Femtosecond laser induced damage of optical coatings

Lei Yuan (袁 磊)[1,2], Yuan'an Zhao (赵元安)[1], Hongbo He (贺洪波)[1], and Jianda Shao (邵建达)[1]

[1]*Shanghai Institute of Optics and Fine Mechanics, Chinese Academy of Sciences, Shanghai 201800*

[2]*Graduate School of the Chinese Academy of Sciences, Beijing 100039*

Single pulse laser-induced damage threshold (LIDT) was investigated for electron beam evaporated optical coatings, including ZrO_2 and HfO_2 single layers, ZrO_2/SiO_2 and HfO_2/SiO_2 high-reflective (HR) coatings, using a 50-fs, 800-nm Ti:sapphire laser. The experimental results showed that the damage thresholds of HfO_2 single layer and HfO_2/SiO_2 HR coating were higher than those of ZrO_2 single layer and ZrO_2/SiO_2 coating, respectively. Namely, the wider the band gap was, the higher the LIDT would be. Meanwhile, single layer showed higher LIDT than corresponding HR coating. A theoretical model based on conduction band electrons produced by photoionization and impact ionization was applied to discuss the damage mechanism. According to the model, the damage thresholds were also calculated and accorded with experimental results. In addition, the surface morphologies of the samples after laser irradiation were observed by Leica optical microscopy to get precise evaluations of damage characteristics.

OCIS codes: 310.1620, 320.7090, 140.3330, 260.3230.

Optical coating, such as high-reflective (HR) coating, anti-reflective (AR) coating and filter, is an essential part in the optical system. For example, HR coatings will serve as the fundamental part of multi-layer dielectric pulse compressor grating and ultra-broadband mirrors used in the chirped pulse amplification (CPA) and optical parametric CPA (OPCPA) laser systems. However, laser-induced damage in optical coatings is always a limiting factor in the development of high-power laser systems[1]. The onset of even small damage sites within an optical coating can eventually degrade the beam quality sufficiently to prevent optimum laser operation. Therefore, it is necessary to study the damage of optical coatings. In the past several years, laser-induced damage of optical coatings with nanosecond laser was performed extensively. The damage threshold, damage morphology and damage mechanism were reported in detail[1−3]. While recent work has concentrated on the damage of bulk dielectric materials with femtosecond lasers[4−6].

In this work, single pulse laser-induced damage threshold (LIDT) was investigated for electron beam evaporated optical coatings, including ZrO_2 and HfO_2 single layers, ZrO_2/SiO_2 and HfO_2/SiO_2 high-reflective (HR) coatings, using a 50-fs, 800 nm Ti:sapphire laser. The damage thresholds of all samples were reported. A theoretical model based on conduction band electrons produced by photoionization and impact ionization was applied to discuss the damage mechanism. In addition, the surface morphologies of the samples after laser irradiation were observed by Leica optical microscopy to get precise evaluations of damage characteristics.

ZrO_2 and HfO_2 single layers, ZrO_2/SiO_2 and HfO_2/SiO_2 HR coatings were used. All the samples were prepared in the same coating chamber by conventional e-beam deposition and at the same deposition conditions. Designs of coating stacks for single layers and HR coatings are given by

$$G|4H|Air, \quad (1)$$

$$G|(H3L)(2H2L)^{14}2H|Air, \quad (2)$$

where G indicates BK7 glass substrate ($\Phi 30 \times 3$ mm). L and H stand for low refractive index material (SiO_2) and high index oxide (HfO_2 or ZrO_2), respectively, with quarter wavelength optical thickness (QWOT).

Laser-induced damage test for all samples was performed in the 1-on-1 mode, that is, each location on the sample was irradiated by only one laser pulse. The experimental setup is shown in Fig. 1.

Laser pulses with 50-fs duration, 800-nm center wavelength, and 1-kHz repetition rate were emitted from an amplified Ti:sapphire laser system. A half-wave plate and a polarizer were used to vary the laser energy. The pulse energy was measured by energy meter from a split-off portion of the beam. The surface of the sample was positioned perpendicular to the direction of the incident laser beam in the focal plane of a lens with a focal length of 160 mm. A Gaussian spatial beam profile with a radius ($1/e^2$) of ~ 20 μm in the spot was achieved. The sample surface was monitored in-situ with a charge-coupled device (CCD) and a cold light source.

The surface morphology after laser irradiation is an important part to get precise evaluations of damage characteristics. The damage morphologies of all samples were observed by Leica microscope as shown in Fig. 2.

For the damage morphologies of all samples (Figs. 2(a)−(d)), the damage induced by defects or impurities and thermal melting or boiling around the spot presented in the long pulse regime are not evident. There is clear edge. This indicates that thermal diffusion and defects or impurities do not play an important part in the damage of optical coatings for femtosecond laser. Moreover,

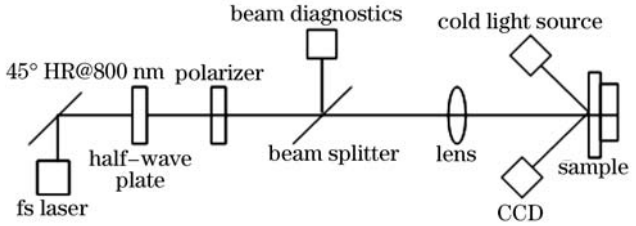

Fig. 1. Experimental setup used for laser damage test.

damage crater is confined to a small region and occurs only over an area with sufficient intensity to produce ionization. Meanwhile, from Figs. 2(b) and (d), it can be seen that the damage morphologies of ZrO_2/SiO_2 and HfO_2/SiO_2 HR coatings are obviously layered. That is, damage in HR coatings may occur layer by layer. This is different from that observed for single layers (Figs. 2(a) and (c)).

The damage morphology was used not only for precise view of laser damage but also for determining the LIDT. The areas of spots damaged were measured with an optical microscopy. The relation between spot area and laser fluence is given by[6]

$$D^2 = 2\omega_0^2 \ln\left(\frac{\phi_0}{\phi_{\rm th}}\right), \tag{3}$$

where ϕ_0 denotes the maximum laser fluence, $\phi_{\rm th}$ is threshold fluence, D is the diameter of a damage crater.

Then it is possible to determine the threshold fluence from a plot of the square of the crater diameter versus the logarithm of the laser fluence. The damage threshold fluence is calculated (Eq. (3)) from linear extrapolation ($D^2 \to 0$) of the data points depicted in the plot. The relative error of the LIDT-determination amounts to $\pm 15\%$, which is mainly due to the uncertainty in the spot size measurements. Then the damage thresholds of all samples were calculated by this method and are shown in Fig. 3.

It can be seen that the thresholds of HfO_2 single layer and HfO_2/SiO_2 HR coating were higher than those of ZrO_2 single layer, ZrO_2/SiO_2 HR coating, respectively. Meanwhile, HfO_2 single layer has higher LIDT than HfO_2/SiO_2 HR coating does, and ZrO_2 single layer has higher LIDT than ZrO_2 /SiO_2 HR coating does. That is, the LIDT of single layer is higher than that of the corresponding HR coating.

In general, the damage of bulk material in femtosecond regime is intrinsic[4−7]. It can be explained by the nonlinear excitation associated with the high electromagnetic field of electrons to the conduction band via processes such as impact ionization, tunneling ionization, and multiphoton absorption[8]. When the conduction-band

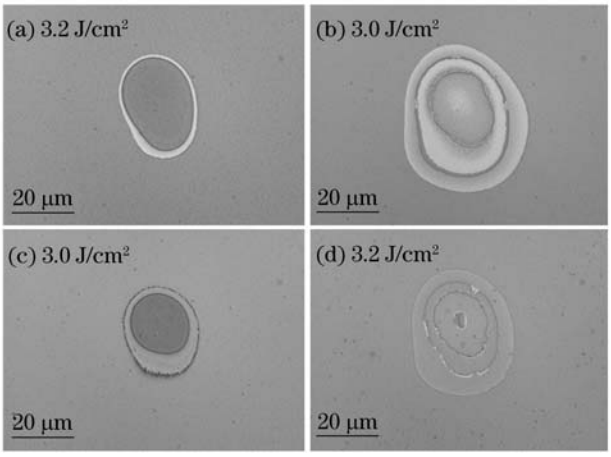

Fig. 2. Damage morphologies of all samples. (a) ZrO_2 single layer; (b) ZrO_2/SiO_2 HR coating; (c) HfO_2 single layer; (d) HfO_2/SiO_2 HR coating.

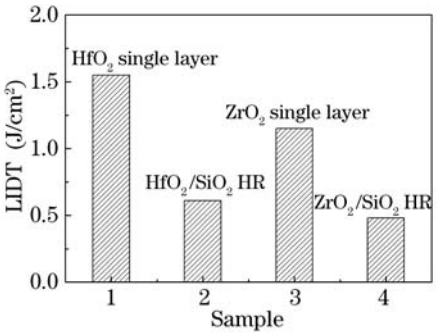

Fig. 3. Experimental damage thresholds of all samples.

electron density reaches a critical plasma density (10^{21} $\rm cm^{-3}$, for $\lambda \sim 800$ nm), the material absorbs strongly through the process of inverse bremsstrahlung resulting in ablation and permanent structural changes[4−8]. While the defects or impurities and thermal diffusion play a negligible role in the damage for the optical coating samples in our experiment as mentioned above, so the damage in the optical coatings is intrinsic, too. Then like bulk material, photoionization and impact ionization are responsible for the damage. As a result, a simple model including producing the conduction band electrons by photoionization and impact ionization was used to explain the damage mechanism.

The rate of electron generation due to excitation is written as

$$\frac{{\rm d}n_{\rm e}}{{\rm d}t} = W_{\rm PI} + W_{\rm II} n_{\rm e}(t), \tag{4}$$

where $n_{\rm e}(t)$ is the electron density, $W_{\rm PI}$ is the photoionization rate described by Keldsh theory[9], $W_{\rm II}$ is the impact ionization rate[10].

In fact, the carrier generation is controlled by the local pulse intensity. Due to interference effects in film, the local intensity enhancements exist. Figure 4 shows the field distributions of ZrO_2 single layer and ZrO_2/SiO_2 HR coating calculated with TFCalc film design software. For HfO_2 and HfO_2/SiO_2 HR coating, the distributions are the same.

Combined the Eq. (4) with the laser intensity distribution of single layers, $n_{\rm e} = 10^{21}$ $\rm cm^{-3}$ taken as the damage criteria the theoretical damage thresholds of ZrO_2 and HfO_2 single layers were calculated and are shown in Fig. 5. It shows that the LIDT of HfO_2 is higher than that of ZrO_2. While the mainly different parameter value for two kinds of samples is band gap energy in our calculation, and HfO_2 (5.6 eV) has a wider band gap than ZrO_2 (5.0 eV). In fact, according to the expressions of $W_{\rm PI}$ and $W_{\rm II}$, the wider the band gap was, the smaller the $W_{\rm PI}$ and $W_{\rm II}$ in Eq. (4) would be. Namely, it would be more difficult to excite HfO_2 than ZrO_2. So HfO_2 single layer would have a higher LIDT, and this is in accord with the experimental result.

Figure 4 also indicates that the ZrO_2/ SiO_2 HR coating is most likely damaged at the first interface between ZrO_2 and SiO_2, which has the maximum of the field distribution. ZrO_2 (SiO_2) has a band gap of 5.0 eV (7.8 eV). Thus damage is likely to occur first in the high-index ZrO_2 layer. Then, for both the single layer and the HR coating, the damages all occurred first in

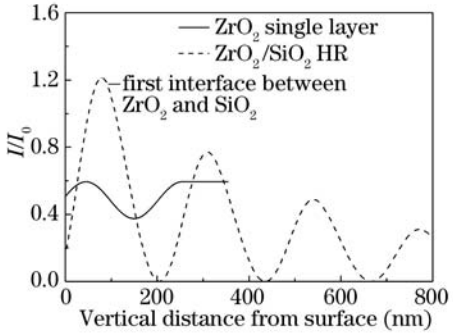

Fig. 4. Distributions of laser intensity for ZrO_2 single layer and ZrO_2/SiO_2 HR coating.

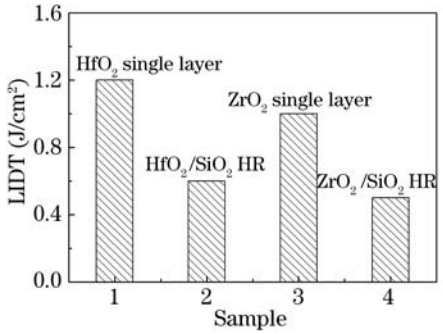

Fig. 5. Theoretical damage thresholds of all samples.

the ZrO_2. As a result, the damage thresholds for ZrO_2/SiO_2 and HfO_2/SiO_2 HR coating were also calculated and are shown in Fig.5. Then it can be seen that the LIDT of the ZrO_2/SiO_2 HR coating is lower than that of the ZrO_2 single layer. This relation would exist between HfO_2/SiO_2 HR coating and HfO_2 single layer, too. These are consistent with experiment. Meanwhile, HfO_2/SiO_2 HR coating show a higher LIDT than ZrO_2/SiO_2 HR coating because HfO_2 has a wider band gap than ZrO_2.

However, there is difference between experimental results and calculated data. Because there are many parameters in our calculation and it is difficult to get the accurate values.

Femtosecond pulse laser-induced damage of optical coatings including ZrO_2 and HfO_2 single layers, ZrO_2/SiO_2 and HfO_2/SiO_2 HR coatings is investigated. The damage morphologies of all samples are different from that induced by nanosecond laser. The heat diffusion plays a negligible role in the damage. The damage thresholds of all samples were reported and showed that the wider the band gap was, the higher the LIDT would be. Meanwhile, single layer showed higher LIDT than corresponding HR coating. A simple model is used to explain the damage mechanisms and calculate the theoretical damage thresholds, which agrees with our measurements.

This work was supported by the National Natural Science Foundation of China under Grant No. 10376040. L. Yuan's e-mail address is yuanlei@siom.ac.cn.

References

1. T. W. Walker, A. H. Guenther, and P. E. Nielsen, IEEE J. Quantum Electron. **17,** 2041 (1981).
2. L. G. Deshazer, B. E. Newnam, and K. M. Leung, NBS Special Publication **509,** 251 (1977).
3. W. Gao, H. He, J. Shao, and Z. Fan, Chin. Opt. Lett. **2,** 493 (2004).
4. B. C. Stuart, M. D. Feit, S. Herman, A. M. Rubenchik, B. W. Shore, and M. D. Perry, Phys. Rev. B **53,** 1749 (1996).
5. J. Jasapara, A. V. V. Nampoothiri, and W. Rudolph, Phys. Rev. B **63,** 045117 (2001).
6. J. Krüger, M. Lenzner, S. Martin, M. Lenner, C. Spielmann, A. Fiedler, and W. Kautek, Appl. Surf. Sci. **208−209,** 233 (2003).
7. A. Kaiser, B. Rethfeld, M. Vicanek, and G. Simon, Phys. Rev. B **61,** 11437 (2000).
8. T. Apostolova and Y. Hahn, J. Appl. Phys. **88,** 1024 (2000).
9. L. V. Keldysh, Sov. Phys. JETP **20,** 1307 (1965).
10. A. Vaidyanathan, T. W. Walker, and A. H. Guenther, IEEE J. Quantum Electron. **16,** 89 (1980).