Damage on HfO_2/SiO_2 high-reflecting coatings under single and multiple Nd:YAG laser pulse irradiation

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The single- and multi-shot damage behaviors of HfO_2/SiO_2 high-reflecting (HR) coatings under Nd:YAG laser exposure were investigated. Fundamental aspects of multi-shot laser damage, such as the instability due to pulse-to-pulse accumulation of absorption defect and structural defect effect, and the mechanism of laser induced defect generation, are considered. It was found in multi-shot damage, the main factors influencing laser-induced damage threshold (LIDT) are accumulation of irreversible changes of structural defects and thermal stress that induced by thermal density fluctuations.

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Laser-induced damage to optical materials as one of the fundamental problems in laser physics has been extensively studied for more than 40 years[1]. The most attention was paid to studies of single-shot damage. As a result, damage mechanisms for single-shot irradiation are now well understood[2,3]. However, whether or not accumulation effect takes place during multi-shot irradiations is still a somehow pending question for optical materials of different types: silicate glasses, crystals, polymer, and optical coatings[4,5]. In this work, we were seeking to understand the feature of multi-shot damage and analyze physical processes leading to the irreversible changes in optical materials, and then to consider possible methods of their suppression to increase laser damage resistance through studying the single- and multi-pulse Nd:YAG laser damages on HfO_2/SiO_2 high-reflecting (HR) coatings.

The HR coatings were prepared by conventional e-beam deposition, using the same deposition technology and in the same coating chamber. The film structure was 9(HL)H, where H and L represented a quarter-wave optical thickness of high (HfO_2) and low (SiO_2) refractive index materials, respectively. The laser damage was based on the small-spot technique for 1-on-1 or S-on-1 investigations according to the ISO standards 11254-1.2[6]. The laser source used for the experiments was a commercial Nd:YAG laser producing pulse of 12 ns duration in TEM_{00} mode at 1.06 μm. The pulse was Gaussian in time with a repetition rate of 1 Hz. A He-Ne laser was used to help in monitoring the test, as shown in Fig. 1. A rotating half-wave plate, followed by a thin-film linear polarizer, adjusted the incident laser intensity. The laser beam was focused with a 250-mm focal-length lens to a 450-μm diameter spot ($1/e^2$) on the sample surface. Online damage detection was possible by measuring video micrographs of the site before and after irradiation with the Nd:YAG laser of each investigation site on the sample. Leica micropolariscope allowed the final decision on laser damage and investigation of the damage morphologies.

The corresponding results of the laser-induced damage threshold (LIDT) measurements for the HfO_2/SiO_2 HR coatings are listed in Table 1. In the case of single-shot damage, the coating showed a typical damage behavior of HR coatings, the LIDT was 16.8 J/cm^2. In the case of multi-shot damage, the measured values were 9.3, 7.1, 6.8, and 6.6 J/cm^2 for 5-on-1, 10-on-1, 50-on-1, and 100-on-1, respectively. A distinct drop in LIDT was observed with the increase of laser shots, but the LIDT has no much difference when the laser shots go beyond 10, the LIDT difference maybe associated with statistic fluctuations in pulse energy. Obviously, this drop can certainly not be explained with a stochastical effect (i.e., due to an increased number of pulses). More likely, an accumulation effect may be caused[5,7].

In attempting to explain this effect, damage morphological investigations were conducted by a Leica micropolariscope. Figure 2 shows the single-shot damage morphologies under different irradiation intensities. From Fig. 2, it is easy to find that all the catastrophic macro-damages show typical morphologies of flat bottom pits. The damages are located close to the coating surface. They have a typical cylindrical shape and a small melted zone is clearly visible at the center of the damage. These damages were well described in Ref. [8] that this kind of damage is not related with a visible preexisting defect like a nodule, but initiated by some invisible almost punctual absorbing center.

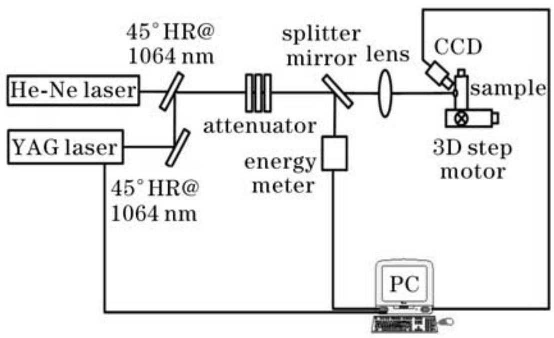

Fig. 1. Measure instrument of LIDT.

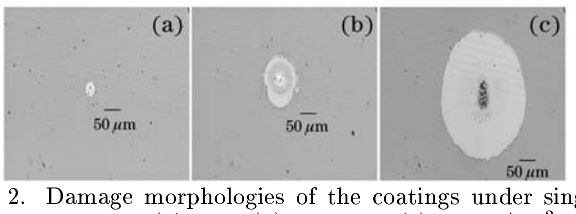

Fig. 2. Damage morphologies of the coatings under single-pulse irradiation. (a) 19.7, (b) 27.1, and (c) 45.4 J/cm^2.

Table 1. The LIDTs of the HR Coating under Different Laser Shots

Irradiation Mode	1-on-1	5-on-1	10-on-1	50-on-1	100-on-1
LIDT (J/cm^2)	16.8	9.3	7.1	6.8	6.6

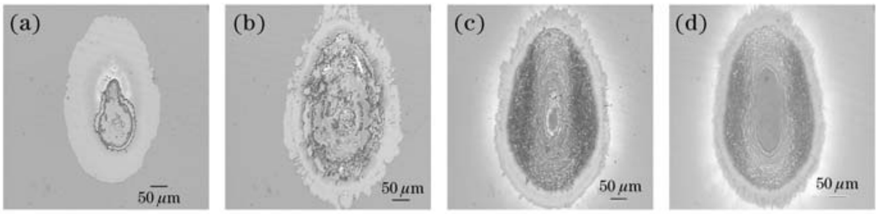

Fig. 3. Typical damage morphologies of laser-induced sites under different shots. (a) 7.6 J/cm^2, 5-on-1; (b) 7.6 J/cm^2, 10-on-1; (c) 7.4 J/cm^2, 50-on-1; (d) 7.5 J/cm^2, 100-on-1.

Figure 3 shows the damage craters of the coatings obtained under different irradiation shots with nearly constant laser irradiation density. For multi-pulse irradiation, the coating shows damage of cracking and etching, not ablation under the irradiation of the laser pulses. The layered structure in the crater depicts in Figs. 3(a)—(d). While the glass substrate remains intact for these irradiation conditions. The damage threshold of the dielectric coatings is clearly below that of the substrate. Therefore, an etch-stop can be observed.

As we know, under single-shot damage, the damage is often associated with absorbing defects and structural defects, and the absorbing defects often play the key role in the single-shot damage. The multi-shot damage features have been attributed for accumulation of pulse-to-pulse irreversible changes of optical properties of absorbing inclusion and ambient material matrix, and the laser-induced damage is often associated with the thermal instability because of the temperature nonlinearity of inclusions and ambient matrix[4]. But in our experiment, a particular phenomenon was found. The mirror is often damaged within the first ten shots. Usually, the mirror can hardly be damaged by the subsequent shots, if it cannot be damaged by the first ten shots. Moreover, the damage only extends, when the subsequent shots' irradiation intensities exceed some critical value, below this value, the damage morphologies keep undisturbed. Obviously, the customary conclusion, which is mainly dominated by the accumulation effect of thermal defect, cannot give a satisfied explanation about this phenomenon, because the temperature rise caused by the absorption of the absorbing defect and ambient material matrix in the multilayer is not too high to induce the failure under relatively lower irradiation density and the accumulation of the temperature rise in the multilayer also does not exist at such a low repetitive frequency.

One possible explanation is the accumulation of thermal stress, which induced by the thermal density fluctuations from pulse-to-pulse irradiations. In multi-shot damage, accumulation of pulse-to-pulse irreversible changes of the structural defects cannot be neglected and play an important role in the multi-shot damage, because it can enhance the local stress and degrade the strength of the multilayer[9]. When the local stress goes beyond the breaking point, the coating is cracked and the stress is discharged. Once the local stress is discharged, it needs new nucleation of local stress to extend the damage. This process probably needs higher laser irradiation intensity. For this reason, in the experiment, the damage only extends with the subsequent shots, when its irradiation intensity exceeding some critical value. This kind of accumulation has been seen from experiment process, during laser radiation the scatter of the He-Ne laser is enhanced step by step. Under the repetitive laser radiation, the damage process is as following: at first several shots, no visible change is observed, after that, there shows He-Ne laser scattering and the scattering light is strengthened as the shot number increases, at last, the visible damage can be observed.

Through the above discussions, it is clear that the methods of real material modification should be good choices to improve the multi-shot LIDT. The methods of the material modification, which minimize the influence of thermal density fluctuations and decrease the initial and laser produced stresses, should be aimed. Further work has been in progress.

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