

The Role of Urbach-Tail Optical Absorption on Subpicosecond Laser-Induced Damage Threshold at 1053 nm in Hafnia and Silica Monolayers

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Hafnia and silica are widely used as the principal materials in multilayer dielectric (MLD) mirrors for petawatt-class laser facilities such as the OMEGA EP Laser System at LLE. Damage initiation in such dielectric coatings under excitation with pulses shorter than about 2.5 ps is associated with the formation of plasma that facilitates, via a complex energy deposition process, superheating of the affected volume.¹ Assuming a crystalline material, the buildup of the electron density in the conduction band originates with multiphoton excitation between the ground state and conduction band. However, the layers in MLD coatings are generally amorphous with a structure that varies based on the deposition method. Consequently, the optical absorption edge properties can be characterized by an optical gap that most often is analyzed using two methods. At photon energies above the optical gap, the absorption (α) behaves according to the Tauc formula, i.e., $\alpha\hbar\omega \propto (\hbar\omega - E_T)^2$, where E_T is known as the *Tauc gap*. Below the optical gap, the absorption is described by the Urbach tail² that arises from localized states in the band gap. In addition, defect states can further extend the absorption edge toward lower energies. As a result, transitions from the ground state to the conduction band are not limited to intraband transitions but include additional pathways through intermediate states at the Urbach tail and defects. It has been previously discussed that red shifting of the Urbach tail can lead to a reduced damage threshold in silica.³ The role of defects in decreasing the damage threshold has also been documented in various materials including hafnia monolayers.⁴

The optical gap and Urbach tail are in the 200- to 350-nm spectral region for both silica and hafnia layers. Based on the above considerations, this work explores the relationship of the laser-induced–damage threshold (LIDT) (using 1053-nm, 800-fs laser pulses) of silica and hafnia layers obtained by different vendors, using different deposition methods, to the characteristics of the Urbach tail in each material. The damage threshold is estimated following normalization for the electric-field distribution within each layer and is typically referred to as the “intrinsic” LIDT.⁵ The damage thresholds are investigated as a function of the estimated optical gap (Tauc gap) of each material and further evaluated as a function of the red-shifted Urbach tail absorption. Analysis of the absorption-edge characteristics is performed via (a) spectroscopic analysis in the UV spectral region and (b) photothermal absorption imaging using 355-nm excitation.

The results suggest that although the fabrication process has a large influence on the intrinsic LIDT, it only marginally affects the estimated optical gap energy. The samples (both silica and hafnia) that exhibit the highest intrinsic LIDT also exhibit the lowest absorption in the three- to five-photon absorption spectral range (≈ 200 to 350 nm), while the lowest LIDT samples exhibit the highest absorption. This trend was quantified in Figs. 1(a) and 1(b) by plotting the absorption coefficient as a function of the intrinsic LIDT for monolayers of hafnia at 351 nm and silica at 266 nm. The choice of the wavelength is based on using a threshold absorption coefficient value of the order of 10^3 cm^{-1} . These results suggest the presence of a correlation between the absorption at the Urbach tail to the intrinsic LIDT at 1053 nm using 800-fs pulses.

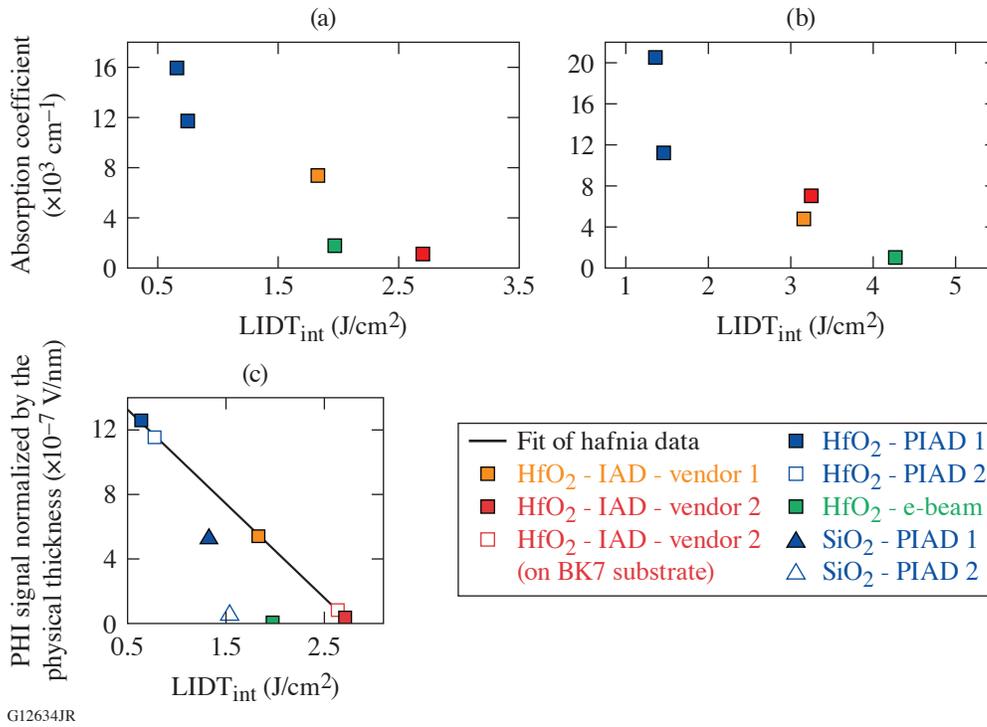


Figure 1

The absorption coefficient of (a) hafnia monolayers at 351 nm and (b) silica monolayers at 266 nm as a function of their intrinsic LIDT. (c) Photothermal heterodyne imaging (PHI) signal at 355 nm normalized by the physical thickness as a function of the intrinsic LIDT for hafnia (squares) and silica (triangles) samples.

The absorption in the UV spectral range was also probed using a photothermal heterodyne imaging system. This technique is based on a pump-probe approach and utilized a 355-nm pump beam to probe the local absorption with spatial resolution of the order of 500 nm. For each sample, several intensity maps were acquired by raster scanning, and an average value of every map for each sample was calculated. The photothermal absorption was normalized by the physical thickness of the layer. The results displayed in Fig. 1(c) demonstrate a direct relationship of the LIDT to the strength of the photothermal absorption signal.

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