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Laser-induced damage threshold in HfO_2/SiO_2 multilayer films irradiated by β-ray*

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Post-processing can effectively improve the resistance to laser damage in multilayer films used in a high power laser system. In this work, HfO_2/SiO_2 multilayer films are prepared by e-beam evaporation and then β-ray irradiation is employed as the post-processing method. The particle irradiation affects the laser induced damage threshold (LIDT), which includes defects, surface roughness, packing density and residual stress. The residual stress that is relaxed during irradiation changes from compressive stress into tensile stress. Our results indicate that appropriate tensile stress can improve LIDT remarkably. In view of the fact that LIDT rises from 8 J/cm^2 to 12 J/cm^2, *i.e.*, 50% increase, after the film has been irradiated by 2.2×10^{13}/cm^2 β-ray, the particle irradiation can be used as a controllable and desirable post-processing method to improve the resistance to laser induced damage.

Keywords: β-ray irradiation, HfO_2/SiO_2 multilayer film, residual stress, laser-induced damage threshold

PACS: 42.79.Wc, 46.70.Hg, 79.20.Eb, 42.70.–a **DOI:** 10.1088/1674-1056/28/2/024215

1. Introduction

Multilayer optical films fabricated from hafnium dioxide (HfO_2) and silicon dioxide (SiO_2) with high laser induced damage threshold (LIDT) are important components in high power laser systems.[1–4] A key use for these films is to improve the damage resistance of high power lasers. Post-processing can effectively improve the resistance to laser damage. In general, the post-processing methods of optical films include surface processing,[3,5,6] heat treatment,[7–9] post hot isostatic pressing,[9,10] *etc*. Among these methods, surface processing can increase LIDT through improving surface quality. Heat treatment can reduce the stress and defects and thus improves the optical characteristics of the film. Post hot isostatic pressing has the advantage of relaxing the residual stress, reducing the density of defects and improving the bandgap of the films. It is well-evidenced that the residual stress, surface quality, defects significantly affect the LIDT. In addition, it has been proven that films of lower density exhibit higher LIDT.[11,12]

Particle irradiation produces defects and changes the crystal structure because of the interaction of an injected particle with the lattice atoms. Previous studies have shown that the particle irradiation can relax residual stress in thin films[13–16] and the mechanism of relaxation of residual stress can be explained by the process of atomic relocation, boundaries relaxation, atomic rearrangement, *etc*. Motivated by this encouraging result, we propose that particle irradiation is a desirable method to improve the LIDT of thin films. However, few results on this issue for HfO_2/SiO_2 multilayer films have been reported. In this paper, the effects of β-ray irradiation of HfO_2/SiO_2 multilayer films which are fabricated by e-beam evaporation are studied and the correlation of the LIDT with the fluence of β-ray irradiation is systematically explored.

2. Experimental details

HfO_2/SiO_2 multilayer antireflective film was deposited on a K9 glass substrate with a size of ϕ50 mm$\times\phi$5 mm by electron-beam evaporation. The multilayer designed structure was 0.5H1.51L1.51(HL)^{14}(HL)^{16}(3.5H)3.1L, where H and L were high index material HfO_2 and low index material SiO_2, respectively. In this work, all of the samples were simultaneously prepared under the same condition and in the same chamber. Irradiation experiments were performed at room temperature by using a ^{90}Sr β-ray source with an activity of 300 mCi in National Space Science Center. The samples were irradiated with accumulated fluxes from 10^{12}/cm^2–10^{13}/cm^2 as shown in Table 1.

The transmittance spectra were measured by a spectrophotometer (NKD 7000, Aquila, UK) at an incident angle of 45° over the spectral range of 280 nm–1300 nm. The surface morphology was investigated through an atomic force microscope (AFM, Dimension 3100, Bruker Nano Inc.) in the contact mode. Multilayer stress was measured by the surface deformation through using a ZYGO Mark III-GPI optical

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interferometer. The photoluminescence of HfO_2/SiO_2 films before and after irradiation were tested by using a LabRam HR 800 Raman spectrometer. According to the standard of ISO-11254, LIDT test was performed in the '1-on-1' mode by using a 1064-nm Nd:YAG pulse laser with a pulse width of 8.9 ns.

Table 1. β-ray irradiation fluences.

Sample	Irradiation fluence/cm^2
#1	As-deposited
#2	4.2×10^{12}
#3	8.5×10^{12}
#4	2.2×10^{13}

3. Results and discussion

The transmittance spectra of HfO_2/SiO_2 films before and after being irradiated by β-ray with different fluences are shown in Fig. 1. It can be seen that the transmittance at 1064 nm reaches to 99.03% before being irradiated and decreases slightly ($< 0.87\%$) with increasing irradiation fluence. In the short-wavelength region, the transmittance decreases apparently, which can be attributed to the color centers generated by radiation-induced defects; as illustrated in Fig. 1. The absorption of light is in consistence with the concentration of radiation-induced color centers.

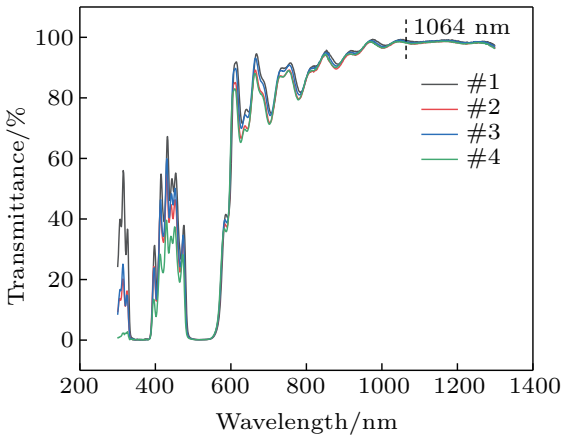


Fig. 1. Transmission spectra of HfO_2/SiO_2 films before and after being irradiated by β-ray.

Figure 2 shows photoluminescence (PL) spectra of HfO_2/SiO_2 films irradiated by different-fluence β-ray, which are excited by laser with an energy of 5.2 eV. The spectrum consists of five wide bands after the film has been irradiated by β-ray, of which the maxima lie in the vicinity of wavelengths of 299 nm–305 nm, 387 nm–405 nm, 492 nm–497 nm, 529 nm–537 nm, and 569 nm–573 nm. The intensity of 387–405 nm wavelength band relates proportionally to radiation fluence and becomes saturated in the high fluence region. Previous studies have concluded that the change in PL spectrum in oxide film related to the oxygen vacancy defects and microstructure transformation.[17,18] High-concentration oxygen vacancy defects cause both PL intensity and light absorption to increase, which are in agreement with the results of transmittance spectra in Fig. 1. The defects are saturated when the radiation-induced damage starts to overlap.

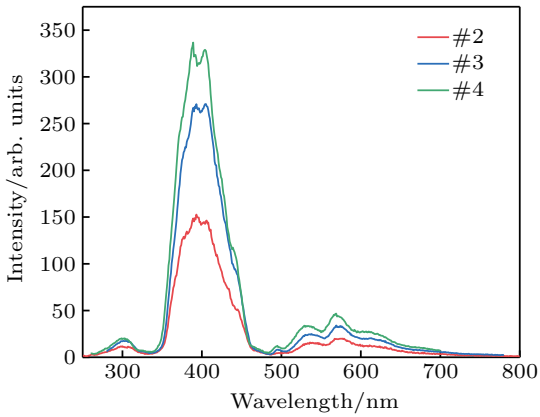


Fig. 2. Photoluminescence spectra and intensities of HfO_2/SiO_2 films after being irradiated by β-ray.

Figure 3 shows AFM images of the as-deposited HfO_2/SiO_2 films before and after being irradiated. The root mean square (RMS) values of the samples are listed in Table 2. It can be obtained from Table 2 that the surface roughness values of HfO_2/SiO_2 films increase after being irradiated. The increase of the surface roughness after being irradiated is probably due to the rearrangement of atoms caused in the process of the electron's transportation. Larger RMS generally indicates more surface defects and more heat absorption in laser-irradiation. In our experiments, the maximum RMS roughness is 5.652 nm at an irradiation fluence of 8.45×10^{12}/cm^2.

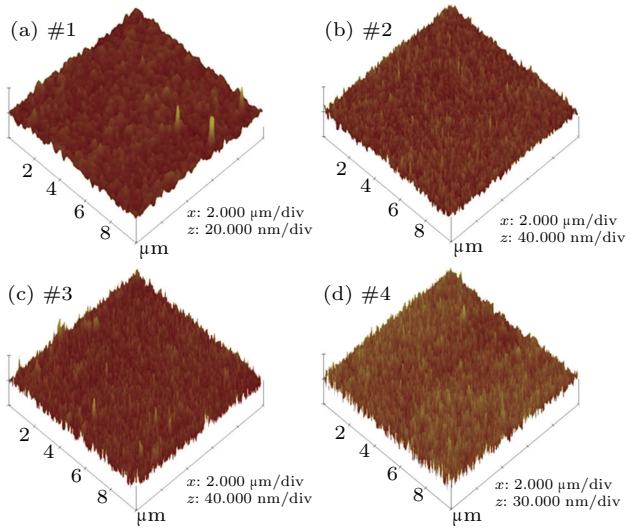


Fig. 3. Atomic force microscope images of HfO_2/SiO_2 films before and after being irradiated by β-ray.

Table 2. Properties of HfO_2/SiO_2 films before and after irradiated by the β-ray.

Sample	Transmitance (1064 nm)/%	RMS/nm	Power ($\lambda = 632.8$ nm)
#1	99.03%	1.959	–0.415
#2	98.16%	3.789	–0.132
#3	98.56%	5.652	0.379
#4	98.20%	5.087	0.025

The surface deformations of the HfO_2/SiO_2 films before and after irradiation are shown in Fig. 4. One can see that the surface shape of as-deposited film is convex and becomes concave after being irradiated. This phenomenon probably results from the increase of packing density.[19,20] In our case, convex surface denotes the compress stress and concave surfaces represents the tensile stress. The measurements of power values are listed in Table 2.

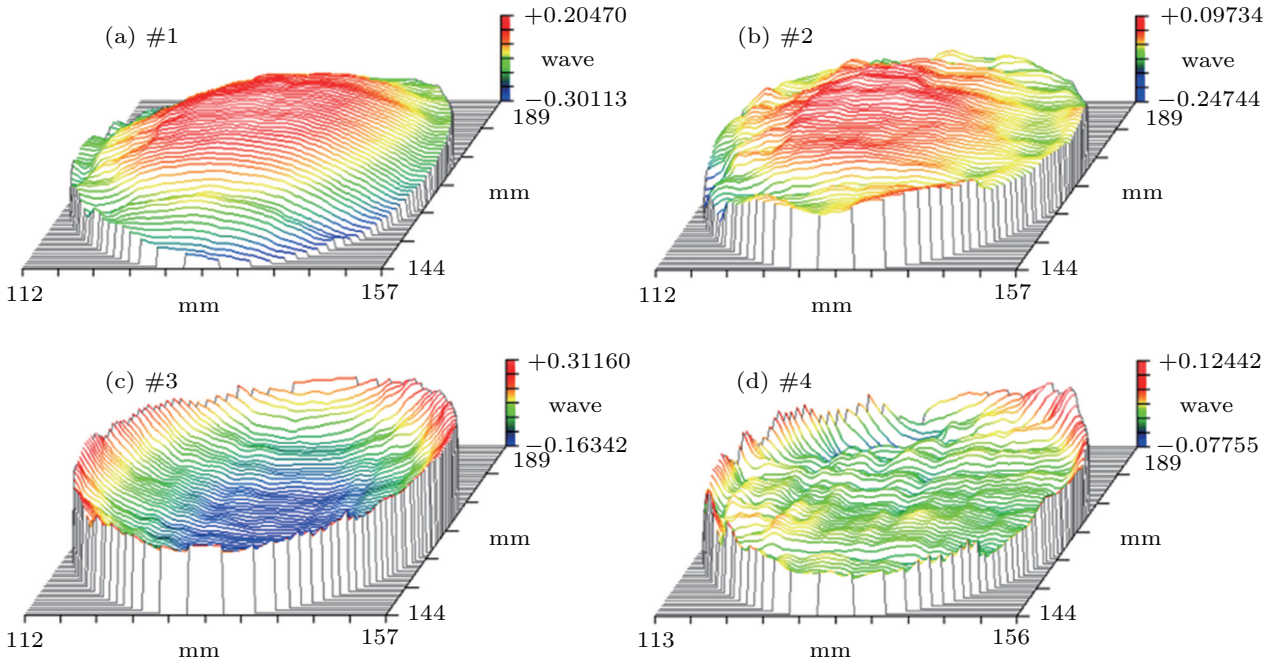


Fig. 4. Surface deformations of HfO_2/SiO_2 films before and after being irradiated by β-ray.

The residual stress can be calculated from the Stoney's formula:[7,21]

$$\sigma = \frac{E}{6(1-\nu)}\frac{t^2}{t_f}\left(\frac{1}{R_0}-\frac{1}{R}\right) = \frac{4E}{3(1-\nu)}\frac{t^2}{D^2}\frac{\Delta Power}{t_f}, \quad (1)$$

where σ is the residual stress of the film, ν is the Poisson's ratio, E is the Young's modulus, D is the diameter of the substrate, t is the thickness of the substrate, t_f is the thickness of the film, R and R_0 are the curvature radius of the substrate before and after deposition. Accordingly, the variation of residual stress can be expressed as

$$\begin{aligned}\Delta\sigma &= \sigma_i - \sigma_0,\\ \Delta\sigma &= \frac{E}{6(1-\nu)}\frac{t^2}{t_f}\left(\frac{1}{R_i}-\frac{1}{R_0}\right)\\ &= \frac{4E}{3(1-\nu)}\frac{t^2}{D^2}\frac{(Power_i - Power_0)}{t_f},\end{aligned} \quad (2)$$

where i refers to the sample after irradiation. The calculated residual stresses of the films after being irradiated varying with the stress after deposition are shown in Fig. 5. The maximum variation of stress reaches 231 MPa, and the tensile stress is formed in the film. It can be seen from Figs. 4 and 5 that the compressive stress generated during deposition is relaxed, and thus changing into tensile stress with the stress variation after the film has been irradiated. The procedure of stress evolution in our experiments can be divided into at least two stages; *i.e.*, compressive stress relaxation and tensile stress transformation. The compressive relaxation process relates to atom displacement and microstructure rearrangements governed by collision between electron and lattice atoms. The microstructural rearrangement is responsible for compression relax effect. In the displacement process, interstitial atoms and vacancies are generated and when the irradiated fluence is greater, the concentration in the film is higher, which is consistent with the results of Fig. 2. Finally, the stress changes into tensile stress. Thereafter, the local vacancies begin to coalesce, leading to the relaxation of tensile stress. We can conjecture that HfO_2/SiO_2 film with zero stress could be obtained by tuning the irradiation parameter.

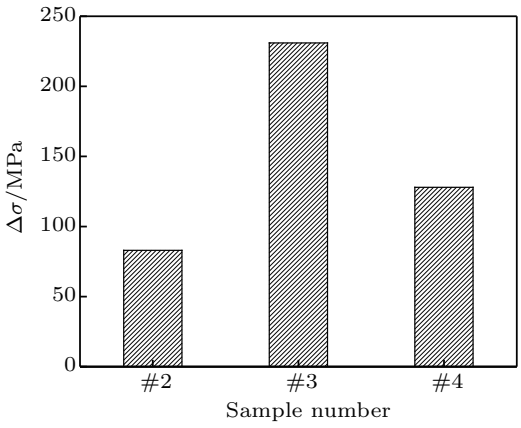


Fig. 5. Calculated residual stress variations for HfO_2/SiO_2 films after being irradiated by β-ray compare with stress induced after deposition

Figure 6 shows the LIDT values of the HfO_2/SiO_2 films before and after being irradiated. The values of LIDT in the

figure are all at the 0% probability damage threshold, which is the maximum peak energy density that the film can withstand. The LIDT value of the as-deposited film is 8 J/cm^2 and exhibits a weak change after the film has been irradiated by a fluence of 4.2×10^{12}/cm^2. As the irradiation fluence increases to 8.5×10^{12}/cm^2, the LIDT decreases by 25% compared with the LIDT of the as-deposited film. Then, the LIDT increases to 12 J/cm^2, with a 50% increase, after the film has been irradiated by a fluence of 2.2×10^{13}/cm^2.

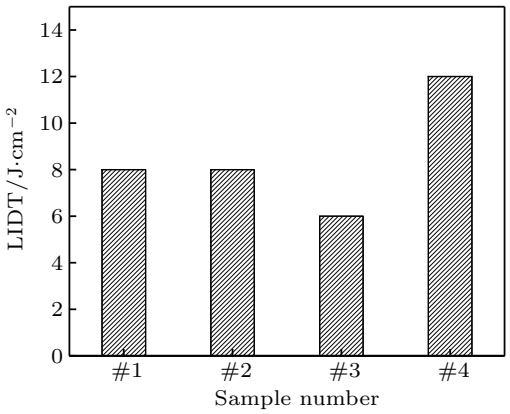


Fig. 6. LIDT for HfO_2/SiO_2 films before and after being irradiated by β-ray.

It is well known that the LIDT relates to multiple factors. The thermal-mechanical damage process is generally regarded as a dominant damage process of dielectric thin film in a nanosecond pulse width region. One of the most useful parameters to predict the laser induced damage to thin-film is the laser-induced thermal stress caused by fast heating for laser absorption. In this model, the thermal stress can be expressed as[12]

$$\Delta P=\alpha\frac{E}{3(1-2v)}\Delta T, \tag{3}$$

where α is the coefficient of thermal expansion, v is the Poisson's ratio, E is the Young's modulus, and ΔT is the temperature rise caused by thermal absorption of defects during laser irradiation. Generally, more thermal absorption of defects leads to large ΔT. In Eq. (3), ΔT can be approximated as

$$\Delta T(t)=(M/K)\ln(1+QKt), \tag{4}$$

where t is the time during which the stress pulse propagates from the absorption point to the free surface, M and Q are constants relating to the laser energy, and K is the thermal conductivity of the film. In this formula, QKt is lower than 1, then $\Delta T\approx MQt$. Under this condition, ΔT is independent of thermal conduction and linearly relates to t. Less dense film exhibits smaller t, thus smaller thermal stress. Besides radiation-induced defects, surface defects can be considered as absorption sites. The more the defects in the film or at the surface, the more efficient the heating will be to produce damage in the film; as illustrated in Eq. (3). Thermal conductivity indicates the capability of thermal expanding outward from the center of laser irradiation. Packing density in this study relates to thermal conductivity. The thermal conductivity increases as the packing density decreases; as illustrated in Eq. (4). Highly inner stressed multilayers can cause irreversible damage such as peeling, cracking and curling during laser irradiation. The best strategy to improve the resistance to laser-induced damage is to control the total stress and to fix it close to zero. Total stress includes residual stress in the as-deposited film and thermal stress induced by laser irradiation. The thermal stress induced by laser irradiation is always existent in compressive form; therefore, appropriate residual tensile stress will lower total stress in the film and, hence, higher LIDT for the film could be achieved.[22] Particle irradiation is a controllable way to modify the material properties and it can improve the LIDT of optical thin film controllably.

Therefore, the discrepancy in LIDT in this study can be explained. Although the compressive stress in sample #2 is relaxed after being irradiated by β-ray, high concentration defects increase the heat absorption and produce greater thermal stress. Consequently, the LIDT of sample #2 shows no change compared with that of sample #1. Lower LIDT in sample #3 is mainly caused by highly tensile stress due to the packing density increasing and the number of defects increasing. The higher LIDT in sample #4 is mainly caused by appropriate tensile stress in the film. Therefore, appropriate tensile stress in the thin film can achieve high LIDT, which is consistent with the result obtained by Ling *et al.*[22]

4. Conclusions

In this paper, β-ray irradiation is employed as a post-processing method for improving performance of HfO_2/SiO_2 multilayer films. The transmittance spectrum, PL intensity, surface morphology, and the LIDT of the film are tested before and after irradiation with different fluences. The results show that the irradiation affects the multiple factors relating to laser-induced damage, such as defects, surface roughness, absorption, packing density, and residual stress. The LIDT increases from 8 J/cm^2 to 12 J/cm^2 after the film has been irradiated by a fluence of of 2.2×10^{13}/cm^2. The residual stress relaxes during the β-ray irradiation and changes from compressive stress to tensile stress. Irradiation-induced appropriate tensile stress which may improve LIDT. The results reveal that β-ray irradiation can be a controllable post-processing method to improve the LIDT of multilayer thin films.

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