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## Dynamics of femtosecond laser pulse induced damage in multilayers

Haiyi Sun <sup>a,\*</sup>, Tianqing Jia <sup>a</sup>, Zhongchao Wei <sup>b</sup>, Jianrong Qiu <sup>a,c</sup>, Xiaoxi Li <sup>a</sup>, Chengbin Li <sup>a</sup>, Shizhen Xu <sup>a</sup>, Donghai Feng <sup>a</sup>, Hezhou Wang <sup>b</sup>, Zhizhan Xu <sup>a</sup>

<sup>a</sup> State Key Laboratory of High Field Laser Physics, Shanghai Institute of Optics and Fine Mechanics,
 Chinese Academy of Sciences, P.O. Box 800-211, Shanghai 201800, PR China
 <sup>b</sup> State Key Laboratory of Ultrafast Laser Spectroscopy, Zhongshan University, Guangzhou 510275, PR China
 <sup>c</sup> Photon Craft Project, Shanghai Institute of Optics and Fine Mechanics, Chinese Academy of Sciences,
 and Japan Science and Technology Corporation, Shanghai 201800, PR China

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#### **Abstract**

We report the single-shot damage thresholds of  $MgF_2/ZnS$  omnidirectional reflector for laser pulse durations from 50 fs to 900 fs. A coupled dynamic model is applied to study the damage mechanisms, in which we consider not only the electronic excitation of the material, but also the influence of this excitation-induced changes in the complex refractive index of material on the laser pulse itself. The results indicate that this feedback effect plays a very important role during the damage of material. Based on this model, we calculate the threshold fluences and the time-resolved excitation process of the multiplayer. The theoretical calculations agree well with our experimental results.

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#### 1. Introduction

Laser-induced damage in optical materials is always a limiting factor in the development of high-power laser systems. Since the invention of laser, laser-induced damage has been extensively studied [1–4], such as damage morphology, threshold fluence and mechanisms, etc. Recent work has concentrated on the damage of bulk dielectric materials with femtosecond pulses, and only a few studied on the multilayers [5].

Dielectric damage in the femtosecond regime can be described as three major processes: (i) the excitation of conduction-band electrons (CBE); (ii) deposition of laser energy in the CBE gas, namely, the heating of CBE; (iii)

transferring the CBE kinetic energy to the lattice [6,7]. A lot of studies have investigated the first process, but only considered the electronic excitation of material by laser and ignored the counteraction of this excitation to the laser pulse itself [8–11]. In fact, this is an interactional process. During the irradiating of laser, the refractive index and extinction coefficient of materials change with the CBE number density, which leads to the changes of reflectivity, transmissivity and distribution of laser intensity in materials. Many experiments about dielectric materials and semiconductors have testified these changes using the pump-probe technique [12–17]. Our recent experiments on another optic films (SiO<sub>2</sub>/TiO<sub>2</sub>) also observed the related phenomenon [18]. The change of reflectivity is related to the material and pump laser intensity, which mainly results from the CBE excitation, that is, plasma formation. Generally, high pump laser intensity leads to the great change of reflectivity. Inversely, this change may further

<sup>\*</sup> Corresponding author. Fax: +086 69918021.

E-mail address: shy780327@siom.ac.cn (H. Sun).

affect the excitation of electrons to conduction band. However, no detailed theoretical investigation was reported on this feedback effect.

In this paper, we measured the dependence of damage thresholds on the pulse durations for MgF<sub>2</sub>/ZnS omnidirectional reflector. A coupled dynamic model was used to investigate the damage process. We explored the influence of the single-pulse laser-induced changes in complex refractive index of material on the laser pulse itself. The results show this feedback effect plays a very important role during the damage of material. Using this model, we calculated the threshold fluences and the ultrafast excitation process of the multilayer. The theoretical results were in good agreement to the experimental measurements.

#### 2. Experiments and results

Experiments were carried out using a Ti: sapphire laser ( $\lambda=800$  nm) with 50 fs full width at half maximum pulse duration. The highest output energy is 0.6 mJ. With a half wave plate and a polarizer, we could vary the pulse energy continuously. The pump pulse was focused on the front surface of material with a biconvex lens with 150 mm focal length. The diameter of focused spot was measured to be around 30  $\mu$ m. We utilized dispersive materials (ZF6 glasses) to adjust the pulse duration  $\tau_p$  from 50 fs to 900 fs.

MgF<sub>2</sub>/ZnS omnidirectional reflector was studied in the experiment, and its reflectivity is more than 99% for incident angles of 0–75° at the wavelength of 800 nm. It is deposited by heat evaporation technique on silica substrate. Its construction is S(HL)<sup>11</sup>(HL)<sup>13</sup>, where S indicates substrate, (HL) are made from high and low refractive materials (ZnS/MgF<sub>2</sub>). Their refractive indexes are 2.38 and 1.35, respectively. The optical thickness of each layer is a quarter-wave, and the peak wavelength of the 11 and 13 pairs of layers is 880 and 1040 nm, respectively. The

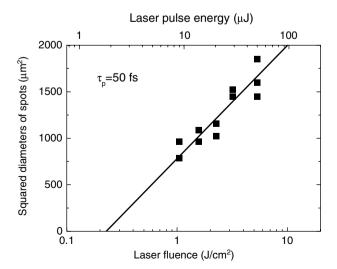


Fig. 1. Squared diameters of the ablation spots as a function of laser pulse energy and laser fluence.

sample was set on a three-dimensional precision stage, and damaged at normal angle. Each location on the sample was irradiated by only one laser pulse.

Fig. 1 shows the process of calculating the threshold fluences, and the method used in our work is adopted from Refs. [12,19] and [20]. This method determines both the threshold energy  $E_{\rm th}$  and the  $e^{-2}$ -beam radius  $\omega_0$ , and thus the damage threshold  $F_{\rm th} = \frac{E_{\rm th}}{\pi\omega_0^2}$ . The empty circles in Fig. 2 indicate the threshold fluences of omnidirectional reflector irradiated by 800 nm laser, which are about  $0.30~{\rm J/cm^2}$ .

#### 3. Theory

In the bulk dielectric materials, it is proposed the damage is induced by electron avalanche. About the damage threshold, this theory has successfully explained various experimental results [21–23]. A recent report about Ta<sub>2</sub>O<sub>5</sub>/SiO<sub>2</sub> multilayer also utilized electron avalanche model to explain the dependence of the threshold fluences on the pulse widths, and their theoretical results agree well with the experimental measurements [5]. Additionally, we find that the damage thresholds (Fig. 2) don't increase so fast as those expected from the photo-ionization alone. Considering all these results, we calculate the threshold fluences of the reflector based on electron avalanche model. In this model, the feedback effects of the excitation of CBE on the laser are also calculated.

Fig. 3 indicates the flow chart of this feedback model. During the calculation, each layer of the sample is divided into 10 thin layers. At a time t, the laser intensity distribution, CBE density and dielectric constant at a specific location in each thin layer are first calculated. Then adding a temporal step  $\Delta t$ , the above process is cycled using the results obtained at the previous time as the initial condition. The temporal step  $\Delta t$  is 2 fs.

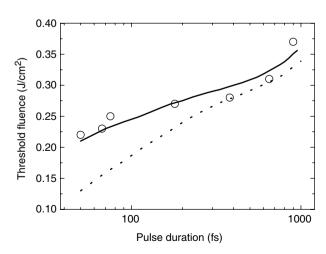


Fig. 2. Threshold fluences vs. pulse durations. The empty circles represent our experimental results with error of  $\pm 20\%$ . The solid and dotted lines are theoretical results simulated by the models with and without the feedback effects, respectively.

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