteristics of the samples resulted from the differences in the material properties. Bulk damage filamentation inside the fused silica with associated exit-surface damage was caused by self-focusing. The damage-growth threshold of the fused silica under the 263 nm laser was lower than that of CaF_2 because the refractive index changed and a color center was formed when the fused silica was exposed to UV irradiation. The initial and damage-growth thresholds of CaF_2 were higher than those of fused silica under the 263 nm laser; therefore, the performance of CaF_2 was better than that of fused silica. In conclusion, the material for the UV laser output window, as well as for any subsequent windows (such as the target chamber window), should be carefully chosen, With its excellent characteristics, CaF_2 can effectively improve the output level and service life of current high-power laser devices.

Author Contributions Qi Zhang and Xiuqing Jiang performed the conceptualization, data analysis, methodology and investigation; Qi Zhang performed the writing - original draft and writing - Review & Editing; All authors reviewed the manuscript.

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Data availability No datasets were generated or analysed during the current study.

Declarations

Conflict of interest The authors declare no Conflict of interest.

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