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Abstract: The coupling model of defect and organic contamination was presented to analyze the coupling effects on laser-induced damage and understand the damage degradation mechanism of optical films in a closed vacuum environment. The distributions of temperature rise around an absorbing defect in multilayer ZrO_2/SiO_2 films were computed based on the finite-element simulation. Laser induced damage tests were made to compare with the theoretical model. Test results were in an agreement with the presented model and calculated results of a temperature rise.

Index Terms: Vacuum, laser-induced damage, defect, organic contamination.

1. Introduction

Optical films used in vacuum and space environments that can sustain higher laser fluencies are driven by the recent advancements in high power laser facilities. In vacuum, low laser induced damage threshold and short service lifetime of optical films have been revealed in many studies [1]–[3]. Improving the performance and increasing the laser-induced damage threshold (LIDT) of optical film components in vacuum have been the basic pursuing objectives. Organic outgassing contaminants in closed vacuum environments have suspected to be a critical factor for laser induced damage degradation behaviors [4]–[6]. The damaged degradation mechanisms, such as photo-induced chemical damage, thermo-mechanical damage, micro-lensing effect mechanism and so on [7]–[9], have been proposed to explain the corresponding damaged behavior. However, for the nanosecond laser pulses, laser-induced damage of optical films is defect-related damage, and the number of the sensitive defects determines the damage probability and the LIDT of the samples. And studies have also shown that defects and organic contaminants interact on the laser induced damage of optical film when organic contamination is adsorbed on the surface of the film [10]. However, less attention is paid to the coupling effects of defect and organic contamination on laser induced damage of optical films, especially in closed vacuum environments.

In this paper, we presented the coupling model of defect and organic contamination to understand the coupling impact of defect and different organic contamination modes on the laser induced damage. The evolutions of temperature rise around defects in multi-layer ZrO₂/SiO₂ films were



Fig. 1. The coupling model of defects and organic contaminations.

TABLE 1 Summary of Parameters Used for the Calculations

	Refractive	Extinctive	Specific	Heat	Thickness
Layer	Index	Coefficient	heat	conductivity	
	Ν	Κ	С	Κ	d
			$(J/cm^{3}.^{0}C)$	$(W/cm.^{0}C)$	(nm)
H(ZrO ₂)	1.89	5x10 ⁻⁵	2.7	$1.8 \mathrm{x} 10^{-4}$	60
L(SiO ₂)	1.46	1×10^{-5}	2	$1.7 \mathrm{x} 10^{-3}$	85
a(toluene)	1.49	0	1.479	1.38×10^{-4}	0/10/50
G (glass)	1.52	0	2	0.014	1000

calculated based on the finite-element simulation. Laser induced damage tests results agreed well with the presented model and calculated results of temperature rise.

2. Theoretical Model

There are two different deposition modes for organic contamination on the optical films. One is a deposited organic contaminations layer caused by classic condensation of organic contaminant on a surface of optical film, the other is organic contamination droplet adsorbing on the surface of optical film. We mainly pay our attention to transparent or weak absorption organic contamination for irradiated laser, which accords to the practical situations in vacuum laser systems. The coupling model of defects and organic contaminations is shown in Fig. 1. As has been mentioned before, in nanosecond pulse width region, laser induced damage of dielectric thin films was mainly laserinduced thermal damage initiated by nanometric absorbing defect. So, we simulated the temperature rise induced by an absorbing inclusion in ZrO_2/SiO_2 high reflective films. The temperature evolution in the spherical defect with the size of 30 nm can be obtained by the program of a finite-element algorithm. We considered the changes of the temperature rise around the defect when adsorbing toluene organic contamination layer and organic contamination droplet.

3. Results and Discussions

3.1 Organic Contamination Layer

Based on the finite-element algorithm, we computed the temperature rise around an absorbing defect in the ZrO_2/SiO_2 HR coatings with adsorptive organic contamination layer of different thickness irradiated by 1064 nm laser with energy density of 10 J/cm². The thermal parameters used for the simulations are shown in Table 1 [10].



Fig. 2. Finite-element simulation without (left) and with (right) adsorptive organic contamination layer.



Fig. 3. The temperature rise with adsorptive organic contamination layer of different thickness.

Fig. 2 showed the finite-element simulation result with two dimensional distributions due to adsorptive organic contamination layer of 50 nm. The temperature rise with adsorptive organic contamination layer of different thickness was plotted as a function of distance from the center of the defect, as shown in Fig. 3. Fig. 3 showed the maximum temperature rise had little increase with increasing thickness of surface adsorptive organic contamination layer. However, the thermal diffusion regions from the center of defect become smaller with increasing thickness of surface adsorptive organic contamination layer from Figs. 2 and 3. It is believed that the occurrence of laser-induced thermal damage of optical films means the temperature rise exceeding critical value of damage. Different thickness toluene organic contamination layer induced almost the same maximum temperature rise around a defect, which denoted that inter-coupling effects between defect and organic contamination layer had little impact on the thermal damage of optical film.

3.2 Organic Contamination Droplet

K. Bien-Aime'studies [14] showed that adsorptive organic contamination droplet acted as a microlensing for irradiated laser, inducing a local increase of power density. Droplets were considered



Fig. 4. Finite-element simulation without (left) and with (right) adsorptive organic contamination droplet.



Fig. 5. The temperature rise with adsorptive organic contamination droplet.

to be spherical. So, the focal distance is $F = \frac{n}{n-1}R$ for a spherical droplet, where *R* is the radius of curvature, *n* a refractive index of the droplet. In our cases, the curvature radius of the toluene droplet is more than ten microns in magnitude. So, the droplet with radius of 15 μ m attains a focal distance of 45.6 μ m. A single longitudinal mode, Gaussian-shaped laser beam with the spot size ϖ_0 of 300 μ m on the sample was incident perpendicularly on the droplet, so the spot size ϖ' of the transmission Gaussian-shaped laser beam was followed as [15]:

$$\overline{\omega}' = \frac{\omega_0}{\sqrt{1 + \left(\frac{\pi \omega_0^2}{\lambda F}\right)^2}} \tag{1}$$

According to equation (1), the spot size of the transmission Gaussian-shaped laser beam will reduce to 51 μ m. It's six times smaller than before. So, the laser power density incidence on the optical film due to adsorbing the organic contamination droplet increases by a factor of 6.

Figs. 4 and 5 showed the finite-element simulation result and corresponding temperature rise of the ZrO_2/SiO_2 HR coatings when adsorbing organic contamination droplet of 15 μ m radius. It can



Fig. 6. Tests of 0% damage probability in vacuum.

be found that organic contamination droplet had the significant impact on the temperature rise of HR coating, and the temperature rise was increased by a factor of more than 5, compared with clean sample.

3.3 Laser Induced Damage

Laser induced damage was tested by 1-on-1 mode for ZrO₂/SiO₂ HR film deposited by electron beam evaporation. Toluene organic contamination layer and toluene organic contamination droplet was introduced to adsorb on the deposited ZrO₂/SiO₂ film, respectively. And the laser induced damage experimental setup, test method and contamination experiment can be seen in the paper [16], [17]. The intentional organic contamination layer is realized in a qualified Micro-chamber with a controlled flow rate. The desired quantity of contaminant is placed in the chambers. The sample is placed on the sample holder that avoids direct contact between the sample and the contaminant. Toluene organic contamination droplet is carried out by spraying the liquid toluene on the surface of the sample by a tube with a desired diameter like a syringe and at the same time, the sample spins with a speed of about 700 revolutions per minute during 30 seconds. For the damage test, the surface area of the film is divided into a matrix of sites, which are well separated in space. A constant number of sites (200 test sites in our experiments) are irradiated with one preselected pulse energy and one pulse per site. For each site pulse, energy and the state of the site (damaged or non-damaged) are recorded. After the exposure, the damage probability is calculated. Then the pulse energy is increased and the sequence is repeated for another test sites. During measurement pulse energy is increased until 100% damage probability is reached. The laser-induced damage probability of the ZrO₂/SiO₂ high reflection film with toluene organic contamination layer with the thickness of about 50 nm shown in Fig. 6 had little change, compared with clean film. Fig. 6 also showed there was about a 1.5 times decrease in 1-on-1 laser-induced damage threshold for the HR film when adsorbing toluene organic droplet.

The crater like features was observed in the scanning electron microscopy (SEM) SEM image in Fig. 7. It was attributed to the plasma formation and expansion that was accompanied by heating due to laser irradiation. The expanding plasma propagated along the direction of the laser incidence and finally a regular crater formed due to defect ejection. The SEM images shown in Fig. 7 also revealed that adsorbing toluene organic droplet induced a serious damage with a mass of ablation



Fig. 7. SEM image with organic contamination droplet (top) and organic contamination layer (bottom).

material in damaged zone around defects, compared with adsorbing toluene organic contamination layer. Larger magnification images denoted that the severe ablation and spalling of film layer occurred in defect sites due to toluene organic droplet. Test results agree well with the model above-mentioned and calculated results of temperature rise. So, the inter-coupling between defect and organic contamination droplet induced high temperature rise in optical film and was accordingly attributed to the decrease of the laser-induced damage threshold.

4. Conclusion

A coupling model of defect and organic contamination was presented to understand the damage degradation mechanism of optical films in vacuum exposed to organic contaminations. The distributions of temperature rise due to the coupling of defect and organic contamination in multi-layer ZrO₂/SiO₂ films were computed by finite-element simulation. Laser induced damage tests were made to compare with the theoretical model. According to our results, the inter-coupling between defects and organic contamination droplet resulted in a fairly high temperature rise around defects, and then aroused the decrease of the laser-induced damage threshold. While, the inter-coupling between defects and organic contamination layer adsorbing on the surface induced nearly the same temperature rise around defects and had little effect on laser-induced thermal damage of ZrO₂/SiO₂ films, compared with clean film.

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