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Study on Brewster angle thin film polarizer using hafnia–silica mixture as high-refractive-index material

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Abstract. Two kinds of polarizer coatings were prepared by electron beam evaporation, using HfO₂ – SiO₂ mixture and HfO₂ as the high-refractive-index materials, respectively. The HfO₂ – SiO₂ mixture layer was implemented by coevaporating SiO₂ and metal Hf, the materials were deposited at an oxygen atmosphere to achieve stoichiometric coatings. The certain HfO₂ and SiO₂ content ratio is controlled by adjusting the deposition rate of HfO₂ and SiO₂ using individual quartz crystal monitor. The spectral performance, surface and interfacial properties, as well as the laser-induced damage performance were studied and compared. Comparing with polarizer coating using HfO₂ as high-refractive-index material, the polarizer coating using HfO₂ – SiO₂ mixture as high-refractive-index material shows better performance with broader polarizing bandwidth, lower surface roughness, better interfacial property while maintaining high laser-induced damage threshold. © 2018 Society of Photo-Optical Instrumentation Engineers (SPIE) [DOI: [10.1117/1.OE.57.2.025101](https://doi.org/10.1117/1.OE.57.2.025101)]

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1 Introduction

Brewster angle thin film polarizer, as one of the essential components in a high-power laser system, could switch the beam out of the primary cavity and protect the system from back-reflected light.¹ The design requirements of a polarizer coating for high-power laser system usually include a high laser-induced damage threshold (LIDT), a *p*-polarized transmittance (T_p) higher than 98%, and an extinction ratio (T_p/T_s) > 100:1, where T_s is the *s*-polarized transmittance. Considering the uniformity of the film deposition, tolerance of angular misalignment, as well as the variations of spectral performance with relative humidity, an allowance of polarizing bandwidth with high T_p and T_p/T_s must be made. The polarizing bandwidth greatly depends on difference between the refractive indices of the two coating materials used. The larger the difference between refractive indices, the easier to achieve broader polarizing bandwidth. SiO₂ is the most commonly used low refractive index material due to its low loss property at 1064 nm. Although HfO₂ has a lower refractive index than Ta₂O₅ and TiO₂, it is still the most favored high refractive index material for high-power laser coatings at 1064 nm since it is the most laser-resistant high-refractive-index material.^{2,3} With the given coating materials, the coating design and thickness monitoring accuracy can be optimized to improve the spectral performance of the polarizer coating. Stolz⁴ theoretically studied the effect of coating design on bandwidth, extinction ratio, and LIDT of the polarizer multilayer coatings. Zhu et al.⁵ achieved the polarizer coating with spectral performance close to the theoretical design, by proposing a monitoring strategy using two pieces of witness glass. Oliver et al.¹ demonstrated high transmission and contrast over an extended wavelength/angular range

suitable 8-nm spectral bandwidth of OMEGA EP, by dynamically correcting the design thicknesses to compensate the change of crystal performance as a function of deposited material on the surface of the crystal. Much work has also been done to improve the LIDT of thin film polarizer. Using metallic Hf in place of HfO₂ as the starting material for evaporation, the nodular defect density was greatly reduced, and the interfacial quality was improved, which ultimately improved the laser resistance.⁶ The effect of SiO₂ overcoat thickness on laser resistance of HfO₂ – SiO₂ Brewster angle thin film polarizer was also studied, and the damage threshold was improved by specific overcoat thickness.⁷ The morphologies of the laser-induced damage on HfO₂ – SiO₂ polarizers were investigated and summarized into four types, which have its particular growth behavior.⁸ Optimization of the deposition process parameters is also the commonly used strategy by coating researchers to minimize the material absorption and defect within coatings, and therefore maximize the LIDT of optical coatings. The refractive index of the coating layer also depends on the deposition process parameter. Taking oxygen pressure used during evaporation of Hf as example, a high oxygen pressure is helpful to minimize the material absorption; however, the refractive index of the HfO₂ layer decreases as the oxygen pressure increases. There might be a compromise between spectral performance and LIDT. The concept of material mixing in e-beam processes has been investigated for several decades.⁹ Previous research work indicated that HfO₂ – SiO₂ mixed composite film with a certain SiO₂ content has higher refractive index and lower surface roughness than pure HfO₂ film,¹⁰ whereas multilayer coating with HfO₂ – SiO₂ mixture interfaces shows a high laser damage resistance.¹¹

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In this paper, $\text{HfO}_2 - \text{SiO}_2$ mixture was used as the high-refractive-index material to broaden the polarizing bandwidth of the polarizer coating. For comparison, polarizer coating using HfO_2 as high-refractive index (POL) was deposited under the identical deposition process parameters. The surface morphology, interfacial property, as well as LIDT were characterized and compared. Polarizer coating using $\text{HfO}_2 - \text{SiO}_2$ mixture as the high-refractive-index materials (M-POL) shows broaden polarizing bandwidth, lower surface roughness, better interfacial quality while maintaining high LIDT.

2 Experimental Details

2.1 Sample Preparation

The polarizer coatings were deposited on BK7 glasses using a Leybold (formerly Leybold Optics GmbH) e-beam evaporation coater. Metal Hf and granular SiO_2 were chosen as high- and low-refractive-index coating material, respectively. For M-POL coating, the $\text{HfO}_2 - \text{SiO}_2$ mixture layer was achieved by evaporating metal Hf and SiO_2 simultaneously. The $\text{HfO}_2 - \text{SiO}_2$ mixture layers were deposited at an oxygen atmosphere enough to achieve stoichiometric coatings. The schematic diagram of the e-beam coater used in this work is shown in Fig. 1, the deposition rates of the HfO_2 and SiO_2 were controlled by the quartz crystal monitors located in the left and right sides of the chamber, respectively. The HfO_2 and SiO_2 content ratios of the $\text{HfO}_2 - \text{SiO}_2$ mixture layer were controlled by adjusting the deposition rate of HfO_2 and silica using individual quartz crystal monitor. The film thickness was controlled by a commercial OMS 5000 optical monitoring system.

In this study, the chamber was pumped to a base pressure of 4.5×10^{-4} Pa, and the substrates were baked to 473 K prior to deposition. $\text{HfO}_2 - \text{SiO}_2$ mixture layers in M-POL coating were achieved by depositing HfO_2 and SiO_2 at a rate of about 0.14 and 0.016 nm/s simultaneously, and at an oxygen pressure of 1.4×10^{-2} Pa. SiO_2 layers in M-POL coating were deposited at a rate of ~ 0.6 nm/s and an oxygen pressure of 3×10^{-3} Pa. For comparison, POL coating was prepared under deposition parameters identical with parameters using in M-POL coating, except that $\text{HfO}_2 - \text{SiO}_2$ mixture layers are replaced by HfO_2 layer deposited at a rate of 0.14 nm/s and an oxygen pressure of 1.4×10^{-2} Pa.

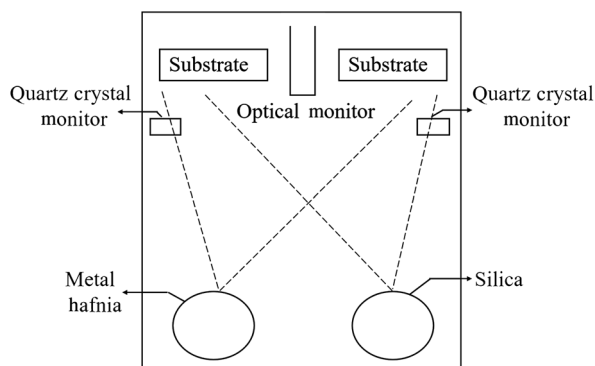


Fig. 1 Schematic diagram of the e-beam coater.

2.2 Sample Characterization

The transmission spectra of the films were measured by spectrophotometer (Perkin-Elmer Lambda 1050). The structure information was measured by x-ray diffraction (XRD, PANalytical Empyrean). Surface morphologies of the substrates and coatings were mapped by an atomic force microscope (AFM, Veeco Dimension 3100). The interface morphologies were characterized by a focused ion beam-scanning electron microscope (FIB-SEM, Carl Zeiss AURIGA CrossBeam). The typical damage morphologies were also characterized by the FIB-SEM, operated at an accelerating voltage of 1 kV. The samples were coated with 10- to 15-nm carbon before the FIB and SEM characterization to prevent surface charging.

The LIDTs of M-POL and POL coatings were evaluated under one-on-one test mode according to ISO 21254.^{12,13} A multilongitudinal mode Nd:YAG laser operated at 1064 nm with a pulse duration of 12 ns was used as the test laser, the laser damage experiments were carried out at normal incident angle from the forward propagating direction. The laser radiation at the sample plane has a near-Gaussian spatial profile. The e^{-2} spot diameters along the x - and y -axes were measured by a knife-edge method, and the effective beam area at the sample plane was 0.107 mm^2 . The site spacing was about five times greater than the laser spot diameter so that it can be sufficiently high to avoid the subsequent pulses influencing the test result on the neighboring sites. At each test laser fluence, 10 unexposed sites on the specimen surface were irradiated by one shot of laser radiation, damage was evaluated by comparing the test areas before and after laser irradiation, the fraction of sites damaged was recorded. The highest laser fluence incident upon the optical surface, for which the linear extrapolated probability of damage is zero, is defined as the LIDT.

The relative error of damage probability was about $\pm 15\%$ mainly due to the uncertainty of the nonuniformity among the samples (3%), the measurement of laser spot area (5%), and the fluctuation of laser energy (5%).

3 Results and Discussions

3.1 Spectral Performance

The transmittance spectra of M-POL and POL coatings were measured under p - and s -polarized light at incident angle of 56.4 deg. As shown in Fig. 2, the transmittance of p -polarized light and s -polarized light of M-POL coating is

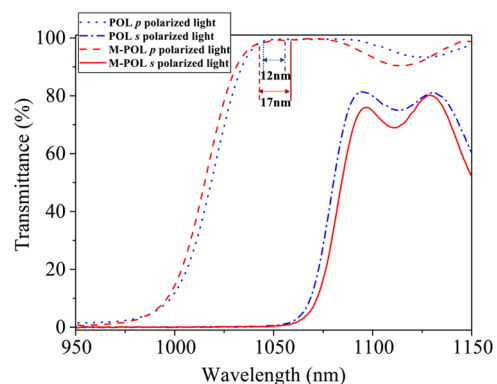


Fig. 2 The transmission spectra of M-POL and POL.

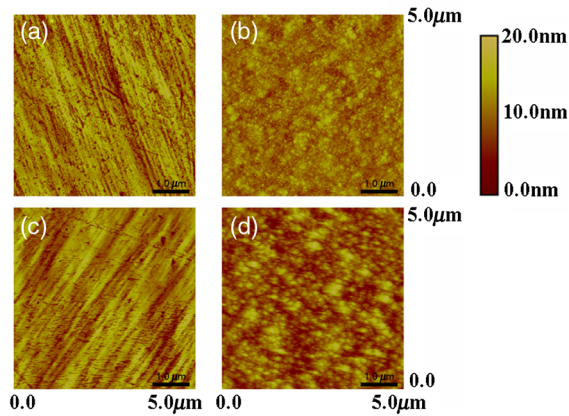


Fig. 3 Surface morphologies of (a) substrate of M-POL and (b) M-POL coating, (c) substrate of POL, and (d) POL coating.

illustrated by red dash line and red solid line, respectively. The transmittance of p -polarized light and s -polarized light of POL coating is illustrated by blue dash line and blue solid line, respectively. As $\text{HfO}_2 - \text{SiO}_2$ mixture layer with a small amount of SiO_2 has a higher refractive index than pure HfO_2 layer,^{10,11} the M-POL coating is expected to show a better spectral performance. Under the deposition parameters used in this work, the bandwidth of the polarizer coating is improved from 12 to 17 nm.

3.2 Surface Morphology

The surface morphologies of the substrates and coatings were characterized by AFM in tapping mode, and the root mean square (RMS) surface roughness was extracted by the software. For each sample, a $5 \times 5 \mu\text{m}$ area was characterized, and the RMS surface roughness was computed by the AFM software.

For comparison, substrates with similar RMS were used. The surface morphology and RMS of the substrate were shown in Figs. 3 and 4. The surface morphologies of the M-POL and POL coating were shown in Figs. 3(b) and 3(d). It is obvious that the M-POL coating has smaller grain size than POL coating, indicating a better surface quality. By replacing HfO_2 layers with $\text{HfO}_2 - \text{SiO}_2$ layers, the RMS

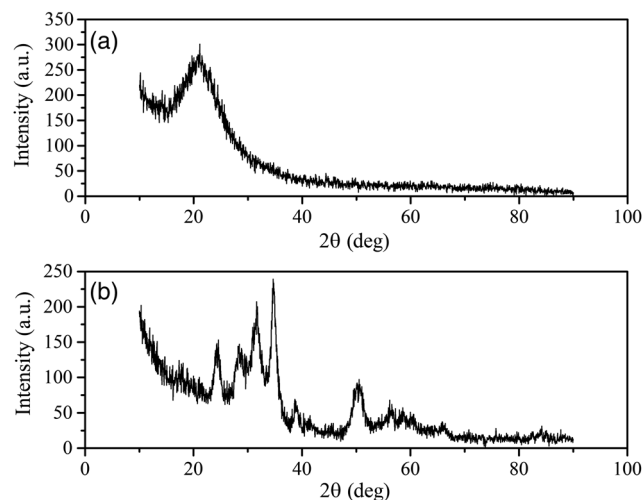


Fig. 4 The diffraction patterns of (a) $\text{HfO}_2 - \text{SiO}_2$ mixture coating and (b) HfO_2 monolayer coating.

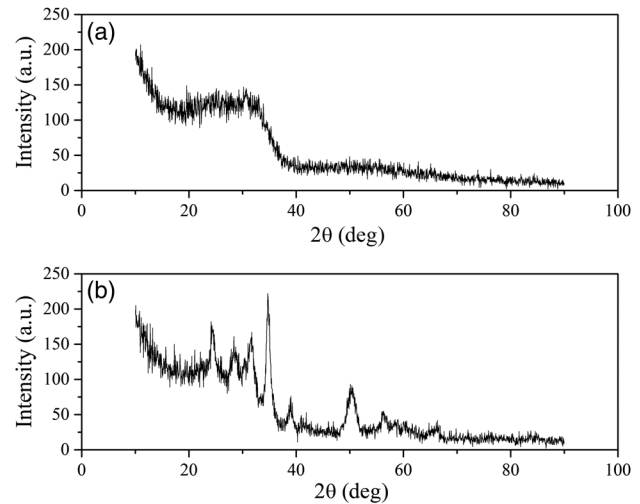


Fig. 5 The diffraction patterns of (a) M-POL and (b) POL coating.

of coating is reduced from 2.47 (POL) to 1.52 (M-POL) while RMS of substrate is slightly reduced from 0.84 (POL) to 0.81 (M-POL), which can be attributed to that the RMS of the high-refractive-index layer in M-POL coating is much lower than that of POL coating.¹⁰

3.3 Structure Properties

The structure information of the HfO_2 and $\text{HfO}_2 - \text{SiO}_2$ mixture monolayer coating, as well as M-POL and POL coating, were investigated using XRD. As shown in Figs. 4(a) and 4(b), HfO_2 monolayer coating is polycrystalline, whereas the $\text{HfO}_2 - \text{SiO}_2$ monolayer coating is amorphous.

As a result, the M-POL is amorphous, whereas the POL is polycrystalline, as shown in Figs. 5(a) and 5(b).

3.4 Interfacial Quality

The cross-section images of two samples were characterized by FIB-SEM and shown in Fig. 6. The interfaces in M-POL coating with mixture layer as high-refractive-index material are flat and smooth, whereas the POL coating with HfO_2 as high-refractive-index material has rougher interfaces. It is not surprising that interfacial quality is different since the $\text{HfO}_2 - \text{SiO}_2$ mixture layers are amorphous and smooth with lower surface roughness, whereas the HfO_2 layer is polycrystalline and rough with higher surface roughness, which means the material grows on two different surfaces.

3.5 Laser-Induced Damage Threshold

The LIDT of M-POL and POL coating under irradiation of s -polarized light was evaluated under one-one-test test mode

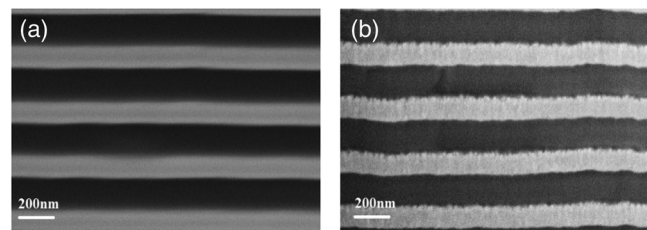


Fig. 6 Cross-section images of (a) M-POL and (b) POL tested by SEM.

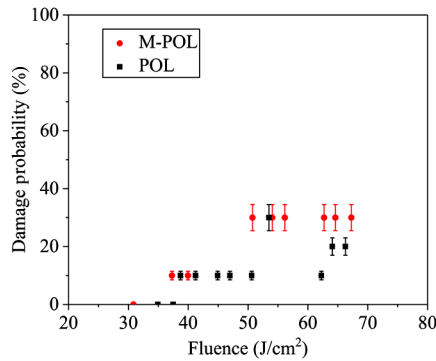


Fig. 7 Damage probability of M-POL and POL coating.

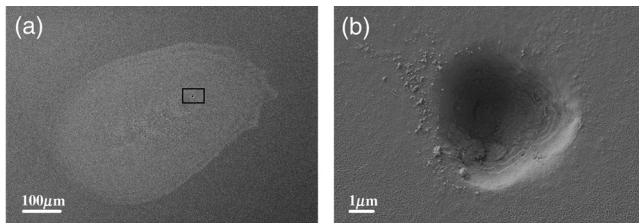


Fig. 8 (a) Typical damage morphologies of M-POL and POL (b) nodular ejection pit.

at 1064 nm with incident angle of 56.4 deg. As shown in Fig. 7, The LIDTs of M-POL and POL are at the same level, the LIDT of the polarizer coating is not degraded by replacing HfO_2 with $\text{HfO}_2 - \text{SiO}_2$ mixture layer.

As shown in Fig. 8(a), the typical damage morphology of M-POL induced by *s*-polarized laser irradiation is small nodular ejection pit surrounded by plasma scald, similar to the damage morphology of POL; Fig. 8(b) shows the high-resolution image of nodular ejection pit, which is the black-framed region in Fig. 8(a). The damage morphology indicates that nodular defect is the main reason that induces damage. It is well known that LIDT of coating depends on the coating materials, E-field distribution, defect type, defect density, and so on. In this work, the coating design structure of M-POL and POL is the same, the small difference in the high-refractive-index materials does not affect the E-field distribution much, which means the M-POL and POL coating have similar E-field distribution. Similar LIDT indicates that the substitution of $\text{HfO}_2 - \text{SiO}_2$ mixture layer for pure HfO_2 layer does not increase the type and density of the defects.

4 Conclusion

The Brewster angle thin film polarizer using $\text{HfO}_2 - \text{SiO}_2$ mixture and HfO_2 as high-refractive material were prepared by dual-source e-beam evaporation and compared in the experiment. The transmittance spectra indicate that M-POL has a larger polarizing bandwidth than POL since the

$\text{HfO}_2 - \text{SiO}_2$ mixture layer has a higher refractive index than HfO_2 layer. The M-POL also shows a better performance in both surface morphology and interface properties than POL since the $\text{HfO}_2 - \text{SiO}_2$ mixture layers are amorphous and smooth with lower surface roughness, whereas the HfO_2 layers are polycrystalline and rough with higher surface roughness. Laser damage test demonstrates that the M-POL maintains high laser damage threshold, which is attractive for the high-power laser application.

Acknowledgments

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