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Femtosecond laser-induced damage in reflector

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Abstract

We have investigated the damage for ZrO_2/SiO_2 800 nm 45° high-reflection mirror with femtosecond pulses. The damage morphologies and the evolution of ablation crater depths with laser fluences are dramatically different from that with pulse longer than a few tens of picoseconds. The ablation in multilayers occurs layer by layer, and not continuously as in the case of bulk single crystalline or amorphous materials. The weak point in damage is the interface between two layers. We also report its single-short damage thresholds for pulse durations ranging from 50 to 900 fs, which departs from the diffusion-dominated $\tau_P^{1/2}$ scaling. A developed avalanche model, including the production of conduction band electrons (CBE) and laser energy deposition, is applied to study the damage mechanisms. The theoretical results agree well with our measurements.

Keywords: 45° high-reflection mirror; Femtosecond laser; Damage threshold; Ablation depth

1. Introduction

Ultra-short and high-power laser technologies are progressing rapidly in recent years. The pulse width τ_p has extended down to 6 fs [1], and the peak power up to TW, even to PW level [2]. Many applications, ranging from materials processing to nuclear fusion and plasma physics, can potentially benefit from the intense short pulse. On the other hand, lots of optical components used in these laser systems are often damaged, especially for filters and reflectors. Therefore, it is meaningful to study the damage mechanisms of reflector.

The study of laser-induced damage of thin films has been an interesting subject since 1970 s. The damage caused by nanosecond pulses is explained on the basis of an absorption-inclusion model, which involves an impurity within the film absorbs the laser irradiation and the absorbed energy subsequently diffuses through the surrounding film [3]. The model predicts a $\sqrt{\tau_P}$ dependence of the threshold fluences on pulse duration. Because the damage occurs through heat

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deposition (heating of lattice), the damage morphologies have general characters. At smaller pulse fluences, the irradiated area is molten and covered with cracks resulting from mechanical tensile stress generated by thermal expansion and cooling process. At larger fluences, the area is evidenced by intense boiling to evaporating [4–8]. In the femtosecond regime, the ablation morphologies and process are also closely related to the laser intensity. This has been verified by many studies about the thermal and ultrafast melting in semiconductors and metals [9–11]. Their results indicate that the threshold of ultrafast melting is larger than that of thermal melting. The morphological features, such as melting, ablation and recrystallization, depend strongly on the laser intensity.

Recent work has concentrated on the damage of bulk dielectric materials with femtosecond laser pulses. Damage morphology, depth and threshold fluence have been studied extensively. Firstly, the absorbed energy has been deposited in conduction band electrons (CBE) gas before it is transferred to the lattice. Hence, the ablated crater has no evidence of heat diffuse, and is characterized by the removal of material, followed by sharp edges [12–14]. Secondly, the evolution of ablation crater depth with laser fluences depends on the pulse duration. The study about α-Al_2O_3 [15] showed that below 1 ps there was a clear threshold, above which the depth suddenly rose to about 100–200 nm. But for long pulses, the threshold was not so sharp, the depth increased more slowly and the holes were more shallow, typically 20 nm. Besides, the similar phenomena have also been found in SiO_2 and CaF_2 [16]. Lastly, as for the measurements of damage threshold, many experiments have yielded significant deviation from the $\sqrt{\tau_p}$ scaling for femtosecond pulses [5,17–19].

There are two opinions about the dielectric damage in the femtosecond regime. One think optical breakdown can be understood in terms of electron avalanche. In the avalanche model [17,18,20], the conduction band electrons are produced by photoionization and impact ionization. Local intensity enhancements due to interference effects were also taken into account in a recent report about multilayer [21]. In these studies, the critical plasma density of $n_{cr} = 10^{21}$ cm^{-3} was often used as the damage criterion. The other opinion thinks photoionization alone can cause the damage of materials, and the impact ionization does not play a dominant role during the damage process [22–24]. With the development of laser and thin films technology, the damage of multilayer is expected to be studied more thoroughly.

In this paper, we show the damage morphologies and the dependence of ablation depths on the pulse fluences for ZrO_2/SiO_2 45° high-reflection mirror. The damage thresholds as a function of pulse duration ranging from 50 to 900 fs are also presented. A developed avalanche model, including the production of CBE and energy deposition, was used to study the damage mechanisms. We calculated the damage thresholds of the mirror taking the critical energy deposited in CBE as the damage criterion, and the theoretical calculations agreed well with the experimental results.

2. Experiment

The output of 800-nm Ti: sapphire laser system used in our experiments was 50 fs, 0.6 mJ. By using a half wave plate and a polarizer, we could vary the pulse energy continuously. The pulse energy was measured with an energy meter from a split-off portion of the beam. We utilized dispersive materials (ZF_6 glasses) to adjust the pulse duration from 50 to 900 fs. The sample is set on a three-dimensional translation stage, and the pulse is focused on the front surface to a diameter of ~30 μm. Each location on the sample was irradiated by only one laser pulse. We monitored the sample surface with a microscope objective (NA ~0.1) and a charge-coupled device (CCD). For the constant pulse duration, the pulse energy was decreased until the damage spot could not be observed through the CCD. After irradiation, the areas of spots were measured with the higher-resolution optical microscopy (NA~0.65). Through the relation between the areas of spots and laser fluences, we could calculate the damage threshold for every pulse width.

ZrO_2/SiO_2 45° high-reflection mirror was used in the experiment. Its construction is $G(H_1L_1)^2(2H_1\,2L_1)^{15}2H_1$, where G indicates fused silica substrate. H_1 stands for ZrO_2 with one quarter wavelength optical depth (QWOD), and L_1 stands for SiO_2 with one QWOD. The peak wavelength of both surface layer and 15 pairs of layers is 428 nm, and 800 nm for

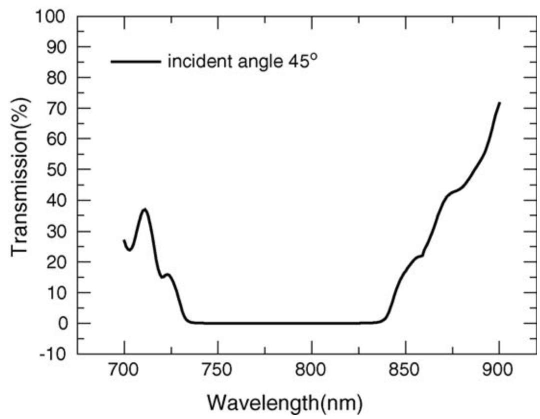

Fig. 1. Transmission of 45° high-reflection mirror.

the 2 pairs of layers. The sample is deposited by electron-beam evaporation. The transmission is showed in Fig. 1. The experimental measurements indicated that its reflectivity is 98% in the 45° incident angle at the wavelength of 800 nm. So the mirror has low optical losses and high quality.

3. Results

Fig. 2(a) and (b) show the ablation morphology of the mirror and corresponding depth profile. Here the laser condition is 50 fs, single shot at 800 nm. The sample was damaged at 0°. With insufficient time for energy coupling to the lattice, the damage crater is no obvious sign of heat diffusion. Although larger laser fluence, ~3 times the damage threshold is applied, there is no evident thermal melting or boiling around the spot as is presented in the long pulse regime. Clear edge is easily seen in Fig. 2(a). This indicates that thermal diffusion does not play an important part in the damage of mirror. From Fig. 2(b), we can find that the maximum depth of the ablation crater reaches more than 1 μm. There exist some levels in the crater. We observed the similar behavior in the ablation morphologies of SiO_2/TiO_2 omnidirectional reflector [25], which is probably related to the removal of one entire layer, and shows the breakdown in multilayers may occur layer by layer. This is different from what is observed for bulk single crystalline or amorphous samples [12,15,20]. The bottom of the crater is not very flat and smooth, which may be due to some inhomogeneities in the intensity distribution of the laser in the focal spot, and amplified by threshold effects.

Fig. 3 shows the ablation depths of the mirror. Solid squares are the average ablation depths, and the line is fitted results. The depths were measured by atomic force microscope (AFM). From Fig. 3, we can find that the ablation depths are linearly proportional to the logarithm of pulse fluences. The linear character indicates that all vaporized material has not redeposited in the cavity but flown out of it. Furthermore, the damage depths suddenly reach hundreds of nanometers once the laser fluences are above the thresholds. Although the ablation depths of multilayers induced by femtosecond pulses have not been investigated yet, many bulk dielectrics, such as fused silica, CaF_2 and α-Al_2O_3 [15,16], show similar phenomena. On the other hand, the ablation depths in multiplayer are quite large compared to the results for bulk dielectric materials, which show ablation

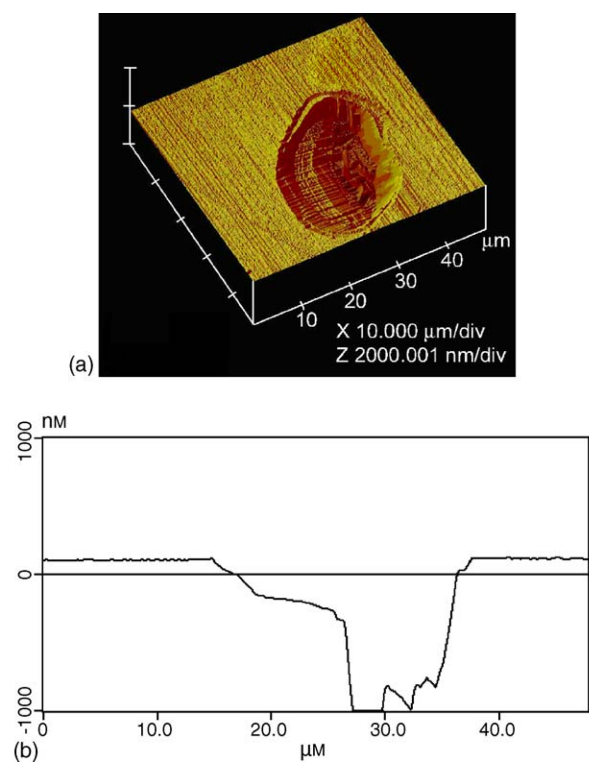

Fig. 2. Ablation pit (a) and its depth profile (b) in 45° high-reflection mirror after a single shot ablation with a laser pulse of 50 fs and a fluence of 1.08 J/cm^2.

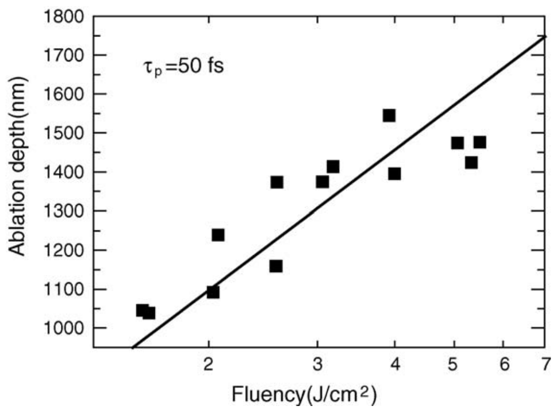

Fig. 3. Ablation depth vs laser fluence. The solid squares and line are our measurement and best-fit results, respectively.

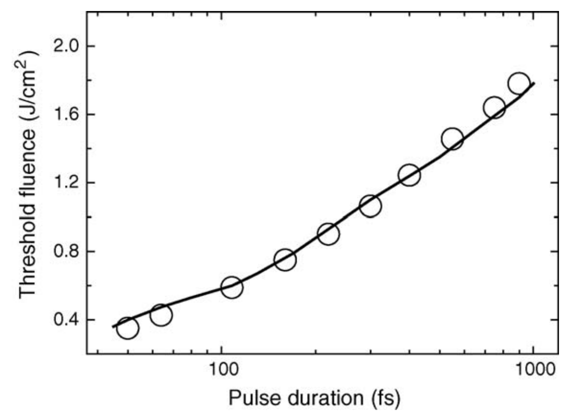

Fig. 5. Threshold fluences vs pulse widths. The hole circles represent our measurements, and the solid curve is our theoretical result. The measurement error is less than ±15%.

depth is below 500 nm, even at intensities corresponding to several times the single shot threshold [15]. We also find the ablation depths (combined Fig. 3 and Fig. 6) locate in the vicinities of the interface between two layers. The similar phenomenon was also observed in SiO_2/TiO_2 omnidirectional reflector [25]. Besides, in the damage of SiO_2/TiO_2 omnidirectional reflector, we also find that only the surface layer is ablated when the laser fluences are slightly lager than the ablation threshold. These are indications that the weak point is the interface between two layers, and an integer number of layers are removed in multiplayer damage. Additionally, the ablation depths in multiplayer are larger than that produced by pulses longer than a few tens of picoseconds because of the reduction of heat diffusion effects [6,15].

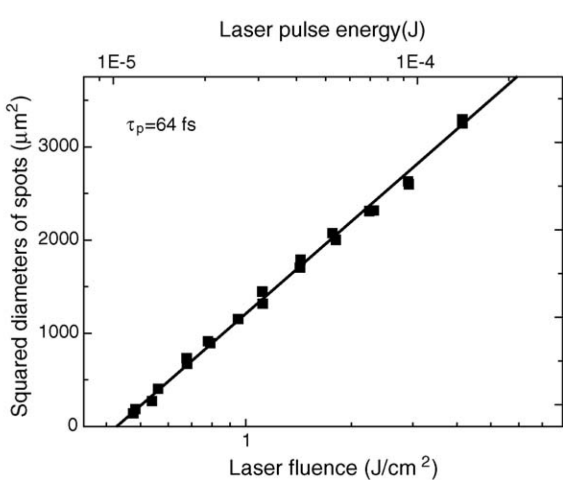

Fig. 4. Squared diameters of the ablation spots as a function of laser fluence.

Fig. 4 shows the process to measure the threshold fluence [26,27]. The damage thresholds as a function of pulse duration are illustrated in Fig. 5. The threshold fluence represents the average laser fluence, which is one half of the peak fluence. The mirror was damaged at its 45° angle.

In Fig. 5, we can find that the damage threshold of the mirror increases slowly from 0.35 to 1.8 J/cm^2 as pulse width ranging from 50 to 900 fs, and it can not be fitted well by $\tau_p^{1/2}$ scaling. This further indicates that the electron kinetic energy does not transfer to the lattice and diffuse during the laser pulse [5].

4. Theory

The results presented above exhibit similar features to that in bulk dielectric materials. Firstly, the ablation morphology is no evident sign of heat diffusion and thermal melting. Secondly, the ablation depths are linearly proportional to the logarithm of pulse fluences and there is a clear threshold, above which the depth suddenly increase to hundreds of nanometers. Thirdly, the damage threshold does not scale as $\tau_P^{1/2}$. In the bulk dielectric materials, it is concluded the damage is induced by electron avalanche. About the damage threshold, this theory has successfully explained different results [5,13,14,16]. A recent report about Ta_2O_5/SiO_2 multilayer also utilized electron avalanche

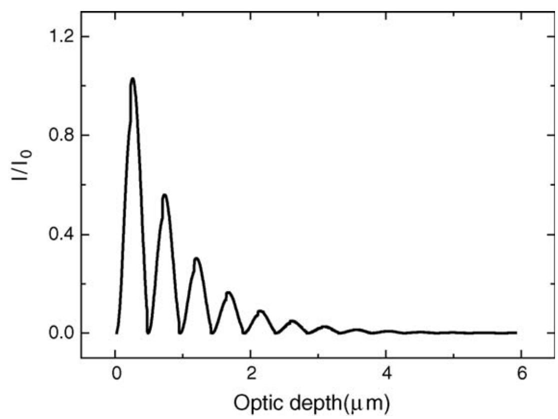

Fig. 6. The distribution of laser intensity for 45° high-reflection mirror.

to model the dependence of the threshold fluences on the pulse widths, and their theoretical results agree well with the experimental measurements [21]. Additionally, we find that the damage thresholds (Fig. 5) do not increase so fast as those expected from the photoionization alone. Considering all of this, we calculate the threshold fluences of the reflector based on electron avalanche model. In this model, laser energy deposition is also calculated taking into account the unique features in multilayer breakdown.

The process of electron avalanche can be understood as follows. CBE oscillate in response to the laser field, transferring energy by scattering from phonons. Once an electron can achieve energy equal to the band gap, the subsequent impact ionization promotes another valence electron up to conduction band [5,18]. The generation of CBE is controlled by the local pulse intensity I. Fig. 6 shows the field distribution in 45° high-reflection mirror. I_0 stands for the incident laser intensity in air. The field distribution indicates that the mirror is most likely damaged at the first interface between ZrO_2 and SiO_2. $ZrO_2(SiO_2)$ has a band gap of ~3.5 eV (9 eV). With our excitation wavelength (~1.55 eV), ZrO_2 need absorb less photons to excite than that of SiO_2. Thus damage is likely to occur first in the high-index ZrO_2 layer [21].

On the basis of avalanche model, the evolution equation of CBE density $n_e(t)$ and energy density E_{dep} deposited in CBE gas can be written as [16]:

$$\frac{\partial n_e}{\partial t} = (R_{PI} + R_{II} n_e)\left(1 - \frac{n_e}{N_0}\right), \tag{1}$$

$$E_{dep} = \int n_e(t) A \mathrm{d}t, \tag{2}$$

where the photoionization rate R_{PI} can be calculated by Keldysh theory [28]. The factor $1 - n_e/N_0$ is introduced for the consideration of the exhaust of valence band electrons (VBE). We consider that only one VBE in a molecule is excited to the conduction band, hence the initial number of VBE (N_0) is equal to that of molecule in the material [29]. The initial densities N_0 of ZrO_2 and SiO_2 are 3.2×10^{22} and 2.2×10^{22} cm^{-3}, respectively. The impact ionization rates R_{II} are calculated by using a flux-doubling model [30,31]. The distribution of laser intensity with the thickness of the reflector is calculated with the theories in matrix optics. We calculate the total absorption rates A by means of the quantum mechanical method [14,31]. When the laser intensity is much larger than 10 TW/cm^2, perturbation theory will fail, and it is substituted by classical method [5,29]. During the calculation, the surface layer of the sample is divided into 10 thin layers, and the other layers are not divided.

Fig. 7 indicates the evolution of CBE density in the surface layer. The CBE density represents the average density over the ten thin layers. The electron density produced by photoionization alone is included for reference. Because photoionization is dependent strongly on the band gap of the material, the electron density produced by it reaches a magnitude of 10^{20} cm^{-3} for narrow band-gap ZrO_2 (3.5 eV). But for the material with wide band gap, the CBE density caused by photoionization alone reaches only a

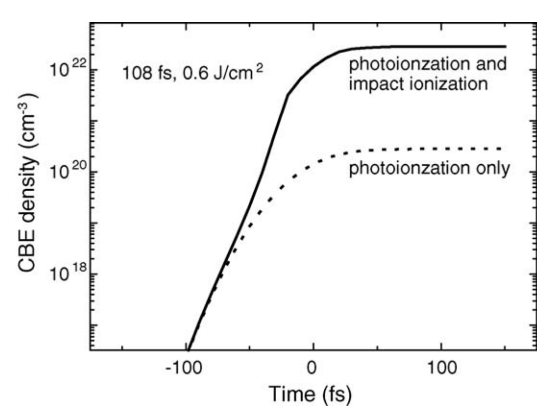

Fig. 7. The evolution of CBE density. The solid curve shows the CBE density produced by photoionization and avalanche ionization, and the dotted curve, only photoionization is considered.

magnitude 10^{18} cm^{-3}, which is generally three orders of magnitude smaller than that produced by photoionization and impact ionization [5]. In our calculations, there is two orders of magnitude difference, which shows that both impact ionization and photoionization play very important roles during the damage. Using CBE density, the corresponding energy density can be obtained by solving Eq. (2).

Fig. 8(a) and (b) show the average energy deposited in the surface layer and the energy density as a function of the depth, respectively. We can find that the energy deposited in the material is inhomogeneous, which is resulted from the inhomogeneous laser intensity distribution (Fig. 6). The energy density increases with increasing laser fluence. Each peak energy density locates in the vicinities of interface between two layers, which again indicates the weak point in damage is the interface. Non-thermal melting in semiconductors and ablation in dielectrics were studied intensively [15,32].

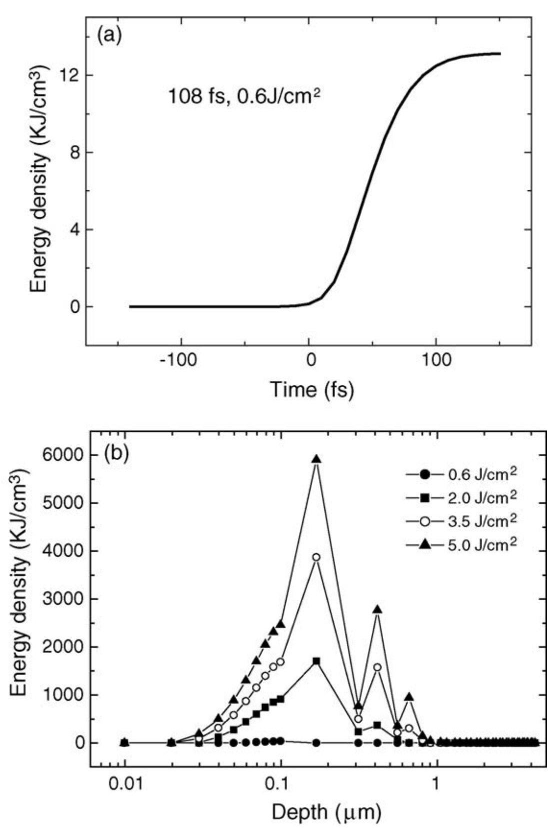

Fig. 8. (a) The evolution of average energy density in the surface layer with time. (b) Energy density as a function of depth.

It was proposed there was an energy barrier to be surmounted before samples were damaged. We think that the damage of mutilayers has similar process. Based on this, the inhomogeneous energy distribution will lead to that the ablation in multilayer occurs layer by layer, and not continuously as in the case of bulk single crystalline or amorphous materials. This is consistent with our observations from the experimental results.

We fit the damage thresholds using 10 kJ/cm^3 as the energy barrier of ZrO_2 film, which is shown in Fig. 5. The results are in good agreement with the experimental measurements. Our threshold fluence increases by ~5 times with increasing pulse duration. For the material with wide band gap, there is a 2–4 times increase in general for the damage induced by femtosecond laser [5,28]. This further indicates that photoionization plays more important part in the material with narrow band gap.

5. Conclusions

The damage of ZrO_2/SiO_2 45° high-reflection mirror induced by femtosecond pulses is investigated. The ablation morphologies and crater depths are considerably different from that induced by pulses longer than a few tens of picoseconds. The damage threshold of the mirror increases from 0.35 to 1.8 J/cm^2 for pulse duration ranging from 50 to 900 fs. A developed avalanche model, including CBE density evolution and laser density deposition, is used to explain the damage mechanisms. When the damage criterion is taken as 10 kJ/cm^3, the theoretical damage thresholds agree well with our measurements. Our results indicate that the ablation in multiplayer occurs layer by layer, and different from that in the case of bulk materials. The weak point in damage is the interface between two layers.

Acknowledgments

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