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Optical characterization of the impact of 100 keV protons on the optical properties of ZrO_2 films prepared by ALD on fused silica substrates

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Atomic layer deposition (ALD)-grown zirconia films underwent irradiation by 100 keV protons at fluences ranging from $1 \cdot 10^{12}$ p^{+}/cm^{2} through $5 \cdot 10^{14}$ p^{+}/cm^{2}. The induced structural damage was modeled using the stopping and range of ions in matter (SRIM) and compared with the change of the optical properties characterized by ellipsometry, spectrophotometry, and x-ray reflectometry. Proton-induced contamination of the optical surface due to deposition of a carbon-rich layer was determined. Correct estimation of the substrate damage was shown to be critical for reliable evaluation of the optical constants of the irradiated films. The ellipsometric angle Δ is shown to be sensitive to both the presence of the buried damaged zone in the irradiated substrate and the contamination layer on the surface of the samples. The complex chemistry in carbon-doped zirconia accommodating over-stoichiometric oxygen is discussed, along with the impact of the film composition change on the refractive index of the irradiated films.

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1. INTRODUCTION

Zirconium oxide (or zirconia, ZrO_2) is one of the most relevant ceramic coating materials for modern technologies. Zirconia is used for creating thermal barriers [1], for electric insulation in semiconductor devices [2], as a chemically persistent material having remarkable catalytic properties [3], for bio- and medical applications [4], and as a promising solid electrolyte [5]. In optics, zirconia is a material of choice for its high refractive index, low phonon energy, and significant luminescent yield [6]. Zirconia is also used in radiation dosimeters and devices for radiation imaging [7].

Zirconia has three crystal phases, with the cubic c-phase being the only one that is stable at high temperatures. For optical coatings, zirconia may be used as both crystalline and amorphous structures. Some applications may exploit the crystal-amorphous transition of zirconia [8].

Zirconia is used as the passivating layer protecting zirconium alloys in corrosive and nuclear environments. In its cubic stabilized crystalline state, it was reported to be exceptionally tolerant to radiation damage without amorphization [9], in particular at low temperatures and high fluence of xenon [10].

Many of the aforementioned properties of zirconia are exploited in space applications. In space, optical devices are exposed to harsh environments [11]. In particular, protons that originated mainly from the sun, with a very wide energy range, are one of the principal sources of optical device deterioration. To better understand deterioration of optical devices based on the use of metal oxides thin films, protons with energies of tens of kilo-electron-volts are of interest, as is well-known [12] because such low-energy protons come to rest within the first few hundreds of nanometers provoking the material damage, while higher energy particles do not affect the surface region.

Here we present a study on a set of amorphous thin films of zirconia manufactured by the atomic layer deposition (ALD) method. For optical applications, zirconia deposited by ALD is a relatively new material. ALD enables conformity of the coating to the coated surface and may yield interesting optical properties, both allowing for implementations in multiple modern applications.

Metal–organic precursors typically used for deposition of oxides by this technique might leave organic traces in the growing layer if the temperature-pressure regime of the chemical reaction is even slightly outside of the optimal range. In general,