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Spectroscopic Properties of Scintillating Hafnium Dioxide Nanocrystals

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In the last decade, many efforts have been devoted to the development of smart multifunctional materials. Among them, inorganic nanostructures have gained importance because of their outstanding luminescence properties and their potential applications as new building block materials for the next generation electronics and in a variety of lighting applications. In particular, many recent researches have been focused on the achievement of progress in the synthesis of nanosized metal oxides. In this field, a significant attention is paid to hafnium dioxide (hafnia or HfO2), which can be employed in optical protective and thermal barrier coatings. Thanks to its mechanical resistance, hafnia finds applications as ceramic, super hard materials and catalysts, or as component in gas sensors and fuel cell electrolytes. Lastly, HfO2 is now evaluated as potential alternative gate dielectric to replace SiO2 in the future generation of electronic nanodevices. Regarding the scintillating properties, the high atomic number Z=72 and the quite high density (9.6 g cm-3) make HfO2 nanocrystals good hosts for phosphor and scintillating applications where a large stopping power for ionizing radiation (X-rays, γ-rays) is required (1).
Bulk hafnia is very difficult to grow due to its high melting point (2774 °C). Actually, HfO2 can be synthetized also in the nanocrystals form and studied to fabricate thin films, optical ceramics and nanocomposite materials (2). Indeed, besides the well-known luminescence of HfO2 nanocrystals activated by the incorporation of rare earth ions, some recent studies evidenced the occurrence of a blue fluorescence from undoped nanocrystals upon UV excitation; remarkably, a broad bluish luminescence appears upon X-ray illumination (RL). These findings suggest the potential of HfO2 nanocrystals as radiation detectors, but the lack of a detailed model that relates their structural and RL properties still hinders the development of efficient nanoscintillators with optimized structure and chemical composition.
We studied the RL features of undoped monoclinic HfO2 nanocrystals and their dependence on the structural properties of the material at the nanoscale in order to elucidate their origin. Upon X-ray irradiation, the nanocrystals show six emission bands in the near UV/visible spectral range, detectable between 10 K and 300 K. The visible luminescence bands at 2.2 eV, 2.5 eV and 2.8 eV are similar to those detected in our previous PL studies (3), while the UV emission at 4.2 eV and 4.6 eV have been observed for the first time. The excitonic behavior of the UV luminescence is evidenced. The strong increase of the 2.5 eV blue luminescence in annealed samples is likely related to the presence of titanium and it might be used for the design of highly efficient blue scintillating materials.

1. LeLuyer C. et al. (2008), HfO2:X (X = Eu3+, Ce3+, Y3+) Sol Gel Powders for Ultradense Scintillating Materials. J. Phys. Chem. A, 112, pp. 10152-10155.
2. Lange S. et al. (2006), Luminescence of Re-Ions in HfO2 Thin Films and Some Possible Applications. Opt. Mater., 28, pp. 1238-1242.
3. Villa I. et al. (2016), Size-Dependent Luminescence in HfO2 Nanocrystals: Toward White Emission from Intrinsic Surface Defects. Chem. Mater., 28(10), pp. 3245–3253.

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