Influence of APS bias voltage on properties of HfO$_2$ and SiO$_2$ single layer deposited by plasma ion-assisted deposition

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HfO$_2$ and SiO$_2$ single layer is deposited on glass substrate with plasma ion assistance provided by Leybold advanced plasma source (APS). The deposition is performed with a bias voltage in the range of $70-130$ V for HfO$_2$, and $70-170$ V for SiO$_2$. Optical, structural, mechanical properties, as well as absorption and laser induced damage threshold at 1064 nm of HfO$_2$ and SiO$_2$ single layer deposited with the plasma ion assistance are systematically investigated. With the increase of APS bias voltage, coatings with higher refractive index, reduced surface roughness, and higher laser-induced damage threshold (LIDT) are obtained, and no significant change of the absorption at 1064 nm is observed. For HfO$_2$, a bias voltage can be identified to achieve coatings without any stress. However, too-high bias voltage can cause the increase of surface roughness and stress, and decrease the LIDT. The bias voltage can be properly identified to achieve coatings with desired properties.

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Optical thin films deposited by conventional electron beam evaporation tend to have lower packing densities; therefore, their optical properties are inferior to sputtered films unless the substrates are heated to elevated temperatures[1]. Plasma ion-assisted deposition is a well-known technique to improve the properties of thermally evaporated thin films[1]. With plasma ion-assisted deposition, a growing thermally evaporated film is bombarded with an energetic ion beam. The momentum transfer from the incoming ions to the condensing molecules causes the mobility of the particles to increase significantly, which results in denser and more stable films with lower substrate heating temperature, or even without heating. A number of studies have been conducted to investigate the packing density, refractive index, and vacuum to air shift of coatings prepared with plasma ion-assisted deposition[2]. The disadvantage of this technique is that densification also results in high compressive stress of the coatings, which may cause disturbing in high-end application[3]. In each case, the parameters of deposition process have to be optimized to achieve a balance between layer densification and other properties, such as evaporation rate, substrate temperature, bombarding gas, ion energy, and ion current density[4]. HfO$_2$ and SiO$_2$ are widely used in multi-layer coating designs for fabricating high-performance reflectors and polarizers used in high-power laser systems[5−8]. However, the optical and mechanical properties of the coatings prepared with conventional electron beam deposition are susceptible to the environment temperature and humidity because the coatings are not sufficiently dense. In order to achieve dense and stable coatings with high optical quality and good mechanical properties, the properties of HfO$_2$ and SiO$_2$ single layer should be investigated thoroughly. In this work, HfO$_2$ and SiO$_2$ single layer is deposited on glass substrates at 140 °C with ion assistance provided by Leybold advanced plasma source (APS). The deposition is performed with different ion energies.

The Leybold APS is composed of a hot cathode, a cylindrical anode tube, and a solenoid magnet. The ion energy is mainly determined by the self-bias voltage between the anode and chamber ground. The magnitude of the bias voltage depends on the applied discharge voltage, the magnetic field strength, and the chamber pressure[2]. In this work, the depositions are performed with a bias voltage of $70-130$ V for HfO$_2$ and $70-170$ V for SiO$_2$. The properties of the samples, such as refractive index, surface roughness, residual stress, film absorption, and laser-induced damage threshold (LIDT), are systematically investigated.

For all the HfO$_2$ single layer, the optical thickness is 8 quarter wavelength at 600 nm, the deposition rate is set to 0.3 nm/s, and the vacuum chamber is operated at a pressure of approximately 4×10^{-2} Pa. The APS is operated at bias voltages of 70, 90, and 130 V. For all the SiO$_2$ single layer, the optical thickness is also 8 quarter wavelength at 600 nm, whereas the deposition rate and the vacuum chamber pressure are changed to 0.6 nm/s and 4×10^{-2} Pa, respectively. The APS is operated at bias voltages of 70, 100, 150, and 170 V. Both HfO$_2$ and SiO$_2$ coatings are deposited at the temperature of 140 °C; all results are obtained from the samples without any other post processing, including annealing; however, the spectra shifts of both HfO$_2$ and SiO$_2$ layers are investigated with samples annealed in air at 300 °C for 3 h. Two runs are made at each bias voltage, and the average values are calculated as the data in this letter.

Transmittance of substrate and coating are measured by a Perkin Elmer Lambda 900 spectrophotometer, and the spectral curves are used to estimate the refractive index n with software Essential Macleod. Surface rough-

ness of the coatings is measured by a atomic force microscope (AFM) (Veeco Metrology Group, Dimension-3100). A 5×5 micron area is sampled in tapping mode. Root mean square (RMS) surface roughness is computed by the AFM software. Stress modification is determined by the magnitude of relative stress-induced curvature of the coating; curvatures of the uncoated glass substrate and coatings are measured by Techo optical interferometer, respectively. Absorption at 1064 nm is measured by a homemade system based on a surface thermal lensing technique, with sensitivity better than 10^{-5} and minimum deviation of 5×10^{-6}[9]. LIDT at 1064 nm is determined under a 1-on-1 test mode with a laser pulse width of 9 ns. Each test spot is irradiated by a single pulse at a certain laser power density, and 40 different spots are irritated for each laser power density.

As the bias voltage increases from 70 to 130 V, the refractive index n of HfO_2 increases significantly, whereas the refractive index n of SiO_2 increases only slightly with bias voltage increases from 70 to 170 V, as shown in Fig. 1. Portions of the samples are annealed in air at 300 °C for 3 h. From the annealed samples, no spectrum shift is observed in HfO_2 prepared at 130 V, and SiO_2 prepared at 170 V, whereas a blue shift is observed in other annealed samples. The shift wavelength is shown in Table 1. According to Fig. 1 and Table 1, we can come to the conclusion that the film density increases as the bias voltage increases.

As shown in Fig. 2, a decrease of surface roughness can be observed as the bias voltage increases for SiO_2 single layer. For HfO_2 single layer, the surface roughness first decreases as bias voltage increases from 70 to 90 V, and then increases when bias voltage increases to 130 V. The increase of bias voltage translates into higher kinetic energy for Ar atoms impinging onto the sample, and some of the energy is converted to thermal energy. This can accelerate the migration of the atoms to the substrate surface and increase the probability of more vacancies in growing films being filled, resulting in the decrease of the surface roughness of SiO_2 layers and HfO_2 layers at lower bias voltage. However, when the
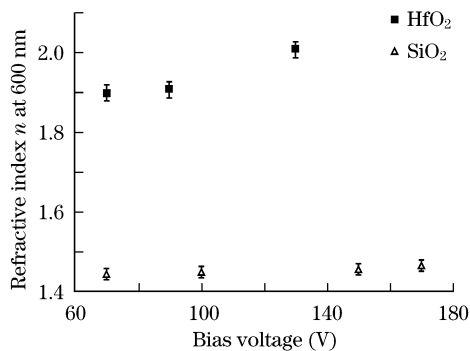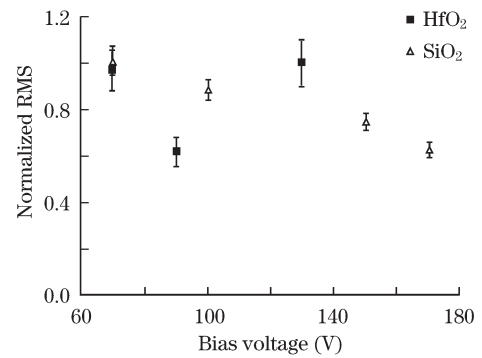
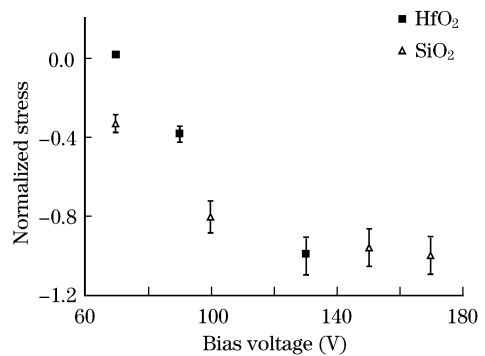Fig. 2. Surface roughness of HfO_2 and SiO_2 single layer as a function of APS bias voltage.

Fig. 3. Stress of HfO_2 and SiO_2 single layer as a function of APS bias voltage.

growing film surface is bombarded with too-high energy ions, the deposited atoms are removed, and surface defects, including vacancies, come into being, resulting in the increase of the surface roughness.

As shown in Fig. 3, the stress of HfO_2 single layer is nearly zero when bias voltage is 70 V, and transits from a tensile state to an increasingly compressive state above this potential. All SiO_2 layers suffer from a compressive stress, which increases as bias voltage increases.

As shown in Fig. 4, for both materials, the absorption at 1064 nm is in the order 10^{-5} and does not change significantly as the bias voltage increases, denoting that the increase of bias voltage does not affect the film absorption.

As shown in Fig. 5, the LIDT of HfO_2 single layer increases as the bias voltage increases from 70 to 90 V, and decreases as the bias voltage increases to 130 V. For SiO_2 single layers, the LIDT slightly increases as bias voltage increases from 70 to 100 V, and then decreases as the bias voltage increases. Defect density in the coatings is investigated under a Leica microscope system to study the effect of bias voltage on the LIDT. The result reveals that HfO_2 films deposited at 70 and 130 V have a higher defect density than film deposited at 90 V, whereas the defect density in SiO_2 film first slightly increases as the bias voltage increases from 70 to 100 V, and then increases significantly as bias voltage increases from 100 to 170 V. The defects in films are likely caused by material sputtering and other particles originating from the vacuum chamber. Since the electric
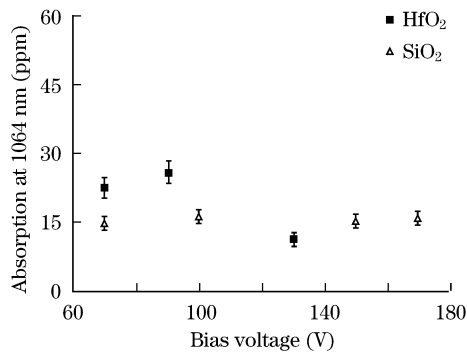
Fig. 1. Refractive index n of HfO_2 and SiO_2 single layer at 600 nm as a function of APS bias voltage.

Table 1. Spectra Shift before and after Annealing in Air at 300 °C for 3 h

	HfO₂			SiO₂			
Bias Voltage (V)	70	90	130	70	100	150	170
$\Delta\lambda$(nm)	8.4	7	0	11	6	1	0

Fig. 4. Absorption of HfO_2 and SiO_2 single layer at 1064 nm as a function of APS bias voltage.
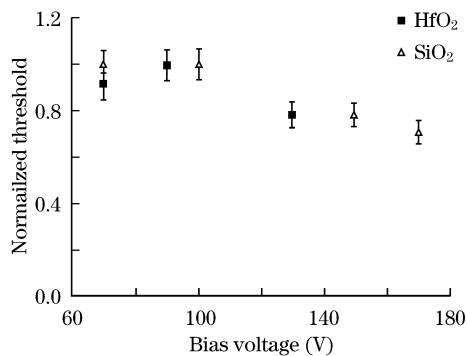

Fig. 5. LIDTs (1-on-1) of HfO_2 and SiO_2 single layer at 1064 nm as a function of APS bias voltage.

beam gun is operated within the same parameters for each run, including sweep pattern and emission current, the defect density caused by material sputtering is at the same level. This indicates that increasing ion energy causes more particles in chamber to enter into the growing films, consequently forming defects. In addition, as the ion energy increases, the contamination caused by the APS source itself becomes more likely.

The O_2 flow for HfO_2 is much higher than that for SiO_2. O_2 cannot be fully ionized at bias voltage of 70 V, and ionization continues at 90 V. This indicates the APS source works more stable at 90 V and correspondingly produces fewer defects in growing HfO_2 film. In contrast, materials are more sufficiently oxygenized with bias voltage increasing, but can also become oxygen-deficient when assisted by too-high bias voltage[4], resulting in the reduction of LIDT. Meanwhile, coatings with larger stress tend to be more easily damaged under laser irritation[10].

In conclusion, HfO_2 and SiO_2 single layer has been deposited onto glass substrate by plasma ion-assisted deposition with a different bias voltage. The coatings have been characterized with respect to the refractive index, surface roughness, mechanical stress, absorption, and laser-induced damage at 1064 nm. With the increase of APS bias voltage, coatings with higher refractive index, lower surface roughness, and higher LIDT are obtained, and no significant change of the absorption at 1064 nm is observed. For HfO_2, a bias voltage can be identified to achieve coatings without any stress. However, too-high bias voltage can cause the increase of surface roughness and stress, and the decrease of LIDT. The bias voltage can be properly identified to achieve coatings with the desired properties.

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