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Wu Zhouling, Fan Zhenxiu, et al.

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Damage Threshold Dependence of Optical Coatings on Substrate Materials

Wu Zhouling, Fan Zhenxiu, Wang Zhijiang

(Shanghai Institute of Optics and Fine Mechanics,
Academia Sinica)

Gao Yang

(Shanghai Institute of Applied Mechanics)

Abstract: Damage threshold dependence on substrate materials was investigated for TiO_2 , ZrO_2 , SiO_2 , MgF_2 , ZnS , and single and $\text{TiO}_2/\text{SiO}_2$ multilayers. The results show that the damage threshold increases with increasing substrate thermal conductivity for single layers and AR coatings and remains the same for HR coatings. With the help of localized absorption measurement and in-situ damage process analysis, these phenomena were well correlated with local absorption-initiated thermal damage mechanism.

1. Introduction

Technically, optical films are the most fragile components to suffer from damage in a laser system. In research on high-power laser technologies, including laser nuclear fusion and laser defense weapons, film damage tends to be a major factor that affects the size of laser devices and output energy level. Consequently, studying intensive laser damage of film and constantly enhancing its resistance to the laser beam become vitally important in research and development of strategic defense weapons as well as in improvement of intense laser systems and expansion of their applications to research and production.

Laser damage to film is a result of interaction between film and laser, associated with both film and laser. As for the film,

there are two factors that substantially affect its optical features, namely the film layer and the substrate. In order to decrease the optical loss in the film and to increase its damage threshold, tremendous efforts have been made in the past few years in selecting substrate materials and in developing substrate preparatory techniques, such as developing a new polishing technology [1-5], improving cleaning technology [6-8], baking the substrate in a vacuum before being coated [9-13] and laser radiation processing of the substrate [11-14]. Despite these efforts, which have led to an improvement of the optical quality of film in many cases, a thorough understanding is yet to be attained on the law of correlation between substrate features and film damage thresholds, as well as on the mechanism that controls such correlation.

Taking as examples, TiO_2 , ZrO_2 , SiO_2 , MgF_2 , ZnS single-layer films, and $\text{TiO}_2/\text{SiO}_2$ multilayer film prepared with electron beams and resistive thermal evaporation, this paper outlines an experimental study on the effect of different substrate materials such as molten quartz (SiO_2), sapphire (Al_2O_3) and calcium fluoride (CaF_2) on the near-infrared laser damage thresholds of optical films, and explains satisfactorily some phenomena related to the local absorption-initiated thermal damage mechanism, coupled with body/surface absorption measurements and analysis of the transient damage process.

2. Experimental Method

In the experiment, sample films were deposited on different substrate materials, which, before being coated, had undergone polishing and cleaning with regular combined polishing and cleaning techniques. Sample film systems and coating technologies are shown in Table 1. All the sample films made of the same material as listed in Table 1 were deposited under the

same cover so that effect of factors other than film material on film damage threshold could be ruled out.

Table 1. Experimental sample films and their preparatory techniques ($\lambda_0=1.06\mu\text{m}$)

膜层材料 1	膜系结构 2	4 基板材料	8 膜层沉积工艺
MgF ₂ ZnS	3 单层膜 $n_s = \lambda_0/2$	5 熔石英(SiO ₂)	9 电阻热蒸发 $P = (2\sim 3) \times 10^{-5} \text{ } ^\circ\text{C}$
TiO ₂ ZrO ₂ SiO ₂		6 蓝宝石(Al ₂ O ₃)	10 电子束热蒸发 $T_s = 250 \text{ } ^\circ\text{C}$ $T_A = 200 \text{ } ^\circ\text{C}$
SiO ₂ /TiO ₂ TiO ₂ /SiO ₂		7 氟化钙(CaF ₂)	
	A(LH)G		
	A(LH) ¹³ HG		

Key: 1. Film materials; 2. Film system structure; 3. Single-layer film; 4. Substrate materials; 5. Molten quartz; 6. Sapphire; 7. Calcium fluoride; 8. Film precipitation techniques; 9. Resistance heat evaporation; 10. Electronic beam heat evaporation;

Threshold measurements were conducted on our own experimental equipment [15]. The laser system consisted of an Nd:YAG oscillator and a bipole Nd:YAG amplifier. The oscillator performed Q-modulation with a LiF crystal and film selection with a small hole diaphragm, and its output beam was at the wavelength $1.06 \mu\text{m}$ with a pulse width of (FWHM) 10 ns, operating in the single-mode state. The incident laser beam was focused on the sample film surface by an image error-erasing, aspherical lens ($f=80 \text{ mm}$) with a flare diameter (I_0/e^2) of $44 \mu\text{m}$. Damage measurements were made in a one-on-one manner, i.e. the same location on the film surface was irradiated by a laser only once,

no matter whether or not this point was really damaged. The film damage threshold was defined as the initial damage threshold [16] which corresponded to zero damage geometric rate. Compared with the conventional definition of the threshold corresponding to 50% damage geometric rate, this definition has the obvious merit in that it can rule out the effect of light speckling [17]. While film damage was defined as an observable, irreversible physical change of the film after it was irradiated by a laser, in our experiment, damage was measured using the light deflection continuous modulation technique [15-18], in which damage referred to an irreversible change to the film light deflection modulation signal.

Absorption measurements were carried out with the repetition-frequency pulsed photothermal deflection technique [19, 20]. Its arrangement was coupled with the damage system [18], available for real-time analysis.

Bulk film absorption and interfacial absorption were achieved through changing single-layer film thickness and multilayer system structure. This experimental method is detailed in [21, 22] and thus will not be discussed here in this paper.

The transient damage process was studied along with the time-resolution pulsed light deflection technique. By analyzing the location of pulsed light deflection signal peak value on the time axis, the damage position can be accurately determined--whether on film surface or inside film or on the interface between film and substrate [15].

3. Experimental Results

Measurements of sample film absorption and damage thresholds are given in Table 2 and Fig. 1.

Table 2. Measurements of absorptance and thresholds of sample films

1 膜层材料	2 吸收率 $A(10^{-4})$			4 阈值 $F_{th}(Jcm^{-2})$		
	3 基板材料: SiO ₂ CaF ₂ Al ₂ O ₃			SiO ₂	CaF ₂	Al ₂ O ₃
MgF ₂	5.0	5.1	4.9	12.8±2.1	16.8±2.3	24.8±1.9
ZnS	15.7	14.9	16.2	4.8±0.9	—	12.3±1.2
TiO ₂	12.5	12.6	13.1	7.8±1.2	10.5±2.0	16.2±1.3
ZrO ₂	7.2	7.6	7.4	11.3±2.3	16.3±1.9	20.3±2.1
SiO ₂	2.3	2.0	2.5	18.3±1.0	19.8±1.3	27.5±2.0
AR: SiO ₂ /TiO ₂	11.3	12.1	11.6	4.3±1.8	8.2±2.5	15.3±2.7
HR: TiO ₂ /SiO ₂	6.8	7.2	6.3	8.8±2.1	8.6±2.7	8.4±2.6

Key: 1. Film materials; 2. Absorptance; 3. Substrate materials; 4. Thresholds

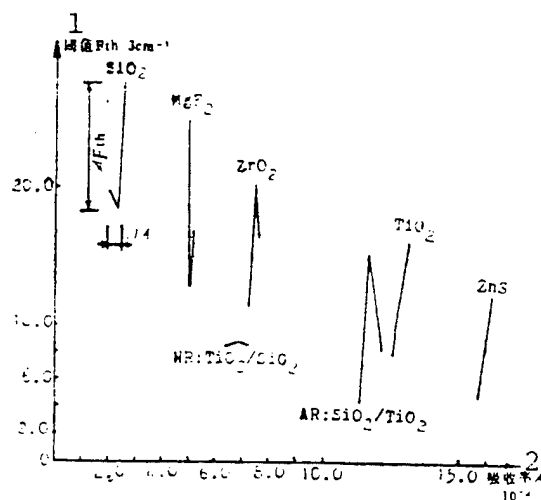


Fig. 1. Graphic presentation of experimental results listed in Table 2: dependence of thresholds on absorptance

Key: 1 Thresholds; 2. Absorptance

Table 2 and Fig. 1 show that the same sample film used to coat different substrates under the same technological conditions exhibits basically the same absorptance, but entirely different thresholds, which proves that the major factor affecting film damage threshold is substrate material rather than total absorptance of the film.

Based on a detailed study of the threshold measurements shown in Table 2, a correlation between those values and substrate thermal conductivity is indicated in Fig. 2. It can be seen from Fig. 2 that the thresholds of all sample films rise with increase in thermal conductivity except for film number 21--the $\text{TiO}_2/\text{SiO}_2$ high inversion film, which suggests that substrate thermal conductivity plays a significant part in the damage process of single-layer films and higher transparency films.

4. Analysis of Experimental Results

Generally, thermal conductivity is several orders lower than thermal conductivity of corresponding bulk materials. Under the action of a 10 ns pulsed laser, film thermal diffusion length ranges from merely orders of several to dozens of angstroms [23]. Thus, if the film-substrate interface is subject to greater local absorption, its heat primarily diffuses toward the substrate, as shown in Fig. 3.

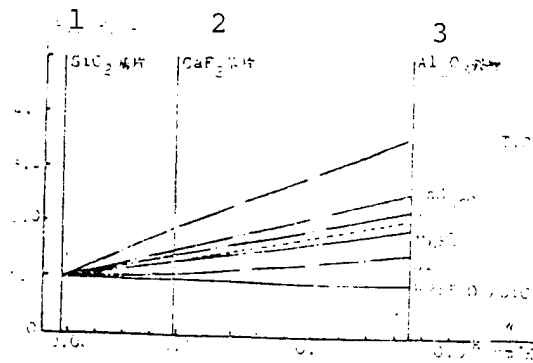


Fig. 2. Correlation between film damage thresholds and substrate thermal conductivity; K-substrate thermal conductivity; F_{th}/F_{th}^{-1} —normalization of film damage threshold over film with quartz glass substrate.

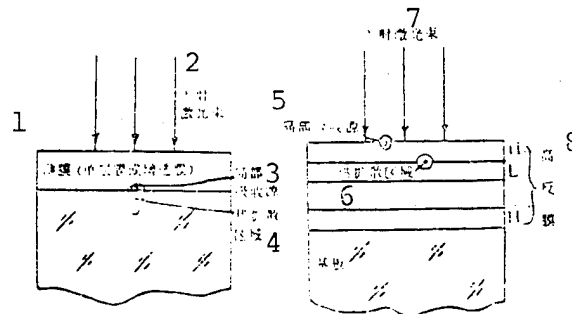


Fig. 3. Block diagram of film interface thermal diffusion: film-substrate interface indicates a rather intensive optical field and high absorptivity and substrate characteristics impose a strong effect on thermal diffusion for single-layer film and higher transparency film, while for high-inversion film, the local intensive absorption source is usually located at the film-air interface or the outermost H-L interface and the optical field at the film-substrate interface is basically equal to zero and therefore, the substrate characteristics in principle do not affect thermal diffusion

Key: 1. Films (single-layer film and higher transparency film); 2. Incident laser beam; 3. Local absorption source; 4. Thermal diffusion region; 5. Local absorption source; 6. Thermal diffusion region; 7. Incident laser beam; 8. High-inversion film

Thus, if local absorption-initiated thermal damage serves as the major mechanism for film near-infrared laser damage, effect of substrate thermal conductivity on the film damage threshold can easily be comprehended.

To further substantiate the local absorption-initiated thermal damage mechanism, a study made of the body/surface absorption, transient damage process and high-inversion film, and the protective film effect. The study results are highlighted as follows:

1) For single-layer films ZrO_2 , MgF_2 and ZnS , film-substrate interface absorption, air-film interface absorption and film in-body absorption are of the same order of magnitude, while for single-layer films TiO_2 and SiO_2 , film-substrate interface absorption, much larger than air-film interface absorption and film in-body absorption, is the major source for absorption loss [21]. When related sample films interact with the laser, the energy precipitation density at their interface should be much higher than that within the film as long as the thinness of film interface is considered.

2) Analysis of the transient-damage process shows that initial damage mostly takes place at the film-substrate interface in the case of single-layer film and higher-transparency film, while for high-inversion film, it usually occurs at the air-film interface [14]. This agrees with the results of studying substrate thermal conductivity as described in this paper.

Figure 4 shows the time-resolution pulse light deflection signals generated during critical damage to two typical sample films.

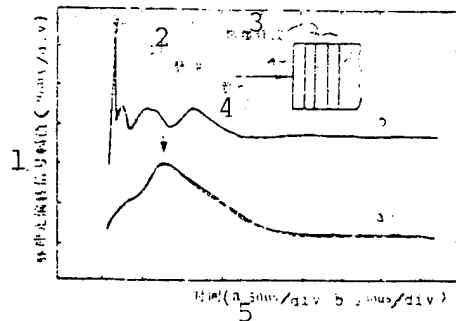


Fig. 4. Time resolution pulse light deflection signals during critical damage of sample films

a - TiO_2 coating on molten quartz substrate;
b - $\text{TiO}_2/\text{SiO}_2$ high-inversion film coating on molten quartz substrate

The arrow indicates the location of peak value on time axis, which corresponds to the position of thermal explosion damage inside film during critical interface damage

Key: 1. Amplitude of pulse light deflection signal;
2. Pulse; 3. Thermal explosion wave; 4. [illegible];
5. Time

3) Protective film can substantially increase the laser damage threshold of high-inversion film [24] but appears much less effective for higher-transparency film, which suggests that local thermal protection at the film-air interface is the major reason that protective film can raise the high-inversion film damage threshold (thermal conductivity of protective film is much larger than that of air). This local thermal protection mechanism corresponds closely to substrate thermal conductivity effect, as discussed in this paper.

5. Conclusion

Technically, substrate material has a rather obvious effect on the near-infrared laser damage threshold of single-layer

medium film and double-layer higher-transparency film. The general law of such influence is that threshold rises with the increase in substrate thermal conductivity. This experimental result, together with authors' earlier research on substrate preprocessing technology [14] and the effect of protective film [24] on the optical film damage process can provide rather convincing support to the "local absorption-initiated thermal damage" mechanism.

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On the authors: Wu Zhouling, male, born on November 2, 1964 is a Ph.D graduate who is now engaged in research on interaction between film optical laser and materials, photoacoustic/photothermal spectroscopy and optical information processing.

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