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Transition from isolated submicrometer pits to integral ablation of HfO_2 and SiO_2 films under subpicosecond irradiation

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ABSTRACT

Damage behavior of HfO_2 and SiO_2 films under subpicosecond irradiation is investigated experimentally and theoretically in this work. The typical damage phenomenon is the transition from isolated submicrometer pits to integral ablation at transitive threshold. The experimental damage thresholds for both coatings are consistent with the theoretical calculation. The rate equation considering the feedback effect of electron number density is applied to calculate the deposited energy density, which illustrates the evolution of damage morphology.

1. Introduction

Recently the rapid development of ultrafast pulse technology has accelerated many practical applications such as highly precise micromachining [1,2], study of ultrafast processes [3,4] and high power laser systems. Such technologies require functional dielectric films to control the light temporally and spatially [5]. However, laser-induced damage of thin films seriously limits the peak power and beam quality.

Dielectric breakdown in the subpicosecond regime has been understood well in the early studies and explained by the nonlinear processes such as multiphoton excitation and impact ionization, which defines a highly deterministic phenomenon depending on the intrinsic material properties [5–7]. Dielectric coatings based on oxides are commonly used in both the practical laser systems and damage experiments. Abundant investigations indicate that the damage phenomenon of thin films under subpicosecond laser is similar with that of bulk materials [8]. The effects of the pulse duration, wavelength, optical band gap energy, material composition and deposition technique on the damage resistance have been explored for a variety of pure and mixed oxide coatings [9–14]. In addition, the rate equation (RE) including the Keldysh thoery for photoionization, Drude model for avalanche and electron relaxation from conduction band to the lower electronic states [8] is used to illustrate the intrinsic fundamental mechanism. Further electric field correction is suggested to include the optical interference effects of the thin films. When the electrons in conduction band surpass the critical density ($\sim 10^{21}\,\mathrm{cm}^{-3}$), the dielectric properties are strongly dependent on the electron density and then lead to the redistribution of electric field, which is known as the feedback effect [5]. Therefore a relatively well-developed RE model including the feedback effect is successfully developed to explain the threshold behavior of the thin films [5,15].

Although the threshold behavior has been well studied by abundant researchers, damage behavior related with the energy deposition process and morphology evolution is only concerned recently [12,15]. Damage features of the HfO_2, SiO_2 and their mixtures under the subpicosecond irradiation indicate the morphology transition from the high density submicrometer dots at the lower fluence to spallation at the higher fluence [12,15]. More studies show a scattered spalling feature with the indistinct and ambiguous boundary for the damage site under higher fluence in the subpicosecond regime, which is distinctly different from the legible and clear cut boundary usually observed under the pulse less than 100 fs [16]. Although several preliminary discussion have been made, a more detailed investigation about the finer damage behavior and morphology evolution is required to illustrate the fundamental damage mechanism. In this work, transition phenomenon is experimentally and theoretically studied to reveal the fundamental mechanism affecting the morphology transition in the subpicosecond, which is beneficial for applications like laser machining and developing optical coatings for high power laser system.

2. Experiments

Hafnia and silica single layers are deposited on fused silicas (Corning 7980) by electron beam evaporation technology. Detailed deposition parameters were given in our previous work [17]. The substrates are ultrasonic cleaned prior to coating. The physical thick-

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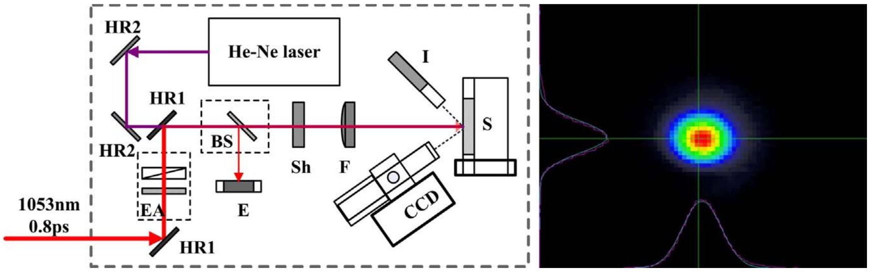

Fig. 1. (Left) Experimental setup for laser damage test at 0.8 ps, 1053 nm. HR1, high-reflective mirror for 1053 nm; HR2, high-reflective mirror for He-Ne laser; EA, energy attenuator (half-wave plate and polarizer); E, energy meter; BS, beam splitter; Sh, mechanical shutter; F, focusing lens; I, white light illumination system; S, thin film samples. (Right) Image of beam intensity distribution at the sample location and the e^{-2} intensity diameter of laser spot is about 331 μm.

ness of the high-refractive index (1.98) material (HfO_2) and low-refractive index (1.47) material (SiO_2) is about 545 nm and 570 nm respectively.

The damage test configuration is shown schematically in Fig. 1. A 10 Hz, 1053 nm laser with pulse duration of 0.8 ps is used to normally focus on the surface of the coatings with a quartz lens. The damage test is conducted in the “1-on-1” mode and a mechanical shutter is used to extract the single pulse. There are 15 sites to be irradiated for each fluence stage. A He-Ne laser is used to collimate optical path. The laser spot close to the target surface is measured with a CCD camera. The e^{-2} intensity diameter of laser spot is about 331 μm and the fluence used in this work is average fluence deriving from the ratio between recorded energy and laser spot. A white light illumination system is used to illuminate the sample and another online CCD image system is used to detect the variation of light scattering from the laser irradiation region. The method combining the online CCD image and offline Nomarski microscope with a magnification of 100 is applied to define the damage event. Due to the indistinct and ambiguous boundary for the damage site, the widely accepted quadratic fitting method [18] in the sub-picosecond regime for the damage diameter and fluence is not applied. We employ the definition of the laser-induced damage threshold (LIDT) suggested in the Refs. [12,19], which is the mean value between the highest fluence of zero damage probability and the upper fluence. The horizontal error bar is linked with the fluence fluctuation during the practical energy measurement for each fluence stage.

The surface morphology of the damage site is characterized by the Nomarski optical microscope, field emission scanning electron microscope (FE-SEM, Zeiss Auriga S40). The cross section of the damage site is checked with the focused ion beam (FIB) module that equipped inside the SEM.

3. Results

3.1. Damage threshold

The statical results of the damage test for HfO_2 and SiO_2 thin films are shown in Fig. 2. The LIDTs for HfO_2 and SiO_2 coatings are 1.18 J/cm^2 and 2.14 J/cm^2. The fluence interval between 0% and 100% probability is small when compared with the energy fluctuation, which depicts a deterministic behavior with respect to the intrinsic properties of material or very large defect density. However, there are several fluence level between 0% and 100% probability, which indicates certain stochastic factor about the damage initiation in addition to the deterministic behavior. Generally stochastic factor during the damage process is related with the defects like the nanosecond case. More discussion is needed to illustrate the stochastic factor as well as the way to affect the LIDT.

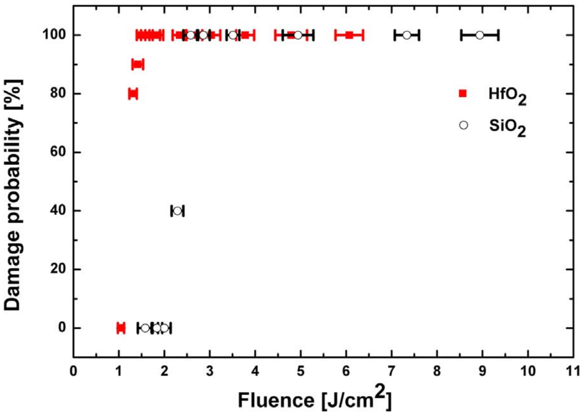

Fig. 2. Damage probability *versus* fluence for HfO_2 and SiO_2. The error bar is corresponding to the practical measured energy fluctuation.

3.2. Damage features of HfO_2

The optical morphologies of HfO_2 film depending on the incident fluence are shown in Fig. 3. At the lower fluence, the damage site is comprised of high density isolated submicrometer pits related with high density damage initiators. However, defect-induced damage is generally responsible for the material irradiated by the nanosecond [20–25]. When the incident fluence surpasses 2.3 J/cm^2, the high density isolated submicrometer pits joint together gradually and form a large-scale surface ablation like an integral. High resolution SEM images in Fig. 4 depict that the isolated pits are all in the submicrometer scale unless they joint together. Moreover, FIB cross section on the surface of the high density isolated pits indicates that the damage pit is shallow and close to the coating surface.

A transitive threshold (~2.3 J/cm^2 here for HfO_2) is defined to label the fluence required to transfer the isolated submicrometer pits to integral ablation. Fig. 5(a)–(c) depicts SEM morphologies on the whole, edge and bottom of the damage site just above the transitive threshold. We clearly observed the joint process of the isolated submicrometer pits near the edge of damage site in Fig. 5(b). The bottom of damage pit is very rough and corresponding to the joint process of high density isolated submicrometer pits. Similar features are also observed at the higher fluence, as shown in Fig. 5(d)–(f).

3.3. Damage features of SiO_2

The optical morphologies of SiO_2 film depending on the incident fluence are shown in Fig. 6 and damage phenomenon similar with that of HfO_2 film is observed. The damage morphologies are not symmetric since the laser spot is nearly elliptical, as shown in Fig. 1. Nevertheless, the flaw of the laser spot does not affect the major phenomenon investigated in this work. High density isolated submicrometer pits at the lower fluence and integral ablation by the joint process at higher fluence are observed. Finer features for both the isolated pits and integral ablation are shown in Fig. 7. Isolated submicrometer pits are found at 2.6 J/cm^2 (Fig. 7(a)) and slightly joint process of the isolated pits are observed at fluence 2.9 J/cm^2 (Fig. 7(b)). FIB cross section in Fig. 7(c) indicates that the damage pit is shallow and close to the