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Effect of Femtosecond Laser Polarization on the Damage Threshold of Ta_2O_5/SiO_2 Film

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Abstract: The study used linearly and circularly polarized femtosecond pulsed lasers to irradiate a Ta_2O_5/SiO_2 film. Firstly, the damage thresholds of the film for linearly and circularly polarized femtosecond pulsed lasers were measured in 1-on-1 mode. The results showed that the damage threshold (1.70 J/cm^2) under a circularly polarized laser was higher than that (1.68 J/cm^2) under a linearly polarized laser. For femtosecond lasers, the multi-photon ionization cross-section under circular polarization was lower than that under linear polarization. The lower ionization rate under circular polarization led to a higher damage threshold compared to the case under linear polarization. Secondly, the damage morphology of the film irradiated by linearly and circularly polarized femtosecond lasers was observed by microscope. The damage caused by linearly polarized laser was more evident than that caused by the circularly polarized laser. Finally, the damage thresholds induced by linearly and circularly polarized femtosecond pulsed lasers were measured in S-on-1 (S = 2, 5, and 10) mode. For the same S value (2, 5, or 10), the damage threshold under the circularly polarized laser was higher than that under the linearly polarized laser. The damage thresholds under two polarized laser pulses decreased with an increase in the number of laser shots, indicating that repeated laser pulses had a cumulative effect on the damage of the film.

Keywords: femtosecond laser; optical thin film; damage threshold; damage morphology; multi-photon ionization cross-section; cumulative effect

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1. Introduction

Optical thin film is a critical component in the laser system, and its optical performance determines the quality of laser output to a large extent. However, the laser-induced damage threshold (LIDT) of thin films is one of the crucial bottlenecks that limit the development of laser technology to high power and high energy. It is also one of the critical factors affecting laser systems' stability and service life. As the output power of laser systems increases, the requirements for the ability of optical films to resist laser damage are getting higher and higher. Before a new type of optical film is used, evaluating its ability to resist laser damage is a necessary procedure. Among them, laser parameters include laser wavelength, pulse width, energy density, spot area, polarization state, etc.; film properties include film material, thickness, processing technology, preparation methods, etc. From the above discussion, it can be seen that the laser damage of optical films is a very complicated and accidental problem.

Many researchers have studied the laser-induced damage threshold (LIDT) of optical thin films. Kumar et al. investigated the LIDT of Ta_2O_5 and Ta_2O_5/SiO_2 films at

532 and 1064 nm [1]; their results showed that the values of LIDTs of all samples, single layer, all quarter-wave multilayer and multilayer having some upper layers non-quarter wave thickness for 1064 nm wavelength were higher than LIDTs at 532 nm wavelength. Gallais et al. studied the damage characteristic of the GaAs PHEMT [2]. The results show that the pulse width, pulse period, and duty ratio of the injected signal have a significant influence on the damaging effect of the device. Zhu et al. investigated the electric field distribution, the LIDT, and the damage morphology of the prepared four mirror coatings [3]. Their results showed that the SiO_2 coating and electric field distribution greatly influenced the LIDT and damage morphology of the mirror coating. Wang et al. studied the effect of optical parameters (such as extinction coefficient, thickness, and refractive index) and laser parameters (such as laser irradiation number and pulse width) on the LIDT of the SiO_2 thin film [4], finding that optical parameters had little effect on the LIDT, laser parameters had a great effect on the LIDT. Bonse et al. investigated the effect of pulse duration on the LIDT in thin triazenepolymer films on glass substrates [5]. They found a significant dependence of the LIDT on the pulse duration in the sub-picosecond regime. Mueller et al. used 1-on-1 and 10-on-1 modes to measure the LIDT of Mo/Si multilayers with an extreme ultraviolet pulse at a wavelength of 13.5 nm [6]. According to their results, the differences between these two measurement methods were compared and analyzed, finding that the damage threshold at the 10-on-1 mode was 60% lower than that at the 1-on-1 mode. Chen et al. studied the melting and thermal-stress damage thresholds of silicon induced by long pulsed laser at 0.532, 1.064, and 10.6 μm [7] and analyzed the damage mechanism of silicon. According to their results, the LIDT of silicon with 0.532 μm was lower than that with 1.064 and 10.6 μm, and the damage mechanism of silicon was mainly the melting damage.

It can be seen from the above discussion that LIDT is not only limited by laser parameters, including laser wavelength, pulse width, energy density, spot area, polarization state, etc. It is also limited by the characteristics of the film, including the film material, thickness, processing technology, preparation method, etc. [5,8–13]. At present, there are few systematic studies on the damage mechanism of thin films under different polarization femtosecond lasers. This experiment studied the damage of thin films irradiated by laser with different polarization based on LIDT and microscopic morphology. The damage mechanism and damage effect of polarized laser on thin-film are understood by conducting the experiments of thin-film laser damage to provide a basis for improving the damage threshold and laser protection of optical thin film.

In addition, among many thin films, the Ta_2O_5 thin film has excellent potential application due to its advantages and is one of the research hotspots in recent years. Firstly, it has a high dielectric constant (30–35), chemical stability, and thermal stability. Secondly, it is considered to be the most advanced technology in the field of microelectronics due to its easy compatibility with semiconductor integrated circuits/processes. Thirdly, it has been widely used in the optical field due to its high refractive index, low absorption rate, and wide spectral transmittance range (300 nm–10 μm). Finally, its high hardness can be used as a protective film [14–17]. So, the laser damage of a Ta_2O_5/SiO_2 multilayer film prepared by electron beam evaporation was studied by 1-on-1 and S-on-1 measurements under linearly polarized and circular-polarized femtosecond lasers. The damage threshold of the film for both polarized lasers was obtained, and a microscope observed the damage morphology of the film for both polarized lasers.

2. Experiment

2.1. Sample Preparation

In the experiment, a filter based on fused quartz is customized by electron beam evaporation, and the corresponding sample image is shown in Figure 1a. Due to the special spectral requirements of the filter, the film layer adopts an irregular structure layer. The primary layer structure is the alternating stacking of high and low refractive index materials (Ta_2O_5 and SiO_2) with different optical thicknesses. The transmission spectrum curve of the film is shown in Figure 1b, with a spectral range of 400 nm–900 nm. As the membrane layer is an irregular membrane system with different thicknesses in the center, it presents different electric field distributions from the conventional filter. Figure 1c shows the electric field distribution of the film at the wavelength of 800 nm.

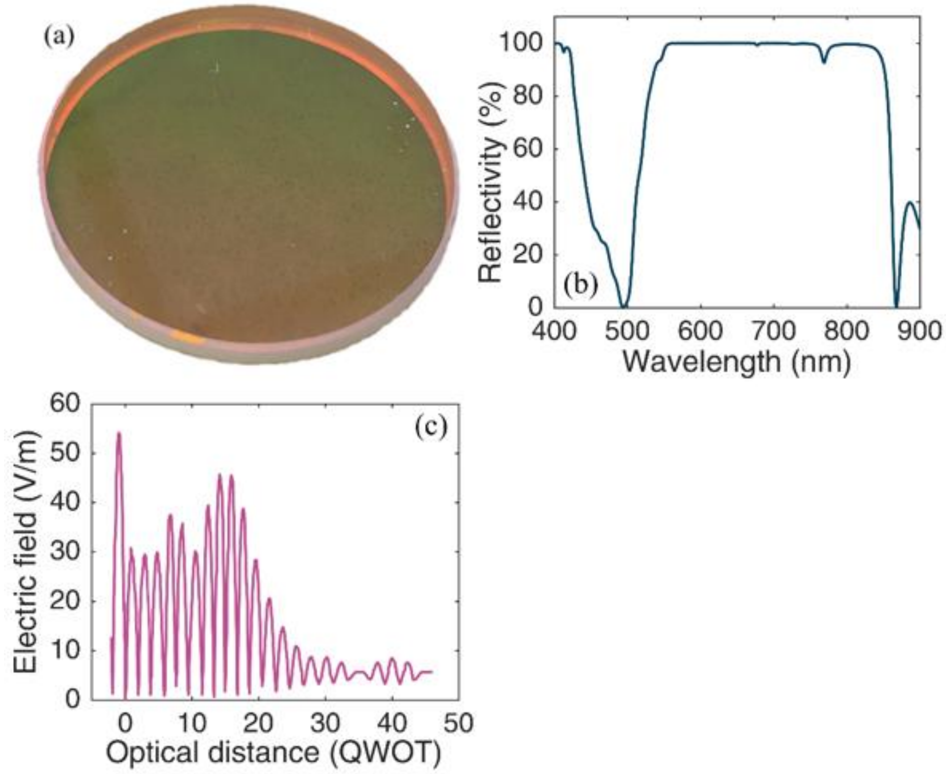


Figure 1. (**a**) Ta_2O_5/SiO_2 multilayer film sample; (**b**) reflectivity curve of the film; (**c**) electric field distribution of the film at 800 nm.

2.2. Experimental Setup

The schematic diagram of femtosecond laser damage to optical thin film is displayed in Figure 2. The whole experimental setup consists of a pulse laser, a half-wave plate, a Gran laser polarizer, a focusing mirror, an energy meter, a CCD camera, a microscope, an experimental sample, and a three-dimensional translation stage. The excitation source was an ultrafast Ti:sapphire femtosecond laser system (Coherent (Santa Clara, CA, USA), Libra-HE) with a center wavelength of 800 nm, a pulse width of 50 fs, and a repetition frequency of 1 kHz. The laser system was modified to the single-shot mode by sending a command ('man:trig') to the serial port (RS232) of the synchronization and delay generator. The laser energy was measured by an energy meter (Coherent, LabMax-TOP laser energy meter + EnergyMax-RS J-10MB-LE Energy Sensor). The energy attenuation system was composed of a computer-controlled rotating half-wave plate (HWP) and a Glan-laser polarizer (Glan); the fluctuation of pulse energy was less than 0.5% within 8 h. A quarter-wave plate (Q) was used to adjust the polarization of the femtosecond laser. For circularly polarized pulsed beam, the angle of the quarter-wave plate was $45° + n \times 90°$; for linearly polarized pulsed beam, the angle of the quarter-wave plate was $n \times 90°$. The n represented an integral multiple of 90°. Next, a plano-convex lens (L, focal length of 25 cm, diameter of 25 mm) was used to focus the pulse beam vertically onto the sample surface. The diameter of the focused spot at the sample surface was approximately 30 μm. In addition, the sample to be measured was placed on a computer-controlled three-dimensional translation stage (Thorlabs, PT3/M-Z8, with an accuracy of ±1.5 μm), and the minimum movement step of the translation stage was 1.0 μm. During the experiment, the translation stage was used to complete the positioning of the sample and control the movement of the sample test point. The damage morphology was monitored by a microscope and a CCD camera for real-time observation, and a more precise microscope (Olympus, BX51) was used to observe the damage morphology of the film after the experiment. All measurements were performed in an atmospheric environment, room temperature (22 °C), and 40% relative humidity.

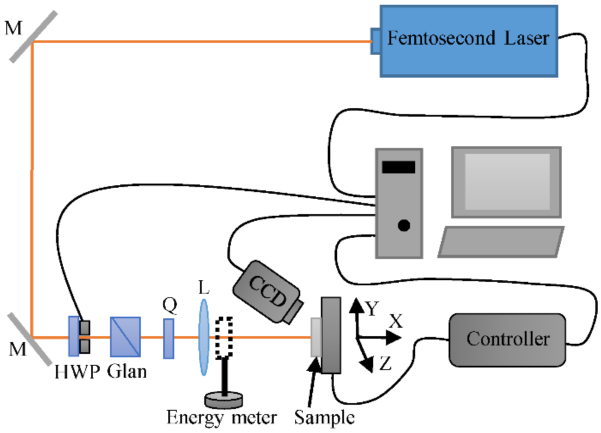


Figure 2. Experimental setup of femtosecond laser damage to optical thin films (M is the 800 nm mirror; HWP is the half-wave plate; Glan is the Glan-laser polarizer; Q is the quarter-wave plate; L is focusing lens).

2.3. Experimental Process

According to the damage standard of optical film from the international standard ISO/1125454 [18], the damage threshold of Ta_2O_5/SiO_2 multilayer film was measured by using the method of "1-on-1" and "S-on-1" for linearly and circularly polarized femtosecond lasers. The 1-on-1 measurement method, also known as single-pulse damage measurement, referred to multiple test points irradiated by laser with the same energy to the sample surface. When the 1-on-1 measurement method is used, each test point was irradiated only once and then moved to the next un-irradiated test point regardless of whether damage occurred. The S-on-1 measurement method, also known as multi-pulse damage measurement, refers to the cumulative effect of repeated laser irradiation on the sample surface. The multiple pulses with the same energy are measured on the same test point. During the experiment, the samples were irradiated by pulsed lasers with different laser polarization and fluences, and each laser fluence irradiated 30 points with a spacing of 200 µm. Additional values (2, 5, and 10) were set for the number of laser irradiation (S) to find the measurement points with non-0% and 100% damage probability. Then the radiation measurement was carried out in the order of increasing energy, using the zero-probability damage threshold to determine the measurement results [19]. The damage probability is the ratio of the number of damage points on the film surface to the number of laser irradiation points.

3. Results and Discussion

When the laser pulse interacts with the dielectric material, the atoms or molecules in the dielectric material absorb one or several photons and ionize, which is called photoionization. For ultrafast laser pulses, the effect of avalanche ionization and electron recombination and attachment process on electron density is small and can be ignored. After absorbing multiple photons, bound electrons in atoms or molecules will transition from a bound state to a continuous state, called multi-photon ionization [20]. When the laser field is strong enough and its frequency is low, it can be regarded as a quasi-electrostatic field. This quasi-electrostatic field is superimposed on the Coulomb potential of the matching electron to form a potential barrier. The bound electron has a certain probability of passing through this potential barrier and becoming a free electron. This process is called tunneling ionization [21,22]. According to different ionization mechanisms, photoionization can be divided into multi-photon ionization and tunnel ionization [23,24]. Tunneling ionization of atoms or molecules is the same effect as multi-photon ionization in a sense. Compared with multi-photon ionization, tunneling ionization occurs at higher light intensity. Therefore, the polarization dependence of femtosecond laser largely depends on nonlinear ionization processes, such as multi-photon ionization, tunneling ionization, and avalanche ionization [25].

Firstly, the LIDT of the Ta_2O_5/SiO_2 multilayer film under linearly polarized and circularly polarized femtosecond lasers was measured at 1-on-1. Figure 3 shows the evolution of damage probability with femtosecond laser fluence under linearly polarized and circularly polarized femtosecond laser radiation at 1-on-1. In the experiment, the microscopic method was used to identify the film damage. The working mode was to place the tested sample under a microscope and observe whether there were damaged spots on the film surface. If there were damage spots on the film, the film was considered to be damaged. According to the international standard ISO/1125454 in the optical film damage standard, the film was placed in the laser optical path, and the laser irradiated the film. Next, the irradiated film was placed under a microscope with a magnification of 200 times, observed whether the film had apparent damage, and recorded the number of damage spots at the same time. By dividing the number of damage points by the number of laser shots, the damage probability was obtained. According to Figure 3, the damage threshold of linearly polarized and circularly polarized femtosecond pulsed lasers is 1.68 and 1.70 J/cm^2, respectively. As the laser energy increases, the damage probability of the linearly polarized femtosecond laser is significantly higher than that of the circularly polarized femtosecond laser, which is attributed to the fact that the multi-photon ionization cross-section of the linearly polarized femtosecond laser is considerably higher than that of circularly polarized femtosecond laser [25]. Temnov et al. [25] suggested that six-photon ionization was considered to be the primary ionization mechanism in fused quartz and sapphire, and the dependence of ionization rate on laser polarization was obviously different; that is, the ionization rate of linear polarization was dominant in high-order multi-photon ionization.

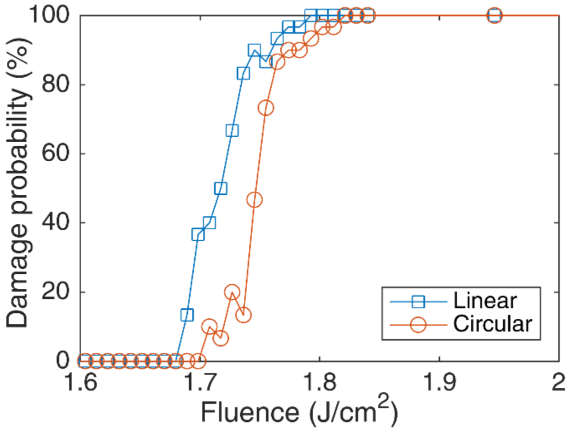


Figure 3. Damage probability with laser fluence at 1-on-1.

It can be seen that the difference of photon ionization process under different polarization was the main reason leading to the difference of damage process [26]. Therefore, the multi-photon ionization cross-section under linearly polarized femtosecond laser is stronger than that under circularly polarized femtosecond laser, leading to the stronger ionization rate under linearly polarized femtosecond laser. As a result, the free electron density under linearly polarized femtosecond laser is higher than that under circularly polarized femtosecond laser. The damage threshold under linear polarization is lower than that under circular polarization. To observe the phenomenon more intuitively, we use a microscope to observe the damaged morphology of the film under linearly polarized and circularly polarized femtosecond lasers, as shown in Figure 4. It is found that the damage under the linearly polarized laser is more evident than that under the circularly polarized laser, which better presents the conclusion in Figure 3; that is, the damage threshold under circular polarization is higher than that under linear polarization.

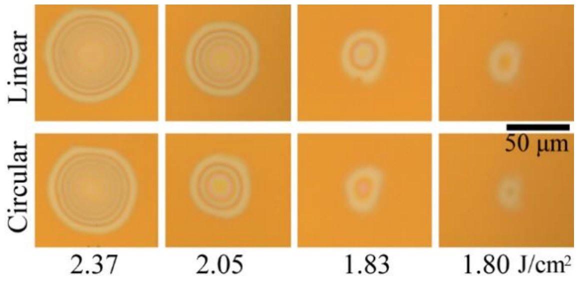


Figure 4. Typical damage morphology under linearly and circularly polarized lasers at 1-on-1.

Secondly, the damage threshold of the Ta_2O_5/SiO_2 multilayer film under linearly polarized and circularly polarized femtosecond lasers was measured at S-on-1 (S = 2, 5, and 10). Figure 5 shows the damage probability of linearly polarized and circularly polarized femtosecond lasers with the femtosecond laser fluence at S-on-1 (S = 2, 5, and 10). According to Figure 5, the damage thresholds (S = 2, 5, and 10) under linearly polarized 1femtosecond laser are 1.56 1.44, and 1.42 J/cm^2, respectively; the damage thresholds (S = 2, 5, and 10) under circularly polarized femtosecond lasers are 1.61, 1.50, and 1.47 J/cm^2, respectively. It is found that the damage thresholds of linearly polarized and circularly polarized femtosecond laser decreased with an increase in the number of laser irradiations. Figure 6 shows the damage morphology of the film under linearly polarized and circularly polarized femtosecond at 5-on-1. It is found that the damage of the film under linearly polarized and circularly polarized femtosecond for 5-on-1, which better verifies the conclusion in Figure 5. Wang et al. conducted laser damage testing in multilayer mirrors under 1-on-1 and S-on-1 modes [27], and their results showed that the damage threshold of 1-on-1 mode was much higher than the S-on-1 mode. At the S-on-1 mode, the main factor influencing the damage threshold is the accumulation of irreversible laser-induced defects and native defects, as shown in the case of circular polarization 5-on-1 from Figure 6. Natoli et al. used single-shot mode and multi-shot mode of laser damage characteristics of silica at 1064 and 355 nm [28], their results showed that the damage threshold of silica decreased with an increase in shot number, and the damage threshold decreased by 35% (1064 nm) and by 20% (355 nm) after 100 shots. The result revealed a systematic fatigue effect of material in shot number. In terms of damage threshold and damage morphology, repeated laser pulses have a cumulative effect on the damage of the Ta_2O_5/SiO_2 multilayer film, and the cumulative effect becomes more obvious with more radiation times. Therefore, as the number of radiation increases from 2 to 10, the damage threshold of the film decreases.

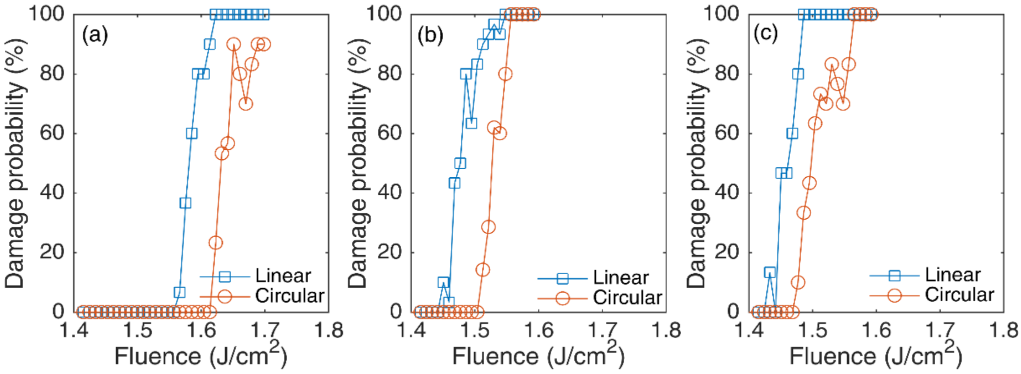
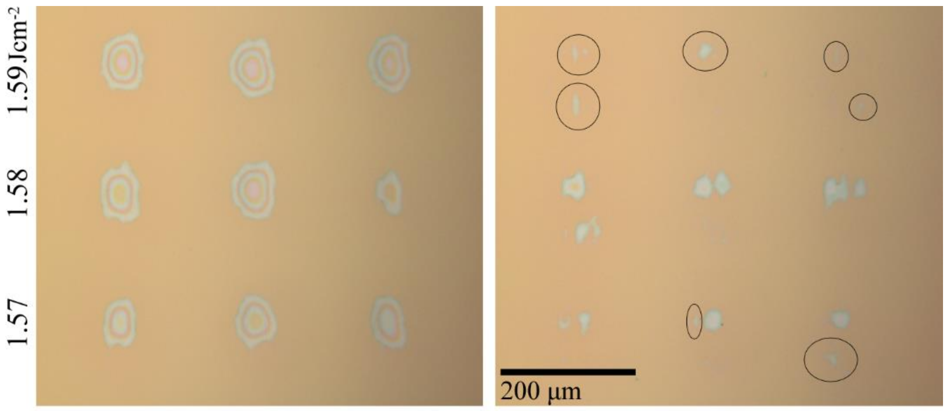


Figure 5. Damage probability of linearly and circularly polarized lasers under 2-on-1 (**a**), 5-on-1 (**b**), and 10-on-1 (**c**).



Linear Circular

Figure 6. Damage morphology near damage threshold for linearly and circularly polarized lasers under 5-on-1. Note: black circle indicates some unapparent damage.

4. Conclusions

The laser damage of the Ta_2O_5/SiO_2 multilayer film on the fused silica substrate prepared by electron beam evaporation was studied by 1-on-1 and S-on-1 measurements using linearly and circularly polarized femtosecond lasers. The damage threshold of the film under linear and circular polarization was obtained, and the damage morphology of the film under linear and circular polarization was observed by microscope. It was found that the damage threshold of linear polarization was lower than that of circular polarization, and the damage morphology of under polarization was more evident than that under linear polarization. The main reason was that the multi-photon ionization cross-section of linear polarization was higher than that of circular polarization, resulting in a higher ionization rate of linear polarization. It was also found that the damage threshold of linearly polarized and circularly polarized femtosecond laser decreased with an increase in laser radiation number. The main reason was that repeated laser radiation had a cumulative effect on the film; the more repeated radiation, the more pronounced the cumulative effect. The result is of great significance for understanding the mechanism of femtosecond laser-induced optical film damage and the femtosecond laser projection of the film.

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