Laser-induced damage of high reflectors for Ti:sapphire laser system

Jianke Yao (姚建可), Weiqiang Zeng (曾维强), Zhengxiu Fan (范正修), Hongbo He (贺洪波), and Jianda Shao (邵建达)

R&D Center for Optical Thin Film Coatings,

Shanghai Institute of Optics and Fine Mechanics, Chinese Academy of Sciences, Shanghai 201800

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A broadband (~ 176 nm, R > 98%, $\lambda_0 = 800$ nm) and high laser-induced damage threshold (LIDT = 2.4 J/cm²) TiO₂/HfO₂/SiO₂ high reflector (HR) for Ti:sapphire chirped-pulse amplification (CPA) laser system is fabricated by the electron beam evaporation. The refractive index and extinction coefficient of TiO₂ and HfO₂ films are calculated from single-layer films' transmittance spectra. The properties of HR are mainly determined by the high refractive index material. The high refractive index leads to wide bandwidth. A low extinction coefficient indicates low absorption and high LIDT. The possible damage mechanism of HR is discussed.

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In the first half of 2006, a 120-TW, 36-fs laser system based on Ti:sapphire chirped-pulse amplification (CPA) was successfully established in China^[1]. To achieve terawatt pulse width in the range of 15 - 20 fs, a broadband mirror with the working spectral bandwidth of $\Delta \lambda = 150 - 200 \text{ nm}$ at center wavelength λ_0 of 800 nm is required, because one of the limiting factors of the pulse width is the bandwidth of mirror in an amplifier chain^[2]. Optical damage owing to the compressed pulses is a major concern in high-peak-power systems of this $type^{[3]}$. Further increases in peak power available from such systems are now limited by damage to optical surfaces due to the intense short pulses. The laser induced damage issue in CPA laser system should take into account an important pulse duration occurring in 800-nm signal pulses with the duration of a few hundreds of picoseconds after the stretcher during the amplification. In this time domain, a laser-induced damage threshold (LIDT) higher than $1 \text{ J/cm}^{2[2]}$ should be maintained.

The coating parameter that most strongly influences LIDT is the choice of materials used to produce a component. TiO₂ is a coating material with highly desired properties. It is hard and chemically resistant. It is transparent in the visible and near infrared (NIR) range, and it has a high refractive index which is useful for multilayer dielectric mirror^[4]. HfO₂ is also one of the most commonly used high-index materials to realize laser damage resistance for its relatively high LIDT and good thermal and mechanical stability^[5].

In this paper, a broadband high LIDT mirror is deposited by electron beam evaporation, using TiO_2 and HfO_2 as high refractive materials and SiO_2 as low refractive material. The optical properties and LIDT of this mirror are measured. Moreover, the possible laser damage mechanism of the mirror is discussed.

In experiment, the single-layer films' optical thickness is $4\lambda_1$, where λ_1 is 500 nm. The structure of TiO₂/SiO₂ high reflector (HR) is (HL)¹³H, for HfO₂/SiO₂ HR it is (ML)¹³M, and that of TiO₂/HfO₂/SiO₂ HR is (HL)¹¹(ML)⁹M, where H stands for the quarter wavelength optical thickness of TiO₂, M stands for that of HfO₂, and L for SiO₂. TiO₂, HfO₂ and SiO₂ are used as high-, middle- and low-refractive-index materials, respectively. High-quality BK7 substrates were cleaned ultrasonically in an alcohol solution before deposition. All films were deposited by electron beam evaporation in the following process parameters. The base pressure was 2×10^{-3} Pa before deposition and the oxygen partial pressure was 3×10^{-2} Pa during deposition. The substrate temperature was kept at 300 °C during deposition. The deposition rate of TiO₂, HfO₂, and SiO₂ were 1.33, 1.33, and 0.2 nm/s, respectively.

The transmittance and reflectance spectra of samples were measured by Perkin-Elmer Lambda 900 spectrophotometer. Refractive index n and extinction coefficient kof films are calculated from the transmittance spectrum by the envelope method^[6].

LIDTs of samples were tested using the chirped pulse train ($\lambda_0 = 800$ nm, $\tau_p = 220$ ps, 10 Hz, incident angle near 0°) from a 23-TW Ti:sapphire laser system^[7]. The detailed test process is shown in Ref. [8]. The effective spot diameter was around 3 mm with the fluctuation of ±10%. The relative uncertainty of the LIDT-determination amounted to ±20%, which was mainly due to the uncertainty in the spot size measurements.

Figure 1 shows the transmittance spectra of films. At the wavelength corresponding to the optical thickness of one half wave, the transmittances of both TiO₂ and HfO₂ films are observed to be nearly the same as that of the substrate. The spectra of HfO₂ film shifts to short wavelength region and has a shorter cutoff wavelength ($\lambda_c = 264$ nm) than that of TiO₂ film ($\lambda_c = 315$ nm). The refractive index and extinction coefficient of films are shown in Fig. 2. It is shown that the refractive index and extinction coefficient of HfO₂ film are lower than those of TiO₂ film.

Figure 3 shows the measured reflectivity of HR. It is found that the bandwidth (R > 98%, $\lambda_0 = 800$ nm) of TiO₂/HfO₂/SiO₂ HR (~ 176 nm) is almost equal to that



Fig. 1. Transmittance spectra of single-layer films.



Fig. 2. (a) Refractive index and (b) extinction coefficient of TiO_2 and HfO_2 films.



Fig. 3. Measured reflectivity of HRs.

of TiO₂/SiO₂ HR (\sim 187 nm), and is wider than that of HfO₂/SiO₂ HR (\sim 146 nm).

It has been established that the standing-wave electric field (SWEF) must be taken into account when evaluating laser damage resistance^[9]. Breakdown most likely occurs at the location of the SWEF maximum in the film. The theoretical result of electric field distributions of $TiO_2/HfO_2/SiO_2$ HR calculated by the thin film



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Fig. 4. Calculated electric field intensity in $\rm TiO_2/\rm HfO_2/\rm SiO_2$ HR.

Table 1. LIDTs of Single-Layer Films and HR

High-Index	LIDT (J/cm^2)	
Material	Single-Layer Film	HR
TiO ₂	0.61	1.20
HfO_2	1.09	2.20
${\rm TiO_2/HfO_2}$	—	2.40

design software TFCalc is shown in Fig. 4. It is found that the SWEF maximum occurs at the interface of the outermost layers of high- and low-refractive-index materials and SWEF decreases from air to substrate. The SWEF in TiO_2/SiO_2 stack is nearly zero.

Table 1 shows the LIDTs of deposited films. The LIDTs of single-layer HfO_2 film and HfO_2/SiO_2 HR are larger than those of TiO_2 single-layer film and TiO_2/SiO_2 HR, respectively. The LIDT of $TiO_2/HfO_2/SiO_2$ HR is almost equal to that of HfO_2/SiO_2 HR.

The absorption coefficient α can be calculated using the relation $\alpha = 4\pi k/\lambda$, where k is the extinction coefficient, λ is the wavelength. The cutoff wavelength λ_c is defined as the wavelength at which the transmittance is zero from the single-layer film's transmittance spectra. λ_c can be considered as a qualitative indicator of the degree of absorption^[10]. So, a low extinction coefficient and a short cutoff wavelength indicate a low absorption. Figures 1 and 2(b) show that the absorption of TiO₂ film is larger than that of HfO₂ film.

According to the theoretical results on the temperature field distribution of a standard HR coating, the temperature rise peaks decrease from air to substrate. The highest temperature rise occurs at the interface of the outermost layers of high- and low-refractive-index materials, which causes massive heat deposition, and thus originates the laser-induced damage^[11]. Moreover, the laser damage resistance of high-index materials is typically less than that of lower-index materials^[12]. Dam-</sup> age is likely to occur first in the high-index layer. Thus the LIDT of HR is determined by the outermost highindex material. Table 1 shows that LIDT is absorption dominated. HfO_2 film has lower absorption than that of TiO_2 film, so the LIDTs of HfO_2 films are higher than those of TiO_2 films. Since the SWEF in TiO_2/SiO_2 stack is nearly zero in the $TiO_2/HfO_2/SiO_2$ HR, the LIDT of $TiO_2/HfO_2/SiO_2$ HR is determined by the outer

 HfO_2/SiO_2 stack and the absorption of HfO_2 , which increase the LIDT of $TiO_2/HfO_2/SiO_2$ HR to nearly equal to that of HfO_2/SiO_2 HR.

The bandwidth of HR is limited by several factors including the difference between high-refractive-index $(n_{\rm H})$ and low-refractive-index $(n_{\rm L})$ materials. The higher the $n_{\rm H}/n_{\rm L}$, the wider is the spectral bandwidth of the HR. The refractive index is closely related to the electronic polarizability of ions and the local field inside the material^[13]. TiO₂ films have higher refractive index than HfO₂ films (Fig. 2) due to the different positions of the metals in the periodic table. Therefore, the bandwidth of TiO₂/SiO₂ HR is wider than that of HfO₂/SiO₂ HR.

In conclusion, we have fabricated a broadband high LIDT HR for terawatt CPA of ~ 36 fs pulses. The usable spectral range and LIDT of HR are traded off against each other. The higher index difference between high-refractive-index and low-refractive-index materials, the wider is the spectral bandwidth of the HR. The laser induced damage in subnanosecond pulses is absorption dominated and relates to material parameters such as extinction coefficient and cutoff wavelength.

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