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## Nanoscale-Thick Thin Films of High-Density HfO<sub>2</sub> for Bulk-like **Optical Responses**

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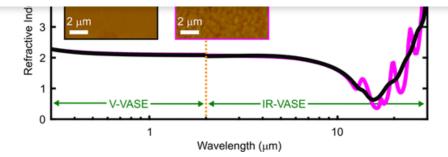
Thin films. ~

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The high refractive index, broadband transparency, and low spectral absorption of HfO<sub>2</sub> (hafnia) make it a suitable choice for thermally stable nanoscale optical filters, UV mirrors, and antireflection coatings. However, achieving dense thin films of HfO<sub>2</sub> with bulk-like optical properties has been a challenge due to differences in film stoichiometric and spatial uniformity at nanoscale thicknesses. Here, we assess HfO2 thin films (i.e., <200 nm thicknesses) prepared using pulsed laser deposition (PLD) at different substrate temperatures (20-675 °C) and the associated chemical, structural, and optical properties are reported. X-ray diffraction analysis reveals that nanoscale-thick thin films of HfO<sub>2</sub> deposited at an ambient substrate temperature (20 °C) using PLD are amorphous with embedded nanocrystallites, whereas films deposited on heated substrates are polycrystalline monoclinic HfO<sub>2</sub>. For the films deposited on heated substrates, further analysis shows that the nanocrystalline phase does not change with increasing substrate temperatures; however, the texture of the crystalline orientation changes to favor (111) at 300-675 °C from the initial (002) orientation at ambient substrate temperature. Such differences in nanoscale-thick thin-film HfO2 PLD process-dependent amorphousness, crystallinity, and surface textures discussed here exhibit minimal influence on the resulting broadband optical properties (250 nm-30 μm). Furthermore, we show that the complex refractive index of high-density nanoscale-thick thin-film HfO<sub>2</sub> prepared at an ambient substrate temperature using PLD resembles that of bulk HfO2 responses. The achievement of fully stoichiometric, high-density, thin-film HfO<sub>2</sub> from PLD is expected to further enable thermally robust nanoscale photonic device integration involving low-dimensional highperformance optical and optoelectronic applications.

**KEYWORDS:** amorphous  $HfO_2$ , hafnium oxide, thin-film optical constants, nanoscale-thick thin films  $\checkmark$ 

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micrographs, XRD results for the ( $\underline{1}11$ ) and (002) orientations as a function of  $T_S$ , XRR patterns at selected  $T_S$ , and V- and IR-VASE optical dispersion data with fits (PDF)

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