Stress mechanism of pulsed laser-driven damage in thin film under nanosecond ultraviolet laser irradiation

Zhenkun Yu (于振坤)^{1,2}, Hongbo He (賀洪波)^{1*}, Xu Li (李 煦)^{1,2}, Hongji Qi (齐红基)¹, and Wenwen Liu (刘文文)^{1,2}

¹Key Laboratory of Materials for High Power Laser, Shanghai Institute of Optics and Fine Mechanics,

Chinese Academy of Science, Shanghai 201800, China

² University of Chinese Academy of Sciences, Beijing 100049, China

 $^{*}Corresponding author: hbhe@siom.ac.cn$

Received January 22, 2013; accepted April 12, 2013; posted online July 3, 2013

An analytical model is derived to describe the stress mechanism in a thin film against the laser-induced damage threshold (LIDT) based on the thermal transfer equation. Different structures of high-reflection films at 355 nm are prepared to validate this model. LIDTs are found to have a linear relationship with stress. Furthermore, predictions from the simple model agree with the experiments.

 $OCIS \ codes: \ 310.0310, \ 140.0140, \ 310.1620, \ 140.3440, \ 140.3380.$

doi: 10.3788/COL201311.073101.

Pulsed laser-driven damage on thin film has been studied over the past five decades. In nanosecond (ns) scale, the thermal effect of thin film is considered as the main reason for laser-induced damage^[1,2]. However, the role of stress in this process has not yet to be fully understood because the initial source of damage is complex. Further research is required to elucidate the relationship between stress and laser-induced damage threshold (LIDT). Stress has certain effects on threshold^[3-6]. For example, tensile stress often has a negative effect on threshold, that is, higher tensile stress is based on a lower damage threshold. Nevertheless, few works systematically explain the relationship between stress and threshold.

In this letter, we aim to derive an analytical model for determining the relationship between stress and damage threshold based on the thermal transfer equation. The temperature distribution (T) under laser irradiation (I)based on the thermal transfer equation can be described as^[7]

$$\rho c \frac{\partial T}{\partial t} = \nabla (k \nabla T) + \alpha (hv) \frac{\mathrm{d}I}{\mathrm{d}t} e^{-\alpha (hv)x}, \qquad (1)$$

where k, ρ , c, and $\alpha(hv)$ are the thermal conductivity, density, thermal capacity, and absorption coefficient, respectively. If the initial damage source is defect, which has a strong absorption coefficient, then $\alpha(hv)$ is the absorption coefficient of such defect; alternatively, it is the absorption coefficient of the thin film. The damage threshold through Eq. (1) is simplified as

$$I_{\rm th} \approx \frac{T_{\rm c}\sqrt{k\rho ct}}{2\alpha(hv)},\tag{2}$$

where $T_{\rm c}$ is the critical temperature and t is the pulse time of the laser. In general, the damage in ns regime is defined as the temperature of the thin film above $T_{\rm c}$. As a result of the thermal stress mechanism, the relationship between $T_{\rm c}$ and the pressure of thin film is given by^[8]

$$P = \frac{\Delta \sigma E T_{\rm c}}{1 - \gamma},\tag{3}$$

where γ and E are the Poisson's ratio and Young's modulus of the material, respectively, and $\Delta \sigma$ is the differential value of the thermal expansion coefficient at the neighboring layers in the film. The critical temperature can then be described as

$$T_{\rm c} = \frac{P(1-\gamma)}{\Delta \sigma E}.$$
 (4)

In Eq. (4), stress P contains two parts, namely, P_c , which is the critical stress of the bulk material, and P_{thin} , which is the extra stress induced by the thin film. In addition, P_c is a constant that is only correlated to the material, whereas P_{thin} is a variable influenced by numerous parameters, such as coating rate, film thickness, and surface roughness of the substrate. However, once the coating materials and technology are chosen, the values of thermal properties, Young's modulus, and Poisson's ratio in the thin film can be considered as constants. Based on Eqs. (2) and (4), the relationship between stress and threshold can be simplified as

$$I_{\rm th} = M(P_{\rm c} - P_{\rm thin}), \tag{5}$$

where M is a constant decided by the features of the coating materials. The effective range of Eq. (5) is that the initial source of the induced damage is the same; otherwise, M is not a constant, and this equation cannot be used.

The theory prediction is compared against the experiments to validate the analytical model. First, the substrate affects the damage threshold through its interaction with the thin film. This situation is not favorable in obtaining the relationship between stress and threshold^[5]. High-reflection (HR), which indicates that only a small amount of laser energy can still reach the substrate, is the best choice to avoid this effect.

In this letter, three structures of HR films were prepared using a Leybold coater. The substrates were polished BK7 ($\Phi 50 \times 5$ (mm)). The coating materials were mainly fluorides, which was also used as a easily induced the change of stress in the HR film. SiO₂ coating material was used to adjust the stress of the thin film; it had no effect on the reflectivity of the optical spectrum or the electromagnetic standing wave field. The coating stacks of the three structures were S/6L V(NV)¹⁰/A, S/V(NV)¹⁵/A, and S/6L V(NV)¹⁵/A, where L, V, and N denote SiO₂, LaF₃, and AlF₃, respectively, and with one-quarter wavelength optical thickness (QWOT) at 355 nm. In addition, S and A denote the substrate and air, respectively. All three structures (Table 1) were manufactured through the same coating process. The details of the coating process are discussed in our previous work^[9]. The stress of films can be obtained using the curvature method. The total stress can be calculated from Stoney's equation^[10]

$$P_{\rm thin} = \frac{4E_{\rm s}}{3(1-\gamma_{\rm s})} \frac{t_{\rm s}^2}{D_{\rm s}^2 t_{\rm f}} \Delta \text{Power}, \tag{6}$$

Table 1. Threshold and Stress of HR Films

Symbol	Structure	LIDT (J/cm^2)	Stress (MPa)
А	$\mathrm{S}/\mathrm{6L}~\mathrm{V(NV)^{10}/A}$	20.1	15
В	$\mathrm{S/V(NV)^{15}/A}$	8.6	78
С	$\rm S/6L~V(NV)^{15}/A$	16.7	30

Notes: V: LaF₃(n=1.71); N: AlF₃(n=1.41); L: SiO₂(n=1.54)

where $E_{\rm s}$ and $\gamma_{\rm s}$ refer to Young's modulus and Poisson's ratio of substrate, respectively; $t_{\rm s}$ and $t_{\rm f}$ denote the thicknesses of the substrate and thin film, respectively; $D_{\rm s}$ is the radius of the substrate, respectively. The value of Δ Power was measured by an interferometer.

The damage threshold was measured by a triple Nd:YAG laser system (355 nm, 8 ns)^[11]. The LIDT test was implemented based on ISO11254-1 standards under the "1-on-1" mode, with 20 sample sites tested using a fixed fluence step. Each sample had 200 total sites. The repetitive frequency of the laser was 5 Hz, and the laser was focused on the sample surface. In this letter, damage was defined as any irreversible modification detected by an online CCD camera. The relative LIDT determination error was approximately $\pm 10\%$. Meanwhile, damage threshold was defined as the laser fluence as the zero damage probability beginning increased.

The damage morphology was observed using a SEM. Figure 1 shows the typical damaged features of the three structures. The depth of the damage zone indicates



Fig. 1. Damage morphologies of the three structures: (a) S/6L V(NV)^{10}/A; (b) S/V(NV)^{15}/A; (c) S/6L V(NV)^{15}/A.



Fig. 2. Damage threshold as a linear function of stress.

that the damage mainly occurs in the outside layers. The outside layers are LaF₃ and AlF₃ in all the three structures. The experimental results agree with our experimental purpose, which is to obtain the laser-induced damage of the thin film without the substrate effect. The initial source of damage can be considered the same based on the damage morphology (Fig. 1) and the coating conditions. All results, which contain the damage threshold and stress, are shown in Table 1 and Fig. 2, respectively. A linear dependency of the threshold with the stress is observed from Eq. (5), which is the line in Fig. 2, where M is 0.1792 and P_c is 125.5 MPa. The result concurs with our theory analysis.

In conclusion, we develop an analytical model to predict the relationship between stress and damage threshold. Our model can capture the threshold trend as a function of stress. Moreover, our model proves that we can improve the threshold by adjusting the stress in the film. Finally, this model only works when the initial damage induced by the source of the thin films is the same.

The authors would like to express their gratitude to Prof. Zhengxiu Fan for the helpful discussions. This work was supported by the National Natural Science Foundation of China under Grant No. 11104293.

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