Design and fabrication of a novel high damage threshold $HfO_2/TiO_2/SiO_2$ multilayer laser mirror*

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This paper describes a new method to design a laser mirror with high reflectivity, wide reflection bandwidth and high laser-induced damage threshold. The mirror is constructed by three materials of $HfO_2/TiO_2/SiO_2$ based on electric field and temperature field distribution characteristics of all-dielectric laser high reflector. TiO_2/SiO_2 stacks act as the high reflector (HR) and broaden the reflection bandwidth, while HfO_2/SiO_2 stacks are used for increasing the laser resistance. The $HfO_2/TiO_2/SiO_2$ laser mirror with 34 layers is fabricated by a novel remote plasma sputtering deposition. The damage threshold of zero damage probability for the new mirror is up to 39.6 J/cm^2 (1064 nm, 12 ns). The possible laser damage mechanism of the mirror is discussed.

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High laser-induced damage threshold (LIDT) all-dielectric mirrors are vital for high power laser applications[1]. To solve the problem of laser damage, the significant researches have been widely reported in recent years[2-5]. In design of high LIDT mirrors, the electric field design and temperature field design are studied[2,3]. In deposition techniques, the most important thing is to choose intrinsic high LIDT coating materials and to optimize the process parameters of different deposition technologies. The LIDTs of coatings based on SiO_2 and HfO_2 are intensively investigated[4-7]. However, the refractive index of HfO_2 is not very high ($n\approx2.01$ at 1064 nm) compared with other metal oxide coating materials, such as TiO_2 ($n\approx2.25$ at 1064 nm) and Nb_2O_5 ($n\approx2.20$ at 1064 nm). For the materials with high refractive index, the laser resistance is weak because of the intrinsic low melting point effect[8]. Due to this restraint, broad-band high-reflection laser mirrors are not easy to fabricate with only HfO_2/SiO_2 coating materials, and in the same reflectivity level they need more layers.

In this paper, a new laser mirror stack is proposed, which can realize high reflectivity, wide reflection bandwidth and high LIDT. This mirror is constructed by three materials: SiO_2, HfO_2 and TiO_2 based on electric field distribution characteristics of high reflector (HR). The laser mirror with 34 layers is fabricated by plasma sputtering deposition. The optical properties and LIDT of the mirror are measured. The possible laser damage mechanism of the mirror is discussed.

According to film optics theory, the standard quarter-wave stack substrate/$(HL)^S$H/air can achieve the highest reflectivity, and the reflectivity and bandwidth of HR are limited by several factors, including the number of stack layers and the refractive index difference between high refractive index (n_H) and low refractive index (n_L) materials. It has been established that the standing-wave electric field must be taken into account when the laser damage resistance is evaluated[9]. Here we calculate the electric field of a standard HR coating substrate/$(HL)^{10}$ H/air at λ_0=1064 nm with TiO_2/SiO_2 as shown in Fig.1. In the interior of TiO_2/SiO_2 stack, the electric field intensity is nearly zero. Moreover, the laser resistance of high index materials is typically less than that of lower index materials[10]. Thus the LIDT of HR is determined by the outermost high index material. However, there are few choices for the material with high refractive index. So the design of a new combined high LIDT laser mirror is provided, using $HfO_2/TiO_2/SiO_2$ materials. In the inner stacks of mirror, the high reflectivity is satisfied by the higher n_H/n_L and less stacks (TiO_2/SiO_2), and in the easily damaged outer stacks, the high LIDT is satisfied by the intrinsic high LIDT properties of

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HfO_2/SiO_2. To achieve better laser resistance, the half-wave SiO_2 overcoats are inserted between the HfO_2/SiO_2 stack and TiO_2/SiO_2 stack, and coated on the top of the whole stack. The schematic diagram of the combined mirror stack is shown in Fig.2.

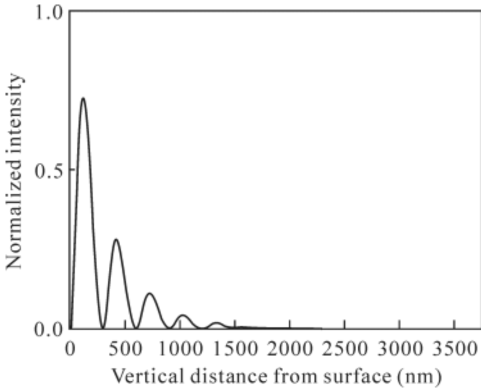


Fig.1 Electric field distribution in high reflector sub/(HL)^{10} H/air at λ_0=1064 nm with TiO_2/SiO_2

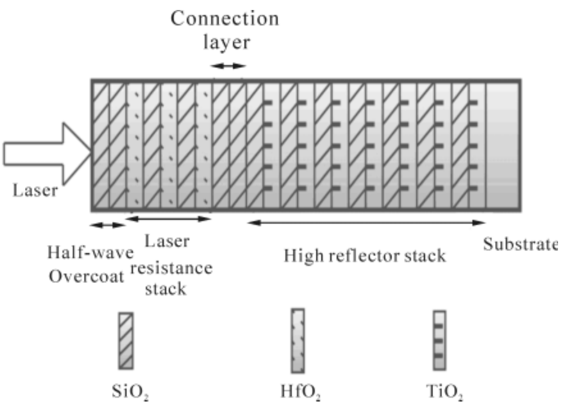


Fig.2 Schematic diagram of the combined $HfO_2/TiO_2/SiO_2$ laser mirror stack

Firstly, a quarter-wave mirror stack with 34 layers is given as substrate/(HL)^{16} H4L/air with reflectivity of 99.992% at 1064 nm. It is found that except for the distribution in half-wave SiO_2 overcoat, the higher electric field intensity occurs at the interface of outer three pairs of the layers of materials with high and low refractive indices, and the maximum intensity is 0.72 V/m. The three pairs of stacks, which are the easiest damaged, are replaced by the HfO_2/SiO_2 stacks. Then the new stack is given as substrate/(HL)^{13} 2L(ML)^3 M4L/air. Because of using high LIDT stack HfO_2/SiO_2, the laser resistance can be obviously improved. The LIDT of $HfO_2/TiO_2/SiO_2$ HR is determined by the outer HfO_2/SiO_2 stack. In Fig.3, the higher electric fields distribute in HfO_2/SiO_2, and the maximum electric field intensity in TiO_2 is only 0.074 V/m.

The characteristics comparison of different mirrors is shown in Tab.1. It is found that the bandwidth (R>99%, λ_0=1064 nm) of HfO_2/SiO_2 mirror with 34 layers is only 170 nm, the reflectivity is only 99.961%, and the whole physical thickness is 5890 nm. The bandwidth of HfO_2/SiO_2 mirror is 80 nm lower than that of TiO_2/SiO_2, and the reflectivity at central wavelength is 0.03% less than that of TiO_2/SiO_2. Using the new $HfO_2/TiO_2/SiO_2$ mirror, the reflectivity at central wavelength is still up to 99.982%. The first main reflectance bandwidth is 230 nm. The whole physical thickness is 6072 nm. Therefore the new combined mirror can simultaneously realize the high reflectivity, wide bandwidth and high laser resistance at the fixed physical thickness.

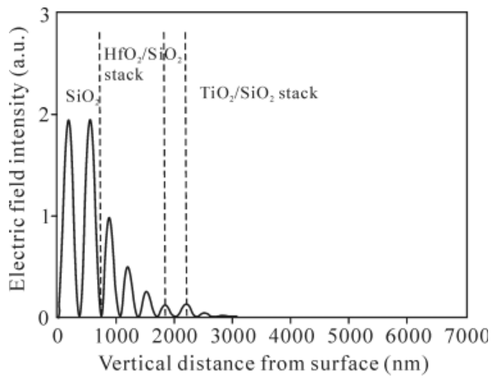


Fig.3 Electric field intensity distribution in $HfO_2/TiO_2/SiO_2$ laser mirror with structure of substrate/(HL)^{13} 2L(ML)^{3}M 4L/air at λ_0=1064 nm

Tab.1 Characteristics comparison of different mirrors

Mirror stack	Reflectivity at 1064 nm (%)	Reflectance bandwidth (nm)	Physical thickness (nm)	LIDT (J/cm^2)
(HL)^{16}H4L	99.992%	250	5650	24.6
(ML)^{16}M4L	99.961%	170	5890	43.8
(HL)^{13} 2L(ML)^{3}M4L	99.982%	230	6072	39.6

In experiment, the laser mirrors are deposited by a novel remote plasma sputtering technique[11,12]. The systems use cryo-pump with a base pressure of 6×10^{-6} Torr. Pure argon gas is introduced to the chamber, and the oxygen is fed into the chamber through another diffusion ring placed as close as possible to the substrate. Silicon (99.999% purity), titanium (99.99% purity) and hafnium (99.99% purity) targets are used in direct current (DC) sputtering mode. The quartz glasses with high surface quality of 10-5 scratch-dig are used to deposit the films. The optical transmittances of the samples are measured using Perkin-Elmer Lambda 750 spectrometer. LIDTs of samples are tested using the Nd:YAG laser as radiation source with 12 ns pulses at 1064 nm and the maximum pulse energy of 100 mJ, and the output laser beam TEM_{00} mode is focused onto the sample using a lens with focal length of 30 cm to generate 400 μm diameter laser beam at normal incidence. 1-on-1 testing mode is performed by the facility. LIDT is determined by a linear extrapolation of the damage probability data to zero damage probability.

The measured transmittances of multilayer mirrors are shown in Fig.4. It is found that the bandwidth (R>99%, λ_0=1064 nm) of $HfO_2/TiO_2/SiO_2$ mirror is up to 230 nm. The transmittance at 1064 nm is 0.00035%. From the measurement data, it is concluded that the reflectivity exceeds 99.8% at 1064 nm.

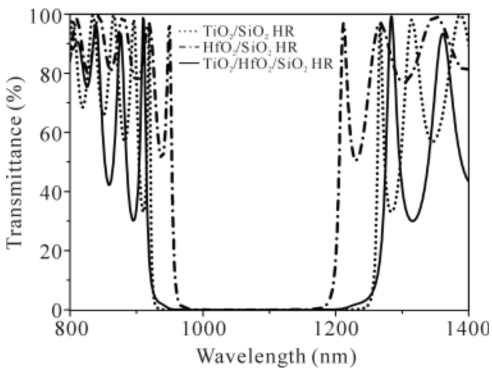
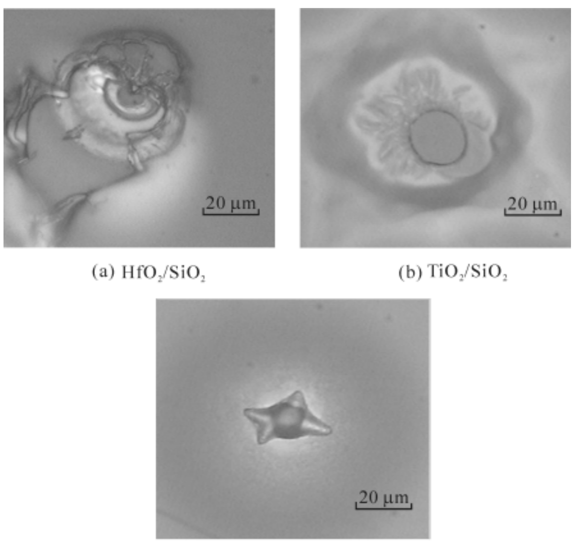


Fig.4 Experimental transmittance spectra for different laser mirror stacks at 1064 nm

All samples during damage testing have the permanent damage morphology and mechanical breaking on the film surface under 500× magnification. The laser damage threshold is determined by a linear extrapolation of the damage probability data to zero damage probability as shown in Tab.1. The damage morphologies of the mirrors are shown in Fig.5. Damage morphology exhibits the pitting and delamination damage of HfO_2/SiO_2 mirror. The damage morphology of TiO_2/SiO_2 shows obvious thermal melting and crapy morphology because of the low melting point of TiO_2. But for the $HfO_2/TiO_2/SiO_2$ mirror, the above damage morphology can not be seen. It seems that the damage happens in the inner stack of TiO_2/SiO_2, but this thermal melting is confined by the outer HfO_2/SiO_2 layer, which results in that it looks like the sun going to blast. From the damage morphology, it also suggests that the breaking is likely to occur first in the high-index TiO_2 layer. Because of the SiO_2 half-wave protection coating, better contribution to increase the damage resistance can be given.



(a) HfO_2/SiO_2 (b) TiO_2/SiO_2

(c) $HfO_2/TiO_2/SiO_2$

Fig.5 Surface damage morphologies of laser mirrors

In conclusion, a new design method of combined mirror is developed using $HfO_2/TiO_2/SiO_2$ based on the electric field and temperature field distribution characteristics of all dielectric HR. The design simultaneously realizes high reflectivity, wide reflection bandwidth and high LIDT by using less film layers. The combined laser mirror with 34 layers is fabricated by remote plasma sputtering deposition. The LIDT of the mirror is up to 39.6 J/cm^2. The method is also realized by other high and middle index materials with SiO_2 combination, such as Nb_2O_5, Ta_2O_5, Y_2O_3, Al_2O_3, etc. So the development of higher damage threshold middle index materials is underway to expand the capability of the method.

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