Laser-induced damage threshold in n-on-1 regime of Ta_2O_5 films at 532, 800, and 1064 nm

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Ta_2O_5 films were prepared with conventional electron beam evaporation and annealed in O_2 at 673 K for 12 h. Laser-induced damage thresholds (LIDTs) of the films were performed at 532 and 1064 nm in 1-on-1 regime firstly, and then were performed at 532, 800, and 1064 nm in n-on-1 regime, respectively. The results showed that the LIDTs in n-on-1 regime were higher than that in 1-on-1 regime at 532 and 1064 nm. In addition, in n-on-1 regime, the LIDT increased with the increase of wavelength. Furthermore, both the optical property and LIDT of Ta_2O_5 films were influenced by annealing in O_2.

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Ta_2O_5 is always used as one of the most important high index materials for optical coatings. Moreover, Ta_2O_5 films have other excellent properties of thermal and chemical stability, so they could be widely used in many fields. However, a disadvantage lies that Ta_2O_5 films are liable to non-oxidant and have high optical loss, thereby annealing is performed as an important method to improve the optical properties[1,2]. Laser-induced damage threshold (LIDT) is the heat point in the researches of laser films. The LIDT in n-on-1 regime is different from that in 1-on-1 regime. In addition, film and bulk damages have both been observed to depend on wavelength, and the LIDTs have been found to be different at different wavelengths[3−5]. In this paper, we studied the optical property, structure, absorption, microdefect density, and LIDT of Ta_2O_5 films before and after annealing. The LIDT of the annealing films was studied at 532 and 1064 nm in 1-on-1 regime, and also at 532, 800, and 1064 nm in n-on-1 regime, respectively.

The films were deposited by using Ta_2O_5 crushed aggregates as starting material with the purity of 99.99%. Ta_2O_5 films were deposited on BK7 glass substrates under optical control and all the film thicknesses were kept at about 400 nm. Before film deposition, every substrate was cleaned ultrasonically in petroleum ether, the temperature was held at 353 K, the chamber was pumped to a base pressure of 2×10^{-3} Pa, and oxygen was introduced to keep oxygen partial pressure of 2×10^{-2} Pa. The substrate temperature was 573 K and the deposition rate was 0.32 nm/s. The post-deposition annealing of the films was performed in O_2 at 673 K for 12 h.

Microstructure of Ta_2O_5 films was analyzed by an X-ray diffractometer. Transmittance spectra of the samples were measured using a Lambda 900 spectrophotometer and the measurement error is within 0.08%. The refractive index and extinction index of the films were calculated by Essential Macleod (a thin film design software). The surface thermal lensing (STL) technique permits the measurement of the very low levels of absorption and the sensitivity was 1 ppm. Experimental apparatus was shown in Ref. [6]. Five sites were selected randomly on each sample surface to measure the microdefect density under a Leica-DMRXE dark-field microscope. Damage testing at 532 and 1064 nm in 1-on-1 and n-on-1 regimes was performed by a Q-switch pulsed laser. The experimental setup was shown in Refs. [6] and [7]. Wavelength at 532 nm was generated by potassium titanyl phosphate (KTP). The laser spot size was measured by using scanning knife edge technique and the fluctuation of laser spot size was about 6%. The LIDT in 1-on-1 regime was defined as the incident pulse's energy density when the damage occurred at 0% damage possibility. The 1-on-1 test used a single shot per site. The n-on-1 test used a series of single shots, the fluence of each shot was increased step-wise until the upper bound was reached. The step of the fluence was about 0.2 J. Considering the test error and statistical error, the total error was about 12% in this LIDT measurement. The LIDT at 800 nm in n-on-1 regime was measured using the chirped pulse train from 23-TW Ti:sapphire laser system. The detailed test process was shown in Ref. [8]. The effective spot diameter was around 3 mm and the fluctuation of spot size was 10%. The relative error of the LIDT determination at 800 nm amounts to 20%, which is mainly due to the uncertainty in the spot size measurements.

The transmittances of the samples were measured before and after annealing. It is found from Fig. 1 that the spectra shift to short wavelength after annealing. At the mean time, the optical transmittance increases especially in the short wavelength region, which may be attributed to the decrease of the optical loss by annealing. Figure 2 shows the refractive index and the extinction index of the as-deposited and annealed samples. It can be clearly seen that the refractive indices increase and the extinction indices decrease after annealing.

Figure 3 shows the X-ray diffraction (XRD) pattern of the samples before and after annealing. The crystallinity of the annealed sample is unchanged from the XRD

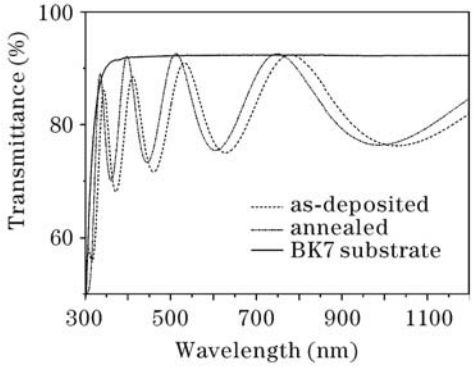

Fig. 1. Transmittance spectra of the samples.

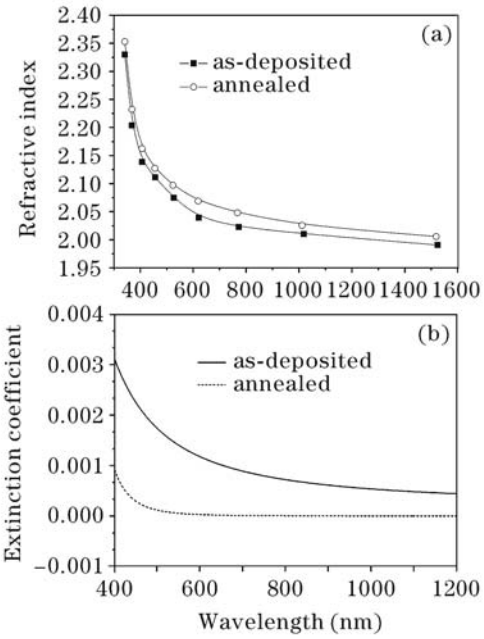

Fig. 2. (a) Refractive index and (b) extinction index of the samples.

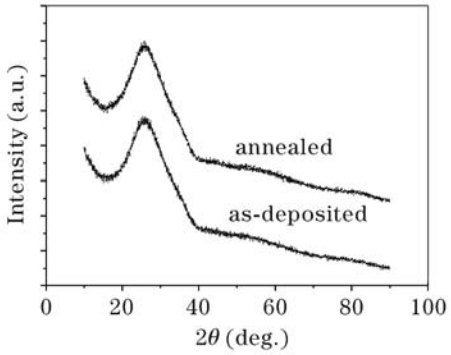

Fig. 3. XRD analysis of the as-deposited and annealed samples.

analysis and both the films are amorphous. It is consistent with some literatures that the change from amorphous to crystalline phase of Ta_2O_5 films should undergo a high temperature annealing (> 973 K)[9,10]. The Ta_2O_5 crushed aggregate as starting material in our study is tetragonal phase and the crystalline point is about 1223 K.

The STL technique is a sensitive method for detecting the thermal absorption of the films. Figure 4 represents the absorption data as measured by STL technology. As one notices from Fig. 4, the absorptions of the as-deposited and annealed samples are 94.6 and 40.4 ppm, respectively. At the same time, a serious fluctuation of absorption for the as-deposited sample is observed. After annealing, the absorption becomes uniform and decreases.

Microdefects are examined by a Leica-DMRXE microscope under dark field. These microdefects are on an approximately micrometer scale. Figure 5 presents the microdefect density of the samples. Five random sites on the sample surfaces are chosen and the average microdefect densities on the five sites are 14.0 and 5.0 mm^{-2}, respectively. It indicates that the microdefect density decreases after annealing.

The LIDT results of annealed samples are tested in the two regimes both at 532 and 1064 nm. As shown in Fig. 6,

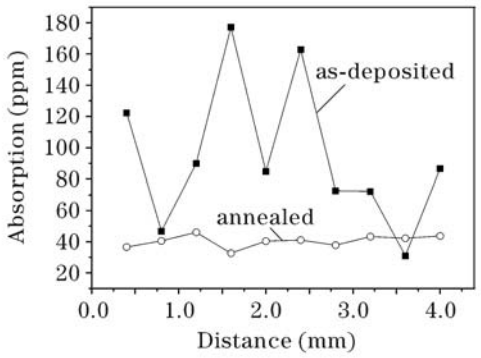

Fig. 4. Absorption of the samples.

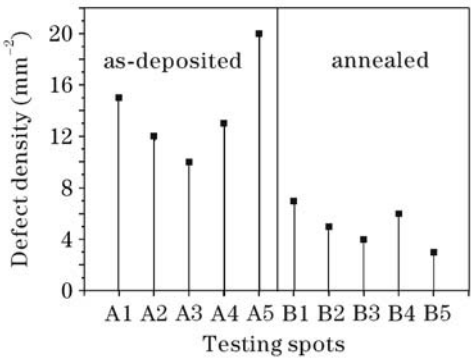

Fig. 5. Microdefect density variation of the samples.

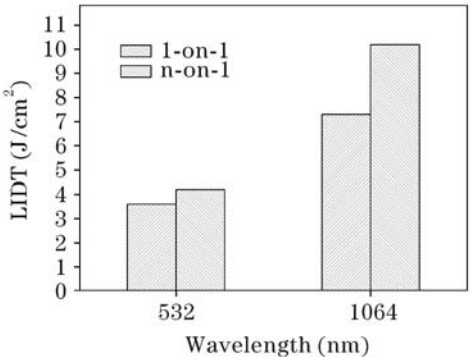

Fig. 6. LIDT results in 1-on-1 and n-on-1 regimes.

the LIDTs are 13% and 40% higher in n-on-1 regime than that in 1-on-1 regime, respectively. It has been reported that the increased LIDT in n-on-1 regime is caused by the laser conditioning effect. The laser conditioning has been linked to the removal of adsorbed water or other atmospheric contamination, and also related to the removal of electrons from intrinsic defects energetically close to the conduction bands in materials[11]. In our previous work, we attributed the laser conditioning enhancement to defect modifications, and it rooted in the improvement of defect stability under low fluence laser radiation[12].

Figure 7 presents bright-field microscopy images of damage morphologies of the as-deposited and annealed samples. The damaged spots on the sample surface are centered on a defect both in 1-on-1 and n-on-1 regimes, which implies that the damage is induced by defect in both the two regimes.

The LIDT results in n-on-1 regime at different wavelength are shown in Fig. 8. The pulse durations are 10, 0.22, and 12 ns at 532, 800, and 1064 nm, respectively. As the pulse duration is different, it is difficult to estimate the LIDT at different wavelengths. Thus, the following relation is adopted to transform all the pulse duration to 1 ns[11]:

$$D_{\rm t} \simeq D_0 t_{\rm p}^{0.30}, \tag{1}$$

where $D_{\rm t}$ is the damage threshold in J/cm^2, D_0 is a constant, and $t_{\rm p}$ is the laser pulse duration in ns.

In summary, we can get the following conclusions.

1) It is well known that in 1-on-1 regime, the LIDT always increases with the wavelength increasing[13]. We find that in n-on-1 regime, the LIDT also increases with the increase of the wavelength. From the discussion mentioned above, the LIDT enhancement in n-on-1 regime is attributed to the laser conditioning effect. According to our observation of the morphologies of the damage spots, we attribute the laser damage to the defect-initiated mechanism in the two regimes. In n-on-1 regime, though the increase of the LIDT is generated by laser conditioning effect, the damage mechanism is unchanged. Therefore, the LIDT results still increase with the wavelength increasing, which is similar to that in 1-on-1 regime.

2) Annealing influences both the optical property and the LIDT of Ta_2O_5 films. The annealed film achieves the increased LIDT at 532, 800, and 1064 nm. It shows that annealing has played a positive role in improving the LIDT at all the wavelength. One possible explanation is brought forward as follows. Since Ta_2O_5 film is always found to be non-stoichiometrically deposited by electron evaporation, in the course of annealing, oxygen diffuses and reacts with the incompletely oxidized Ta inside the film. At the mean time, the film modifies its structure of amorphous state, which accompanies with the destruction of unstable bond and the formation of new stable bond. Thus, the extinction index, absorption, and microdefect density decrease, while the LIDT increases after annealing. Researches about the LIDT at 355 and 266 nm will be done later.

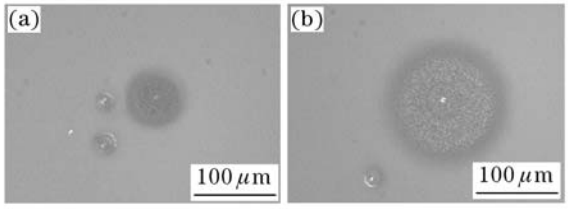

Fig. 7. Typical damage morphologies of the samples in (a) 1-on-1 regime and (b) n-on-1 regime.

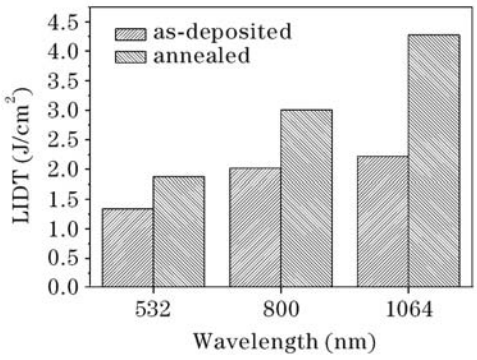

Fig. 8. LIDT at different wavelengths in n-on-1 regime with 1-ns pulse.

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