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Strategies to increase laser damage performance of Ta_2O_5/SiO_2 mirrors by modifications of the top layer design

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Ta_2O_5/SiO_2 high reflection (HR) interference coatings for $\lambda \sim 1$ µm offer superior performance at high irradiance conditions. However, these coatings are not good candidates for high peak power conditions in comparison to HfO_2/SiO_2 multilayer stacks. Here we show that the modification of the top layers design of a quarter wave Ta_2O_5/SiO_2 high reflector leads to 4–5 fold increase in the laser damage fluence compared to a quarter wave $(Ta_2O_5/SiO_2)^{15}$ when tested at $\lambda = 1.03$ µm using pulse durations of 0.19 and 4 ns and peak power densities of 43.5 and 216 GW/cm^2. One of the designs achieved a laser damage threshold fluence of 174 J/cm^2 at 4 ns, which is 10% higher than that of a HfO_2/SiO_2 quarter wave design.

OCIS codes: (310.1620) Interference coatings; (310.6860) Thin films, optical properties; (230.4170) Multilayers; (140.3330) Laser damage.

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1. INTRODUCTION

Ta_2O_5/SiO_2 high reflection (HR) interference coatings are ubiquitous in the architecture of near infrared high average power lasers. Ta_2O_5/SiO_2 HRs are also critical coating components in the mirrors of gravitational wave interferometers [1] and in grating compressors in the 1ω beam line at the National Ignition Facility [2,3]. When Ta_2O_5/SiO_2 multilayer stacks are grown by physical vapor deposition, coatings with absorption and scattering losses in the few to tens of parts per million and very low stress can be realized. Although Ta_2O_5/SiO_2 HRs are capable of withstanding irradiances much greater than 1 MW/cm^2, they perform poorly at high peak power conditions at wavelengths near 1 µm. For high peak power applications, it would be highly desirable to keep the low stress, low absorption and scattering loss of Ta_2O_5/SiO_2 HRs and to increase laser damage performance to the level of HfO_2/SiO_2 HR multilayer stacks, which are the most broadly used for these applications.

In this paper, we describe different strategies in the design of Ta_2O_5/SiO_2 HR coatings deposited by ion beam sputtering that led to a ~5 × improvement of the laser damage threshold when tested at $\lambda = 1.03$ µm at pulse durations of 0.19 and 4 ns. At a pulse duration of 4 ns, the modified Ta_2O_5/SiO_2 coating outperforms a quarter wave HfO_2/SiO_2 HR.

Design strategies are based on two distinct approaches: (1) modifications to the standing wave electric field distribution and (2) modifications in the coating design to incorporate thicker $(3/4\lambda)$ SiO_2 layers for increased mechanical and thermal stability. The importance of the standing wave electric field distribution within the coating has been previously established in the short pulse regime. Apfel have derived a structure with thinner high index layers, reducing the peak electric field in the more damage-susceptible high index material by 33% and shifting the peaks in the standing wave distribution away from the material interfaces [4]. Such a design has been previously tested with 30 ps pulses at $\lambda = 1064$ nm, in TiO_2/SiO_2 stacks where the threshold was increased by a factor of 2.6 higher than a quarter wave design [5]. Evidence of increased thermo-mechanical stability and the relation to laser damage initiation has been demonstrated by Stolz et al, where an added $\lambda/2$ SiO_2 layer provided a 4-fold increase in the 3 ns laser induced damage threshold (LIDT) of HfO_2/SiO_2 Brewster's angle polarizers at $\lambda = 1064$ nm [6]. These improvements were associated with a much lower thermal expansion coefficient of SiO_2 compared with HfO_2 and to differences in the heat dissipation between the two materials that create a thermal and stress profile that affects laser damage. The use of a $\lambda/2\,SiO_2$ top layer is customary in commercially available multilayer dielectric coatings.

2. EXPERIMENTAL DETAILS

HR interference coatings were deposited on superpolished fused silica substrates with a Veeco Spector ion beam sputter deposition system. The accelerating voltage of the Ar^+ beam was set at 1250 V, and the current was 600 mA. No ion assisting or post annealing procedures were performed. The growth parameters for each material were selected to achieve stoichiometric films with low defect densities based on our previous research [7,8]. High reflectors (HR) were designed with a center wavelength of 1030 nm. Several modifications were made to the base coating structure consisting of 15 quarter wave stack layers, $(Ta_2O_5/SiO_2)^{15}$. In all of the modified designs, Ta_2O_5 is replaced with HfO_2 in the top high index layers where the standing wave electric field is most intense. This design takes advantage of the superior optical properties of Ta_2O_5, while utilizing higher damage threshold HfO_2 in the more damage-susceptible top layers. At the fourth high index layer, the electric field in the high index material reaches the $1/e$ point; thus, these top layers have a dominating effect on the laser damage threshold. In combination with this design, another structure with reduced thickness high index top layers was deposited, a technique derived from Apfel [4]. A final modified design employs an added $\lambda/2\,SiO_2$ overcoat and $3\lambda/4\,SiO_2$ top layers to improve the mechanical stability of the HR coating. For comparison, a reference quarter wave $(HfO_2/SiO_2)^{15}$ stack was also fabricated. Figure 1 displays the coating designs along with the standing wave electric field distribution in each structure calculated using Essential Macleod software.

Normal incidence transmission was measured with a Horiba UV-Vis spectrophotometer operating at $\lambda = 190$–1100 nm to confirm a reflectivity greater than 99% at the designed center

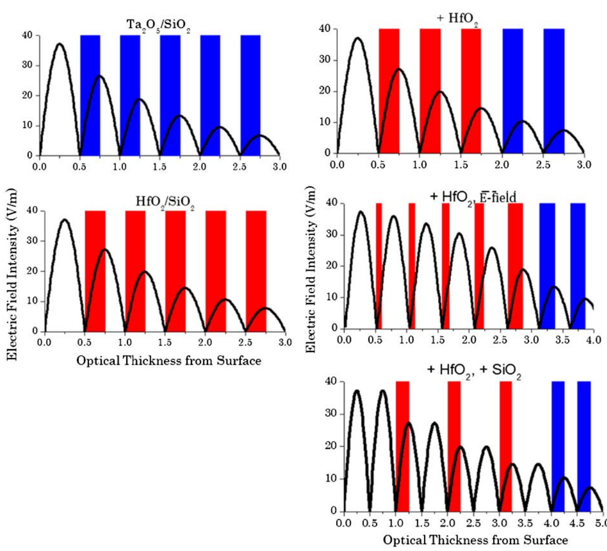

Fig. 1. HR top layer design with the standing wave electric field distribution obtained from Essential Macleod software for an incident intensity of 1 W/m^2 at $\lambda = 1.03$ μm. Red, blue, and white regions identify HfO_2, Ta_2O_5, and SiO_2, respectively.

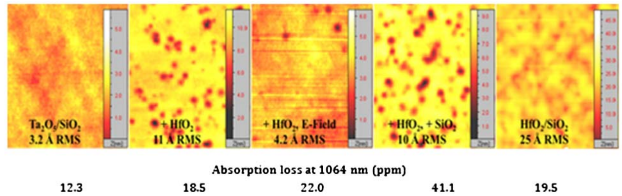

Fig. 2. Atomic force micrographs displaying the surface morphology and RMS surface roughness for each sample. Displayed micrographs are 5 × 5 μm. The corresponding absorption loss is listed.

wavelength of $\lambda = 1.030$ μm. The surface of the structures was inspected with a Novascan ESPM 3D atomic force microscope (AFM). The root mean squared (RMS) roughness was taken from 5 × 5 μm scan areas. The Ta_2O_5/SiO_2 quarter wave surface exhibits an RMS roughness of 3.2 Å, nearly half that of the 6 Å RMS measured for the superpolished fused silica substrate. The HfO_2/SiO_2 structure has an RMS roughness of 25 Å, eight times larger than the Ta_2O_5/SiO_2 structure. The surface roughness of all modified designs is less than half that of the HfO_2/SiO_2 structure. This indicates the excellent surface and film smoothness and uniformity obtained when using Ta_2O_5 as a high index material. AFM images of the surface morphology of the different HR structures are shown in Fig. 2. The absorption loss of the HR coatings at 1.064 μm determined from photothermal common path interferometry [9] is also summarized in Fig. 2.

Laser damage measurements were performed utilizing a Yb:YAG chirped pulse amplification laser system [10,11]. This system begins with 300 fs pulses from a Yb:KYW oscillator that were stretched to 0.19 ns and sent through a 100 Hz repetition rate Yb:YAG regenerative amplifier. Then, an amplification stage consisting of a cryogenically cooled Yb:YAG thick disk amplifier in an active mirror configuration was used to amplify these pulses to several tens of millijoules [12]. Pulses of 4.4 ns duration were obtained by operating the regenerative amplifier as an unseeded Q-switched, cavity-dumped oscillator. The LIDT fluence was obtained following the ISO-21254 protocol in a 100-on-1 configuration. Laser pulses were focused to a ~100 μm full width at half-maximum focal spot size while events were detected from the scatter of an incident HeNe laser and confirmed through post-inspection using a Nomarski microscope with a 100 × objective. The damage onset, 0%, and the 50% damage probability were obtained from a linear fitting of the damage probability curves.

3. RESULTS AND DISCUSSION

Figure 3 shows the 0% and 50% probability LIDT fluence for the different coating designs for (a) 4 ns and (b) 0.19 ns. When testing with 4 ns pulses, it was found that the combination of replacing the top three Ta_2O_5 layers of the base coat HR structure with HfO_2 and using ¾λ SiO_2 layers achieves the highest LIDT performance, with a LIDT fluence of 174 J/cm^2, that is, 4.5 × higher than the base coat $(Ta_2O_5/SiO_2)^{15}$. The peak intensity in these tests was 43.5 GW/cm^2. As shown in Fig. 3, this design outperforms by 10% the LIDT of the HfO_2/SiO_2 HR quarter wave stacks. The other designs also

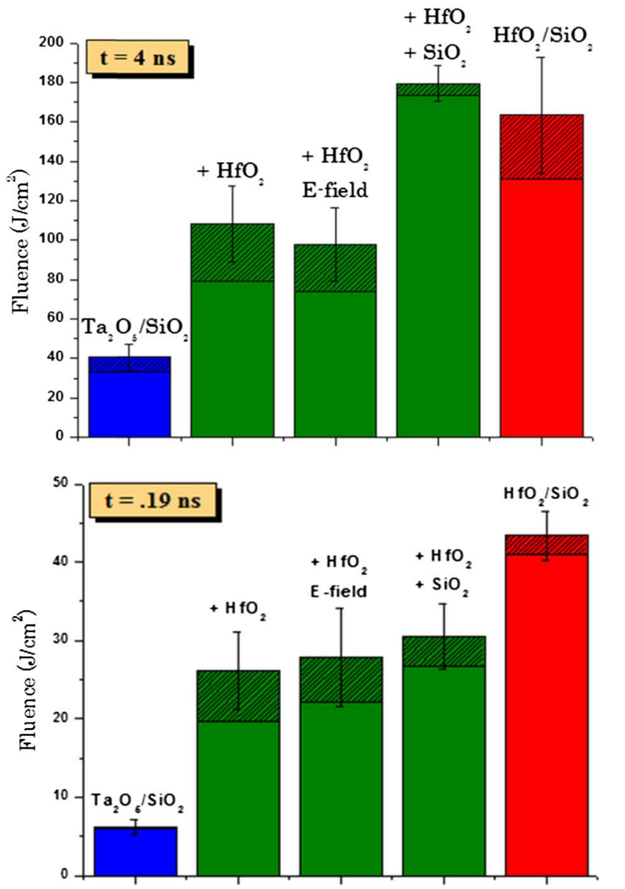

Fig. 3. Measured 0% LIDT (colored) and 50% damage probability fluence (shaded) for each structure at (a) 4 ns and (b) 0.19 ns pulse durations using a 100-on-1 test.

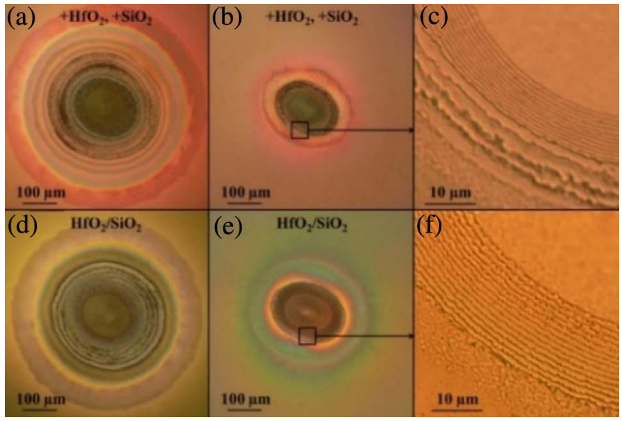

Fig. 4. Normaski microscope images of damage morphologies for the $+HfO_2 + SiO_2$ and HfO_2/SiO_2 structures at pulse durations of 4 ns (a, b) and 0.19 ns (d, e), respectively. (c, f) Higher magnification images of the coatings damage region.

showed improvements in the LIDT of over 2 ×, in comparison to the base coat structure $(Ta_2O_5/SiO_2)^{15}$. Previous experiments have investigated the LIDT of Ta_2O_5/SiO_2 quarter wave stacks, obtaining 55 J/cm^2 when irradiated with 5 ns pulses at 1064 nm wavelength [13]. HfO_2/SiO_2 HR designs operating at 1064 nm wavelength have achieved an LIDT as high as 105 J/cm^2 at 3 ns pulse duration [14], similar to those reported here. Others have used HfO_2 to replace the top three Titania layers in a TiO_2/SiO_2 quarter wave stack, where the modified structure exhibited a LIDT fluence of 39.6 J/cm^2 LIDT when tested at $\lambda = 1.064$ μm, 12 ns source, which is a nearly 60% increase from the TiO_2/SiO_2 quarter wave reference structure [15]. In previous work, we have also shown a 50% increase in the LIDT of quarter wave Ta_2O_5/SiO_2 HRs using a strategy whereby the top three layers of Ta_2O_5 were replaced by Y_2O_3 or HfO_2 [8]. It is important to point out that the design with the electric field modified (HfO_2 + E-field) does not offer any clear advantages in terms of damage performance compared with the $+HfO_2$ design when tested with a 4 ns source.

The LIDT behavior of the base coating design $(Ta_2O_5/SiO_2)^{15}$ HR and the modified designs determined at pulse durations of 0.19 ns and a peak intensity of 216 GW/cm^2 are shown in Fig. 3(b). Under these test conditions, an increase of ∼4× in the LIDT of the modified designs compared to the base coat design is measured. These improvements in LIDT are significant, suggesting that the LIDT performance is dominated by the top layers in the structure. The best performing design has a LIDT fluence that is ∼30% lower than the LIDT of the quarter wave HfO_2/SiO_2 HR. An observation drawn from the results of Fig. 3 reveals that the scaling of the LIDT fluence with the pulse duration is not the same for all structures, and furthermore, it does not follow he square root behavior expected when thermal damage dominates [16].

Damage morphologies for the HfO_2/SiO_2 and $+HfO_2$, $+SiO_2$ structures at a fluence near the LIDT are displayed in Fig. 4 for 4 ns and 0.19 ns pulse durations. The morphologies of the two pulse durations look similar. Each damage site exhibits a central region where the coating has been completely ablated, revealing the fused silica substrate. This crater transitions to a series of visible rings revealing the ablated layers of the coating. In each case, the number of rings is equivalent to the number of bilayers in the design. Further yet, from the center of the crater there is a discolored region, which, in the case of the nanosecond tests, shows ripple formations. These are attributed to the intense shockwave the multilayer experiences during plasma formation. One such investigation into the dynamics of laser induced damage has estimated that nearly 52% of absorbed radiation is dissipated through shockwave-propagation [17]. Damage is initiated in the top layers of the coating where the electric field intensity is maximum. The added SiO_2 enhances the mechanical stability of the top layers by reducing the peak thermal induced strain.

In summary, we have shown that $Ta_2O_5/HfO_2/SiO_2$ HR interference coatings designed for normal incidence at $\lambda = 1.030$ μm achieved a 4–5 fold increase in the LIDT tested at 0.19 and 4 ns when the top layer of the base coat consisting of a Ta_2O_5/SiO_2 quarter wave stack is replaced by HfO_2 top layers. The incorporation of $3\lambda/4\,SiO_2$ low index layers near the coating/air interface is very effective at increasing the LIDT, likely due to improved mechanical-thermal properties of the structure, as previously shown [6]. At 4 ns pulse duration, this modified structure exhibits a LIDT of 174 J/cm^2, higher than a HfO_2/SiO_2 quarter wave reference structure. While laser damage testing results can be difficult to compare, this LIDT value is amongst the highest values reported in the literature.

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REFERENCES

1. LIGO Scientific and VIRGO Collaborations, "Advanced LIGO," Class. Quantum Grav. **32**, 115012 (2015).
2. J. A. Britten, W. Molander, A. M. Komashko, and C. P. J. Barty, "Multilayer dielectric gratings for petawatt-class laser systems," in *Laser-Induced Damage in Optical Materials*, G. J. Exarhos, A. H. Guenther, N. Kaiser, K. L. Lewis, M. J. Soileau, and C. J. Stolz, eds. (2003), pp. 1–7.
3. B. W. Shore, M. D. Perry, J. A. Britten, R. D. Boyd, M. D. Feit, H. T. Nguyen, R. Chow, G. E. Loomis, and L. F. Li, "Design of high-efficiency dielectric reflection gratings," J. Opt. Soc. Am. A **14**, 1124–1136 (1997).
4. J. H. Apfel, "Optical coating design with reduced electric-field intensity," Appl. Opt. **16**, 1880–1885. (1977).
5. B. E. Newnam and D. H. Gill, "Laser damage resistance and standing-wave fields in dielectric coatings," J. Opt. Soc. Am. **66**, 166 (1976).
6. C. J. Stolz, F. Y. Genin, T. A. Reitter, N. Molau, R. P. Bevis, M. K. vonGunten, D. J. Smith, and J. F. Anzellotti, "Effect of SiO2 overcoat thickness on laser damage morphology of HfO2/SiO2 Brewster's angle polarizer at 1064 nm," Proc. SPIE **2**, 265–272 (1997).
7. P. F. Langston, E. Krous, D. Schiltz, D. Patel, L. Emmert, A. Markosyan, B. Reagan, K. Wernsing, Y. Xu, Z. Sun, R. Route, M. M. Fejer, J. J. Rocca, W. Rudolph, and C. S. Menoni, "Point defects in Sc_2O_3 thin films by ion beam sputtering," Appl. Opt. **53**, A276–A280 (2014).
8. D. Schiltz, D. Patel, L. Emmert, C. Baumgarten, B. Reagan, W. Rudolph, J. J. Rocca, and C. S. Menoni, "Modification of multilayer mirror top-layer design for increased laser damage resistance," Proc. SPIE **9237**, 92371G (2014).
9. A. Alexandrovski, M. Fejer, A. Markosyan, and R. Route, "Photothermal common-path interferometry (PCI): new developments," Proc. SPIE **7193**, 71930D (2009).
10. C. Baumgarten, M. Pedicone, H. Bravo, H. C. Wang, L. Yin, C. S. Menoni, J. J. Rocca, and B. A. Reagan, "1 J, 0.5 kHz repetition rate picosecond laser," Opt. Lett. **41**, 3339–3342 (2016).
11. B. A. Reagan, K. A. Wernsing, A. H. Curtis, F. J. Furch, B. M. Luther, D. Patel, C. S. Menoni, and J. J. Rocca, "Demonstration of a 100 Hz repetition rate gain-saturated diode-pumped table–top soft x-ray laser," Opt. Lett. **37**, 3624–3626. (2012).
12. A. H. Curtis, B. A. Reagan, K. A. Wernsing, F. J. Furch, B. M. Luther, and J. J. Rocca, "Demonstration of a compact 100 Hz, 0.1 J, diode-pumped picosecond laser," Opt. Lett. **36**, 2164–2166 (2011).
13. Z. Qiao, P. Ma, H. Liu, Y. T. Pu, and Z. C. Liu, "Laser-induced damage of rugate and quarter-wave stacks high reflectors deposited by ion-beam sputtering," Opt. Eng. **52**, 086103 (2013).
14. H. F. Jiao, X. B. Cheng, Z. X. Shen, B. Ma, J. L. Zhang, T. Ding, P. F. He, and Z. S. Wang, "Study of laser induced damage of high reflector at 1064 nm," Proc. SPIE **7842**, 784205 (2010).
15. Z. Y. Meng, S. I. Huang, Z. Liu, C. H. Zeng, and Y. K. Bu, "Design and fabrication of a novel high damage threshold $HfO_2/TiO_2/SiO_2$ multi-layer laser mirror," Optoelectron. Lett. **8**, 190–192 (2012).
16. B. C. Stuart, M. D. Feit, S. Herman, A. M. Rubenchik, B. W. Shore, and M. D. Perry, "Nanosecond-to-femtosecond laser-induced breakdown in dielectrics," Phys. Rev. B **53**, 1749–1761 (1996).
17. C. W. Carr, H. B. Radousky, A. M. Rubenchik, M. D. Feit, and S. G. Demos, "Localized dynamics during laser-induced damage in optical materials," Phys. Rev. Lett. **92**, 087401 (2004).